NEUTRON IRRADIATION EFFECTS IN REACTOR PRESSURE VESSEL STEELS AND WELDMENTS

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Vienna, Austria, October 1998

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FOREWORD

As a result of the popularity of the report "Neutron Irradiation Embrittlement of Reactor Pressure Vessel Steels", published by the Agency in 1975 as Technical Reports Series No. 163, it was decided that another report on this broad subject would be of use. The intent was not merely to update the earlier report, but rather to produce a document with a number of invited authors.

In this report, background, but contemporary views of specially identified areas of the subject are considered as self-contained chapters written by experts from various organizations from different countries.

An expert group comprising of L.M. Davies (United Kingdom), J. Föhl (Germany) and L. Ianko (IAEA) was convened by the IAEA in November 1989 to consider this proposal and to propose an outline and deal with organizational matters. It was decided to form an editorial board comprising of L.M. Davies, L.E. Steele (USA), J. Föhl, M. Brumovsky (Czech Republic), V. Lyssakov (Russia) and L. Ianko. Members of this board also served as "area co-ordinators", to keep in contact with authors, and as referees.

The IAEA wishes to acknowledge those who contributed to the book, all authors and reviewers.

The situation with regard to this new very late report was reviewed at the IAEA in October 1997. While the report was "dated" it was agreed it should be "cleaned up", copied and issued as a "working document" which would be freely available, because it had intrinsic value, and could be used as a "springboard" for a slimmer TRS covering the same topic area in about two years time.

The cause of the delays were reviewed and these included the very late submission of some of the chapters which resulted in a late review for acceptance by the IAEA's publication committee. Even with the exclusion of the, by then, dated material in the chapter on National Perspective the material was not in a "camera ready" state. The chapters were not on WP disks. The reprography technology had moved on with remarkable progress with the implication that to bring the report to the "house style" of the IAEA would require excessive effort which was not available. This consideration led to the conclusion that the material, unfortunately, was not now suitable for a TRS or TECDOC. However, because of its intrinsic value it was decided to produce the report as a "working document" under an IWG-LMNPP Number for reference purposes.
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CHAPTER 1  INTRODUCTION

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1.1 BACKGROUND

Since the demonstration of a sustained fission reactor in 1942 nuclear power has emerged as a major method for producing electricity. In the light of current knowledge of the amount and location of global fossil fuel resources and their current usage rates, current environmental and public concerns about the effect of burning fossil fuels, the enhanced future demand for electricity from the developing world and the expansion of demand for electricity in the developed world, it is not surprising that there is likely to be an increased demand for nuclear power, an environmentally benign available technology. At the end of 1996 there were 442 nuclear power plants in operation and 36 under construction in the world (1). This represented nearly 400,000 MWe(net) generating capacity of nuclear power plants in 1996. In equivalent terms, the electricity generated avoided the emission of over 2000 million tonnes of carbon dioxide in 1992 alone into the world's atmosphere if that amount of electricity had been produced from burning coal. Therefore, in a strict sense, nuclear power is not merely a benign energy source, it is actually environmentally beneficial.

Of those reactors in operation and construction the commonest type is the Pressurized Water Reactor (PWR). We will also be commenting upon the WWERs (Vodo-Vodianyi Energeticheskiy Reactor) in this paper. WWERs are pressurized water reactors and are generally located in Central and Eastern Europe. Three WWER models have been developed (2). The first, the 440/230 has a capacity of 440 MWe and was developed in the 1960s and produced
exclusively in the then USSR. The second, the 440/213 has the same capacity but an improved design mainly with regard to safety equipment. The third type is the WWER 1000 with a capacity of 1000 MWe and developed in the 1980s.

This book, in the IAEA Technical Report Series (TRS), is about the effects of neutron irradiation on the steel and welds used for the pressure vessels (PV) which house the reactor cores in light water reactors. There have been earlier overviews, for example (3) which can be used to expand upon the historical basis of the topic. The Reactor Pressure Vessel (RPV) is a key component in most Nuclear Power plants and, as it is generally considered to be irreplaceable, its operating life can therefore determine the lifetime of the plant. Its integrity is paramount in avoiding the release of contamination in the event of an, albeit remote, accident. It is therefore necessary to understand those features which affect its mechanical properties and which are used to predict its integrity and lifetime. The lifetime of a nuclear power plant is usually determined by those factors which determine an acceptable margin of safety and allow the production of electricity at an acceptable cost. The objective is to operate the plant safely and produce electricity at a high availability factor over its design life. The design life of the early light water reactors was not originally based on neutron irradiation effects- but was more to do with other design features such as fatigue usage factors (4).

The ability of a pressure vessel to withstand the normal pressure and thermal stresses during the heating and cooling part of cycles associated with plant start up and shut down together with the assessment of the stresses associated with postulated accidents are considered at the design stage. For these reasons it is important that the pressure vessel is made from appropriate materials and manufactured to a high quality standard and also meets the demands placed upon it from the design and operational requirements. (see Chapter 2 on Reactor pressure vessel design by J Fohl, Chapter 3 on Reactor pressure vessel materials by K Suzuki and Chapter 4 on the WWER reactor pressure vessel design by M. Brumovsky and Y. Dragunov). Pressure vessels are subject to 'allowable pressure-temperature limits during normal operation and operational test conditions. Greater sensitivity to neutron irradiation increases the restrictions on the operating 'window of the pressure-temperature relationship. Pressure-temperature operating limits reviews are performed periodically through life to assess the adequacy of operating parameters and to ensure against brittle fracture. Establishing quantitatively the safety margins of a nuclear pressure vessel, during normal operation and under fault conditions requires that reliable mechanical properties information be available (see Chapter 5 on the determination of reliable material properties by E Roos and J Fohl).

However nuclear power plants have been built and operated for many years (Fig
1. The relevance of this particular point is that in the late 1960s the role of the impurity and residual elements copper and phosphorus were identified (5)(6) as increasing the sensitivity of 'model' pressure vessel steels to irradiation embrittlement. The application of the results from this, and other early work, was truly remarkable in its impact on PWR PV technology and the economics of power plant operation. Benefits have been derived in terms of plant life assurance and also in extending the life of modern plant by the removal of the major component lifetime limiting feature.

Subsequently, nickel and many other elements (7)(8)(9) were also found to be important in their effect on the mechanical properties of PV steels and welds on irradiation. There are anecdotes of earlier steelmaking practice which utilised automobile scrap still containing the electrical copper wire for the manufacture of pressure vessel base metal and also of using copper coated electrodes for welding (10). The copper content of PWR PV welds and base metal made after about 1972 was substantially reduced but there remains a significant number of operating vessels with higher levels of copper in their welds. Additionally some early pressure vessels were produced by welding practices employing a flux which resulted in a low toughness in the ductile temperature regime (11).

Thus the reactor pressure vessels typifying this earlier generation of nuclear power plant were seen as the plant lifetime limiting components and the effect of neutron irradiation on the materials in the beltline region of RPVs was an area of concern and much study. The newer generation of pressure vessels, which have benefited from the improvements in pressure vessel technology and improved understanding of irradiation effects, have a much reduced sensitivity to neutron irradiation and therefore are not restricted in life from this cause. However the annealing of the Novovoronezh 3 WWER pressure vessel to mitigate the effects of neutron irradiation, as recently as 1987, is an indication of the relatively continuous nature of the impact of this particular problem.

1.2 IRRADIATION EFFECTS ON MECHANICAL PROPERTIES

Material composition, method of manufacture, metallurgical condition, design and operating temperature can all influence the mechanical strength of the Reactor Pressure Vessel (RPV) steels and welds during neutron irradiation. All these are features to be considered in assessing the behaviour of a particular pressure vessel. There is an increase in yield strength, an increase in tensile strength and a decrease in strain hardening capability of steels and welds. A possibility which also has to be considered is for there to be also a non-hardening component of embrittlement caused by irradiation enhanced temper embrittlement or transmutation and precipitation effects during long term operation.

The main neutron irradiation effects are illustrated by the Charpy test where the amount of energy required to break specimens made from samples of the steel
is measured. The higher the energy required, the less brittle or more ductile is the specimen being tested. A pendulum is swung, with a known energy, which is established by how far the pendulum swings in a free but unloaded state. When a notched specimen of standard shape is inserted in the path of the swinging pendulum then the distance now swung is a measure of the energy absorbed in fracturing the specimen. Steels and welds used for RPVs demonstrate a transitional behaviour as a function of test temperature, see Figure 2 and 3, where the material exhibits brittleness at low temperature and ductile behaviour at higher temperatures. In general terms the effect of neutron irradiation is to shift this transitional behaviour to higher temperatures and to reduce the energy absorbed for fracture in the ductile region. There may also be an increase in the temperature range of the transition region. This shift in Ductile-Brittle Transition Temperature (ΔDBTT) is frequently defined at the reference absorbed energy level of 41J (30ft.lbs.) which generally correlates with the transition temperature defined as the Reference Nil Ductility Transition Temperature (RT_{NDT}) from a separate drop weight test, ΔDBTT is therefore to be considered to be equivalent to the shift in RT_{NDT} (ΔRT_{NDT}) which is commonly employed to 'reference' the fracture toughness of the material.

ΔDBTT varies with the neutron dose (fluence) experienced by the RPV. The energetic neutrons encompass a range of energies. The fluence is usually specified in terms of energetic neutrons above a particular neutron energy level, for example, greater than 0.1, 0.5 or, more usually, greater than 1.0 Million electron Volts, (MeV). Neutron fluence is now usually expressed in units of neutrons (n) per square metre but, much data, many workers and formulations still use neutrons per square centimetre so both will be used here. There is also a current trend to express neutron effects in terms of the damage produced by incident neutrons in the matrix iron crystal lattice. Collisions between the more energetic neutrons and the lattice atoms produce primary knock-on atoms (PKAs), which in turn, lose their energy by interacting with the lattice atoms to produce, under the appropriate conditions, vacancies and interstitials. At higher energies, where collisions occur every lattice spacing, collision or displacement cascades are produced in which the final configuration is generally thought to be a vacancy rich region surrounded by interstitial atoms which may be clustered. The calculated number of point defects produced by PKAs provides a measure of neutron exposure expressed as displacements per atom (dpa).

1.3 NEUTRON FLUENCE

Knowledge of the neutron exposure of a pressure vessel is necessary in order to analyse changes in mechanical properties (see Chapter 6 on Neutron Exposure by G.Prillinger and R. A. Van Konynenburg). The aspect of reporting neutron fluence is a particularly important feature in normalising data derived from different irradiation positions or different reactors and then relating them to a particular location in an actual RPV, where the neutron flux and energy
spectrum are usually substantially different. Information on irradiation effects on the mechanical properties of RPV steels has also been gained from accelerated irradiations carried out in Materials Test Reactors where, again, the neutron flux could be significantly higher and the neutron energy spectrum could be significantly different from that of an actual location in an RPV where the data is to be applied.

Surveillance capsules are located inside the RPV of power reactors, where the neutron flux and energy spectrum is also usually higher than the actual RPV. The method of reporting neutron fluence continues to be important and for example, earlier workers (12) found that "...neutrons with energies 1MeV would account for over 75%...but neutrons with energies greater than 0.1MeV would account for 94% of the transition shift temperature increase observed for virtually every spectrum". Indeed some of the uncertainties associated with calibrating the fluence can also be removed by the use of a 'standard' reference material, (i.e. one having a 'known' response in its change of mechanical properties to neutron irradiation), in surveillance programmes. Such a material is that designated as JRQ in the IAEA Coordinated Research Programme, Phase 3.

1.4 IRRADIATION EFFECT TRENDS

The effects of irradiation on mechanical properties are described in Chapter 8 (by W. L. Server, T. J. Griesbach, Y. Dragunov and A Amaev) The effect of irradiation on the mechanical properties can be followed by irradiating specimens taken from representative archive samples of an operational RPV (or research samples or candidate materials for future RPVs) under representative neutron fluences, and irradiation temperatures. This is accomplished by loading specimens in special assemblies in materials test reactors or in power producing reactors (surveillance assemblies) (see Chapter 7 by R. J McElroy, B. C. Edwards and B. Houssin) and testing them after irradiation to establish the change in mechanical property. For example, the transition temperature increase at a particular neutron fluence can then be derived from knowledge of the equivalent unirradiated curve and this gives one point on the ADBTT-neutron fluence curve (trend curve). The curves shown in Figure 4 (13) show the increase in ADBTT with increasing neutron fluence and are for steels and weldments of particular compositions. As we have mentioned earlier, different compositions, particularly with respect to impurity and residual element concentrations, can have a significant effect on the sensitivity of the steel to neutron irradiation.

The significance of having appropriate and flexible surveillance programmes has continued to increase over the years. The data obtained from tests on surveillance specimens are used to set operating pressures and temperatures of RPVs; the data is used to assess vessel integrity for actual plant transients; the technique can be used to evaluate the effect of re-irradiation of samples from annealed vessels; a rescheduling of the surveillance programme can meet the
needs of a plant life extension programme; the characterised facilities can be used for the irradiation of samples from other reactors which may be temporarily shut down or unavailable, or samples of generic or candidate materials. Indeed for the Spanish reactors (30) problems are emerging of rescheduling their surveillance programmes against a background of surveillance results which show a low irradiation sensitivity and the potential for a much extended PV life.

1.5 EMPIRICAL MODELLING OF IRRADIATION EFFECTS

Earlier empirical models describing irradiation effects on mechanical properties for guidance for Regulatory purposes led to the preparation and use of United States Nuclear Regulatory Commission (USNRC) Regulatory Guide 1.99 (14). This merely took the copper and phosphorous content of the steel and related them to the neutron fluence to give the trend of the shift DBTT for that particular steel. This was later modified and refined into a Revision 2 (15) in order to take into account the increasing base of surveillance data, to recognise the importance of nickel in irradiation sensitivity and to treat the weld data as a separate family. Phosphorus content was not included as a variable in the USNRC Reg. Guide Rev.2 because the role of phosphorus could not be distinguished from the data base, which included results for a restricted range of phosphorus and, perhaps more importantly, for the higher copper content of the US steels where the effect of phosphorus is not so marked. It is 'bulk' copper that is used to describe neutron irradiation sensitivity. For a schematic illustration of the effect of copper see Figure 3.

But, copper is present in these steels and weldments in many forms. A significant observation was the discovery (31) of 'Digenite', copper sulphide inclusions, in these steels. Copper can also be present as coarse particles, precipitated or undissolved, after stress relief heat treatment of the steels and weldments. Copper is also present as precipitates during irradiation. However it is the copper in solution that provides the major potential for future irradiation embrittlement. Thus, depending upon the quantitative and relative amounts of these forms of copper then it may be that the best descriptor of 'effective' copper is the unirradiated level of 'dissolved' copper. However, for the range of steels being used for RPVs elsewhere in the world phosphorus, as well as the major effect of copper, is seen as an important variable in describing hardening and non-hardening changes in mechanical properties. For example, French (16), Japanese and Russian (17) empirical models, deriving from their own national steels' irradiation effects data bases include phosphorous, and, in some cases, other elements also as variables. There is therefore an intent, because of the shortcomings of some of this large number of Guides which may be based on limited or somehow unique data, for an IAEA data base to be established which will encompass a large amount and a greater variety of the international irradiation data and which will reflect the large variety of materials in use. It is now generally thought that phosphorous is also a significant element in promoting irradiation sensitivity. Annealing studies (17)(32) suggest that
phosphorus behaves in two ways as an embrittling agent. Firstly as ultra-fine phosphide precipitates, in the same way as copper, and also as a grain boundary segregate to produce temper embrittlement. Clearly there is still scope for further work.

The empirical models and Guides serve a variety of purposes and usually describe or predict the mechanical properties after the irradiation of RPV steels and weld specimens. These models are produced in the context of a national programme and are usually, as stated in the previous paragraph, within the constraints of the data base used to generate those models. They could therefore be subject to modification or further refinement as new data outside the existing range becomes available. In some cases no surveillance data base exists for a particular plant and also there may be no archive material available for irradiation. The materials may belong to a particular 'family of steels' and these empirical models can be used to predict behaviour in a 'generic' way but confidence in the results can always be enhanced with additional information on the materials such as chemical composition from samples taken from irradiated vessels or by generating more data by irradiating equivalent materials. (see Chapter 12 Safety assessment using surveillance programmes and data base by D.-H Njo)

1.6 MECHANISTIC MODELLING

More sophisticated comprehensive models based on the underlying mechanisms of neutron irradiation effects on pressure vessel steels and welds have and continue to be developed (see Chapter 10 of this TRS by Professor Odette)(18)(19). The detailed underlying irradiation effects and the resulting metallurgical structures which lead to changes in mechanical properties of steels and welds continue to be the subject of detailed investigation. New techniques are available which permit a more detailed description of metallographic structures (see Chapter 9 of this TRS by Drs English and Phythian). The potential of these new developments is being realised to meet technological requirements of operating plants. A full description of the detailed metallography of the pressure vessel materials of a specific plant will augment and underpin the formulation of empirical and mechanistically based relationships between neutron fluence and mechanical property change and also possibly explain the role and significance of other alloying, residual and impurity elements. Increasingly, as these refinements continue to be developed, a detailed metallographic description will become available to help characterise the condition of real pressure vessels and will be of direct value in independently characterising their irradiation and mechanical condition.

Neutron irradiation surveillance results have recently been reviewed and discussed from a consideration of mechanistic models (10). The mechanical property changes are attributed to the interaction of dislocations with the following features resulting from neutron irradiation:
• Defect production (i.e. vacancies, interstitials, dislocation loops, vacancy clusters, caused by neutron displacement cascades

• Formation of ultra-fine copper-rich, coherent precipitates (age hardening)

• Ultra-fine phosphide formation

• Ultra-fine carbide formation

• Temper embrittlement caused by phosphorus segregation

It is now generally thought that the first two mechanisms are the most significant. Phosphorus is thought to behave in the same way as copper, that is, by hardening the matrix by precipitation as phosphides. However there is also the possibility for phosphorus to contribute to a non-hardening but embrittling mechanism by segregation to grain boundaries. Nevertheless, irradiation effects in model (simulation), and actual pressure vessel steels involve complicated processes and there may be differences between the two sets of materials, these continue to be the subject of much investigation. This overall situation will continue to be the case whilst predictions are required outside the boundaries of existing data bases or where further refinement of data is required for interpolation of data.

1.7 CURRENT 'STATE OF THE ART'

Thus neutron irradiation effects a change in the mechanical properties of PV steels and welds and the changes derive from a multiplicity of factors. These factors include the method of manufacture (plates, forgings and welds), fabrication (including variations in heat treatment), chemical composition (the degree of change being mainly dependant on the copper content) and metallurgical structure, neutron fluence, neutron energy spectrum neutron flux, irradiation temperature and time of irradiation. The changes can be predicted by using data derived from materials test reactor irradiations or from lower fluence rates from surveillance irradiations. Predictions can be obtained from the use of empirical guides together with additional confidence from the added knowledge of generic data, the composition of the steel or and metallurgical structure. However, amelioration or mitigation of these irradiation effects can contribute to the extension of operational life of some older plants. For nuclear power plants with 'modern' pressure vessels the problems of irradiation effects have been largely overcome and new problems of plant lifetime limiting features need to be assessed.

The stresses associated with a postulated transient which causes a rapid cooldown of the pressure vessel at high or increasing system pressure, Pressurised Thermal Shock (PTS), in a region sensitive to neutron irradiation containing flaws have
been evaluated in the USA. This led to 'PTS screening criteria values' which, in turn, have led to flux reduction programmes or other measures in several plants to ensure that the screening values will not be exceeded during current (licence) life. However, the USNRC is planning (29) to change (update) the current PTS rules with the result that many US PWR PV will exceed the PTS screening criteria before the end of (licenced) life.

1.8 MITIGATION OF IRRADIATION EFFECTS

Recovery of mechanical properties by annealing irradiation damage has been the subject of many investigations (see Chapter 11 by T. Mager, Y. Dragunov and C. Leitz) (20)(17). Significant recovery of unirradiated mechanical properties, both the shift in ductile brittle transition temperature and upper shelf fracture strength, could be obtained under relatively modest conditions of temperature and time. Interestingly the kinetics of upper shelf recovery is different from the shift in transition temperature indicating that different mechanisms may be operating in these different regimes.

The US Army SM-1A reactor was annealed in 1967 (21) and the BR-3 vessel in 1984 (22). The feasibility and economics of annealing vessels has been considered in the USA (23)(24) and a recommended guide for in-service annealing has been drafted by the ASTM (25). However, no 'commercial' PWR PV has yet been annealed outside the former USSR or former Comecon countries.

After surveillance capsules were withdrawn from the WWER Loviisa plant in Finland, and some of the former USSR plants, and the samples tested and analysed was it realised that the degradation in mechanical properties was greater than expected (see figure 5) (26)(27). The enhanced degradation in mechanical properties was ascribed to a combination of high copper and phosphorus and a high neutron fluence. From the subsequent evaluation of the WWER plants, in the light of these results, ameliorating measures were proposed and implemented to a greater or lesser extent on a plant specific basis. Amongst these measures were included a modification to the pressure-temperature limits, replacement of outer fuel assemblies by dummy elements which acted as neutron shields, use of highly depleted fuel assemblies at the core periphery and annealing to recover the unirradiated mechanical properties.

One of the major developments, and achievements, of the WWER programme (28) has been the annealing and recovery of a large number of actual irradiated pressure vessels (about ten) starting with the Novovoronezh 3 reactor pressure vessel, after 16 years service, in May 1987. This was carried out at a temperature of 420C for 150 hours after extensive studies on irradiation and annealing. The recovery was only partial. Higher degrees of recovery have been achieved in subsequent annealing of the other vessels by raising the annealing temperature
to 475°C. From the underlying studies (17) the residual embrittlement after annealing was found to be independent of neutron fluence (Fig. 6) but significant dependence on phosphorus content was found. The residual embrittlement was inversely proportional to annealing temperature.

1.9 INTERNATIONAL PROGRAMMES

Studies of neutron irradiation effects on pressure vessel steels and weldments continues to absorb much effort worldwide. The emphasis changes from the needs of the older generation of vessels to those associated with the longer anticipated life of the newer vessels.

The IAEA continues to be a focus for international activities in the field of neutron irradiation effects on pressure vessel steels and weldments. The activities were coordinated in the Division of Nuclear Power under the aegis of the International Working Group on the Reliability of Reactor Pressurised Components (IWG - RRPC), which now operates under as the International Working Group on Nuclear Power Plant Life Management (IWG - NPPLM). There are three main activities in this field. The first is through regularly held Specialist Meetings at different venues (e.g. Balatonfured, Hungary in 1990, Paris, France in 1993). The most recent meeting has been held in conjunction with the CSNI Working Group 3. from the OECD. The second has been through the Coordinated Research Programme on irradiation effects. This longstanding, but highly successful, venture has now reached the end of Phase 3. The third area of IAEA activity is in the generation of a more comprehensive international data base on irradiation effects which will cover a greater range of materials and conditions. This work has been initiated and will be coordinated by the IAEA.

The International Group on Radiation Damage Mechanisms (IG-RDM) was founded in 1987 with USNRC sponsorship, to bring together in a 'workshop' atmosphere scientists and engineers involved directly with RPV embrittlement issues. Involvement with the Group has led to many inter-laboratory comparison exercises. This aspect of the international activity has resulted in greater understanding of damage mechanisms because of the obvious advantages of applying a variety of techniques to the same material.

The European Action Group on RPV Materials Irradiation Effects and Studies (AMES) is one of a number of network groups being set up in Europe. There are a large number of objectives and while essentially European it is hoped to include a larger international membership at a later stage.

Indeed with the maturing and rationalisation of nuclear technology worldwide there is a perceived increase in cooperative activity in this area. By these means there is a 'sharing' of advanced equipment and techniques and participation in extra-national activities.
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Chapter 2. Reactor Pressure Vessel Design

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1 Introduction

The design of the RPV has to take into account all functional requirements to provide hot water (pressurized water reactor, PWR) or steam (boiling water reactor, BWR) and all possible deviations from normal operating conditions as well as loads resulting from low probability external events such as earthquake and air plane crash.

With regard to neutron irradiation the discussion of RPV design is limited to the core belt line of the vessel, which is defined as the region where the material accumulates a fluence of more than $10^{21} \text{m}^{-2} (E > 1 \text{MeV}) \ [1, 2]$. However, all loading patterns resulting from normal operation, emergency and upset conditions in the plant have to be considered in their effect on the RPV.

The design process includes a variety of different tasks. Starting with the specification of the functional parameters the main areas are:

- specification of initial material properties with sufficient reserve accounting for time depending degradation.

- reliable determination of material properties and the pressure and state of flaws in the component (quality assurance)
- determination of loading (operating, service) conditions

- time dependent degradation under service conditions

- surveillance of material and flaw state as well as monitoring of service conditions

- assumptions for worst case conditions with respect to loading spectrum, material and flaw state.

For the assessment of overall safety, validated computer codes (thermal hydraulics and finite element codes) and fracture mechanics concepts must be available to determine

- pressure and temperature as a function of time for all possible and postulated events in the plant, considering the plant specific interaction of components and events

- loading (stress, strain) of the component for a given pressure and temperature under steady state and transient conditions

- stress intensity and strain distribution of the component assuming the presence of flaws.

Failure of the reactor pressure vessel can be by leakage or by bursting, however, both cases have to be prevented by adequate design according to the Code requirements for boiler and pressure vessels as it is e.g. the ASME Code in the US or the KTA-Code in Germany. Crack initiation may occur, but arrest of the crack has to be assured at a depth of 75% of the wall thickness [3].
The likelihood of the occurrence of an event is assumed and can be estimated on a probabilistic basis. In consequence of an event two different safety strategies are in current use. In the probabilistic safety approach the vessel is assumed to fail under some severe events which, however, have an extremely low probability. Alternatively, the deterministic safety approach determines design conditions under which catastrophic failure is ruled out even in case of the most severe events. The philosophy of the deterministic approach is based on a material toughness concept with additional independent redundancies as outlined in the German "Basis Safety Concept", Fig. 1, [4]. The redundant measures are:

- "multiple parties testing" to assure and record the initial quality of the component
- understanding of the thermo-mechanical behavior of the material and thus to establish the "worst case" material state
- continuous monitoring of safety-relevant "in-service parameters" to assure that the design assumptions cover all events conservatively
- availability of validated computer codes, fracture mechanics concepts and non destructive examination (NDE) and evaluation methods.

The substantial prerequisite, however, for a "basis safe design" is the production of an ingot with optimized chemical composition regarding alloying, accompanying and trace elements including a reduced segregation pattern. This incorporates low sensitivity against degradation during manufacturing (forging, heat-treatment, welding, post weld
heat-treatment), provides high initial toughness and assures a minimum of defects. Additionally the sensitivity against ageing under service conditions is improved.

With respect to the weld, improvements have been made by adequate selection of flux in combination with requirements on the chemical composition of the weld wire, welding parameters such as weld bead size and sequence to minimize the coarse grained areas in the heat affected zone (HAZ) [5].

2 Service Conditions

For the core belt line region of the reactor pressure vessel, the safety relevant parameters are:

- pressure
- temperature
- neutron irradiation and
- coolant.

Materials used for the RPV are fine grained high strength ferritic steels which are protected against corrosion with an austenitic steel cladding. The operating temperature is commonly about 280 to 290°C, the operating pressure is at 7 MPa for BWR and up to 15 MPa for PWR.

Heavy components are susceptible to potential crack formation during fabrication, particularly resulting from welding and cladding with subsequent stress relief heat treatment. Because some cracks may remain undiscovered by non destructive examination (NDE), fracture mechanics analyses to evaluate their significance are required. In the safety assessment flaw sizes are postulated covering realistic and worst case conditions. The assumptions include subsurface cracks and surface cracks penetrating the cladding.
The degradation of the RPV material is essentially determined by the accumulated neutron fluence, therefore, efforts were undertaken to reduce the neutron flux at the vessel wall. This could be achieved by increasing the distance between the core edge and the vessel wall.

Neutron exposure is defined differently, in different countries. Some consider only neutrons with energies of \( E > 1 \text{ MeV} \), others set the threshold at a lower level e.g. 0.5 or 0.1 MeV. Reactors built in the 1960s were of smaller dimensions and the design life fluence (DLF) for forty years (load factor 80\%) was in the order of \( 4 \text{ to } 5 \cdot 10^{23} \text{ m}^{-2} (E > 1 \text{ MeV}) \), which is not in accordance with today's reduced DLF requirements e.g. in Germany of \( 1 \cdot 10^{23} \text{ m}^{-2} (E > 1 \text{ MeV}) \) \[3\]. Therefore the neutron flux at the vessel wall of those reactors was drastically reduced by means of a changed core loading and shielding elements inserted additionally (see section 2.7.1).

Since it is known that welds incorporate a high potential for material degradation, the number of welds in the RPV was reduced for the advanced reactor design so that seamless forged shells were used rather than plates with axial weld seams.

In the last 25 years the design power of generating units has increased up to 1300 MW (electric) and these require larger RPVs. In particular the shells in the core region were made as large as possible to further minimize the number of welds and increase the water gap between the core and the pressure vessel wall. With the large inner diameter of up to 5000 mm the wall thickness increased to 250 mm, Fig. 2. Producing such heavy ingots and fabricating large vessels of high quality was a big challenge to the industry. Examples of vessel dimensions are shown in Fig. 3.
For the large units (advanced design) the design life fluence is much lower and amounts to about $5 \cdot 10^{22} \text{ m}^{-2}$ for PWRs and $5 \cdot 10^{21} \text{ m}^{-2}$ (E > 1 MeV) for BWRs, respectively. In Germany for example the licensing requirement is not to exceed a fluence of $1 \cdot 10^{23} \text{ m}^{-2}$ (E > 1 MeV) in 32 effective full power years [3] which forced the industry to produce larger vessels than elsewhere. A comparison of water gaps for some typical reactors is shown in Fig. 4. This difference in design becomes obvious by comparing the two 1300 MW plants which represent the German and the US design [6].

Besides the geometrical parameters the neutron flux is determined by the core loading scheme, which is different in BWRs and PWRs and is the reason for the generally low flux in BWRs. Modifications in the fuel element arrangement were also made to reduce the design life fluence for older PWR reactor pressure vessels.

3 Design Criteria

The major criteria on which the design is based with respect to neutron irradiation are:

- pressure/temperature-time relationships and the resulting mechanical and thermal stresses including residual stresses from fabrication (load paths)

- quantitative data on change of material properties due to ageing effects, essentially by neutron irradiation

- minimum material properties requirements which allow safe operation or at least safe shut down in case of emergency and accident conditions.
This is shown in Fig. 5 [7]. Each of the tasks mentioned above was and is still subject of intense research and development (R&D) work. Progress has been made in the determination of the effect of neutron irradiation on materials the knowledge of which can generally be applied in the safety assessment. However, the exploration of the most critical loading path has to be performed for each individual plant.

Great efforts have been made to assess the limit for material properties at which catastrophic failure of a component can be excluded. Investigations in this direction were enhanced because several older reactors will reach their end-of-life (which can be defined in various ways) within the next decade. In this connection, decisions about life extension may have to be made. Furthermore, the weld of some US-reactors had low toughness already at the beginning of life while others have been susceptible to neutron irradiation [8], so the toughness values come close to the limits set by the relevant Codes.

4 Design Concepts

The safety assessment concentrates primarily on the prevention of brittle failure. Therefore the conditions must be defined under which brittle failure can be excluded. This is the case when the loading of the component occurs in a regime sufficiently above the Nil Ductility Transition Temperature $T_{\text{NDT}}$ [9] which is conservatively determined from both the Charpy and the drop-weight test. The reference temperature $T_{\text{RTNDT}}$ is the highest temperature comparing $T_{\text{NDT}}$ with $T_{68J} - 33 \text{ K}$ and $T_{0.9 \text{ mm}} - 33 \text{ K}$, respectively. $T_{68J}$ (temperature at which 68 J is reached) and $T_{0.9 \text{ mm}}$ (temperature at which 0.9 mm lateral expansion is reached) are derived from Charpy specimens testing with at least 3 specimens or from an energy/temperature and lateral expansion/temperature curve [10, 11].
4.1 Transition Temperature Concept

On the basis of the RT_{NDT} a temperature/pressure limit-curve (modified Porse diagram) can be established which separates the regime in which a component can be operated safely, from a so called "not allowed" ("prohibited") regime in which brittle failure has to be considered, Fig. 6. This concept is based upon the failure assessment diagram as introduced by Pellini [9]. It is concluded that even for large flaws brittle failure of the component will not occur at a high enough temperature above T_{NDT}. A basic assumption in this concept is that only primary stresses are acting arising from internal pressure.

The main concern of in-service material degradation is the increase in transition temperature and drop in upper shelf toughness. The degree of material degradation is usually determined with Charpy specimens only, measuring the shift of the transition temperature at an energy level of 41 J (ΔT_{41J}) and the drop in upper shelf energy (ΔUSE) [1, 2, 12, 13]. The energy level of the shift criteria has changed in the past from 68 J to 41 J, because the 41 J level is more closely related to NDT. However, in addition to that criterion, for some materials the upper shelf energy has dropped to a level close to 68 J, so that a shift determined at that level would be unrealistically high and in some cases the shift could not be determined at all. In some countries the shift is determined at other energy levels than 41 J, e.g. 49 J as in the ex-USSR.

To account for material degradation in the safety analysis, the Porse diagram has to be adjusted by shifting the curve to a higher temperature for that amount which can
be derived from the shift of the transition temperature as determined from the Charpy energy-temperature curve at an energy level of 41 J,

\[ RT_{\text{NDT}}^{\text{(adjusted)}} = RT_{\text{NDT}} + \Delta T_{41J}. \] (1)

An example for an adjusted pressure/temperature limit curve is also shown in Fig. 6.

As a preventive measure for RPVs with high shift in transition temperature, an automatic locking system has been introduced in some reactors to assure that no combination of pressure and temperature can occur in the prohibited regime.

4.2 Fracture mechanics concept

The transition temperature approach does not allow the quantification of the safety margin in flawed structures and cannot take into account secondary stresses. For this and other reasons the fracture mechanics approach was introduced. This compares the fracture toughness of the material with the stress intensity in a component, Fig. 7. According to some national Codes [14, 3] this approach is only applied where the linear elastic fracture mechanics regime (LEFM) is valid.

The prerequisites for using this approach are:

- reliable fracture toughness data of the material with regard to its transferability to large structures and complex geometries

- knowledge or reliable assumptions about flaw state in the component

- appropriate codes to compute the stress intensity for complex loading situations and geometries.
4.2.1 Crack initiation

For the design consideration the reference fracture toughness curve is used and fixed by the Reference Temperature RT
NDT as defined in section 4 above. The adjustment necessary to account for the material degradation is also made according to the transition temperature shift at the 41 J level.

From fracture mechanics testing of RPV steels a $K_{IR}$-curve was established as a lower bound curve that has to be used in the design phase [3, 14]. However, if actual and reliable fracture toughness data of the component are available, then these data may be used instead.

The load path has to be calculated for normal operating, emergency and faulted conditions. The size of the crack to be assumed in the safety assessment is either a semi-elliptical surface flaw with the depth $a = 1/4 \ T$ and $b = 1.5 \ T$ (where $T$ = wall thickness) or an infinitely long flaw with a depth depending on the reliability of NDE techniques multiplied by a factor of two to cover uncertainties.

The $1/4 \ T$ flaw is the basis for normal operating conditions for which the stress intensity is to be calculated with a safety margin of a factor of two for the primary stresses as shown in equation (2):

$$K_I = 2 \ K_I(p) + K_I(th) \quad (2)$$

- $K_I(p)$ stress intensity resulting from internal pressure
- $K_I(th)$ stress intensity resulting from thermal stresses
The assumption of an infinitely long flaw is used for the calculation of the behaviour under severe transients, because the extension of shallow cracks along the surface - to grow to a long crack - occurs before the crack penetrates the wall in the thickness direction [15]. In the case of emergency and faulted conditions the stress intensity can be calculated according to

\[ K_I = K_I(p) + K_I(th) + \text{others} \]  \hspace{1cm} (3)

The primary stresses do not have to be multiplied by a factor of two as required for normal operating conditions, however, the theoretical crack depth assumption has to be twice the size certified by non destructive examination.

A typical load path for start-up and shut-down is shown in Fig. 7. The comparison of the load path with the fracture toughness curve shows clearly that the safety margin has to be considered in two directions, one refers to load, the other to temperature. Failure of the component cannot be excluded when the load path intersects the fracture toughness curve (exceptions are discussed in section 4.2.3).

On the fracture mechanics basis the effect of transient loading on cracks can be assessed. The loading path for the most severe transient has to be evaluated for each individual plant. One critical accident assumption is the rupture of a small pipe (small break loss of coolant accident, LOCA) or a leak of corresponding size. In that case, the emergency core cooling pumps come into action feeding cold water into the vessel while the pressure is essentially maintained at saturation pressure or drops very slowly. As a consequence the inner surface of the RPV wall cools down rather rapidly building up thermal stresses which are in addition to the stresses resulting from internal pressure [16, 17, 18].
For a postulated leak of 1960 mm² (I.D. = 50 mm) and a coolant temperature of 30°C the pressure-time and temperature-time path responsible for the load on the pressure vessel is shown in Fig. 8. In this example a slight repressurization occurs after about 10 min. The resulting stress intensity was computed for different crack sizes (crack depth a) and is plotted in Fig. 9 for a circumferential flaw and in Fig. 10 for an axial flaw.

The higher primary stress for the axially oriented flaw leads consequently to high stress intensity values. Because the stress intensity was computed using linear elastic methods, unrealistically high values are reached shortly after start of cool down in a temperature range where ductile behavior of the material can be expected. The linear elastic fracture mechanics approach does not take into account any ductile (stable) crack extension. In the safety assessment, the upper temperature regime is only covered by results of Charpy impact test and associated experience. In this respect, failure of the component can be excluded when the Charpy upper shelf toughness is not lower than 68J. The calculation of the stress intensity for a circumferential crack with respect to brittle crack initiation, shows that small flaws are even more dangerous than large ones [19]. In the case of ductile (stable) crack extension, however, large flaws are controlling the process.

For emergency core cooling (ECC) events the $K_{ic}$-curve (static crack initiation) may be used instead of the $K_{IR}$-curve. The fracture toughness of materials with high sensitivity against neutron irradiation has to be adjusted at high temperatures, according to $RT_{NDT}$ (adjusted). In that case the slope of the $K_{ic}$-curve and the reliable calculation of the load path are of great importance with respect to possible intersection with the load path and thus to the lifetime for which safe operation can be assured.
4.2.2 Crack arrest

Since during ECC a temperature gradient through the wall exists, which depends strongly on the time after cool down has begun, Fig. 11a the secondary stresses are dominant in building up the stress intensity with a gradient across the wall, Fig. 11b. In case of brittle crack initiation during ECC the crack will grow rapidly into a field of higher temperature.

Once the crack tip has jumped to a location at which the material is in the upper shelf toughness regime the crack could arrest although the stress intensity may have increased during this crack extension. In general the stress intensity first increases with time and decreases after a certain time, Fig. 11c, however, as the material temperature at the new crack tip decreases and the stress intensity is still high enough a second crack initiation may occur. For the entire ECC path the correlation between temperature, crack depth, stress intensity and time can be computed. On this basis two curves

\[ K_1 = K_{ic} \quad \text{and} \quad K_1 = K_{ia} \]

can be established as shown schematically in Fig. 12 from which the sequence of crack initiation and arrest can be evaluated [18, 19].

According to some national Codes [3, 20], crack initiation may be considered, when crack arrest can be assured at less than 75% of the wall thickness. If this approach is being applied it becomes obvious that reliable initiation and arrest toughness data require to be guaranteed.
4.2.3 Warm prestressing effect

The warm prestressing effect was discussed as long ago as the early 1970s [21]. It refers to the fact that under conditions of decreasing stress intensity with time a crack cannot initiate. This situation occurs during ECC as shown in Fig. 9 and 10, respectively. There it is supposed that at the time when the load path intersects the $K_{lc}$-curve with a negative slope - indicating decrease in $K_I$ with time - a crack never will initiate. Previously it was only thought that overstressing of a sharp crack in the ductile regime would blunt the crack tip and thus crack initiation at lower $K_I$ values would be rendered more difficult. Further considerations of the mechanism of brittle crack initiation made clear that the warm prestressing effect can be explained on the basis of the mobility of dislocations [22].

When a certain $K_I$ value is applied to a structure in the ductile regime a plastic zone is being formed at the crack tip depending on the stress state and the deformability of the material under the given constraint (see Chapter 5). During cool down of the material at constant stress intensity (path 1) no additional energy has to be stored in the material and thus there is no driving force to move dislocations. Although the load path (horizontal curve) intersects the $K_{lc}$-curve, Fig. 13, crack initiation cannot occur. This is even more so in the case of decreasing $K_I$ (path 2).

For this reason a limit can be established beyond which crack initiation is not possible. This is the point when the maximum stress intensity has been reached. However, attention has to be drawn to the particular case of repressurization. If, after the intersection of the load path due to repressurization the stress intensity increases, crack initiation has to be expected. For highly embrittled materials and severe load paths the warm prestressing effect
is an essential item in the safety analysis. Several studies have been carried out to demonstrate the warm prestressing effect [23]. The benefit, however, is restricted to load cases for which repressurization can reliably be excluded.

5 Design curves to account for material in-service degradation

From fracture mechanics analyses it became obvious that knowledge about the fracture toughness/temperature curve was essential. In the unirradiated condition the lower limit curve for crack initiation has been well established for reactor pressure vessel steels used in the "western" countries [11, 14] as a conservative lower bound. In addition to that the procedure of RTNDT determination implies already some conservatism in the approach. For the adjustment of the limit curve in the irradiated state the $\Delta T_{41J}$ criterion is used as determined from Charpy impact testing (mean curve). The conservatism in this procedure has been demonstrated within research programs where fracture mechanics, drop-weight and Charpy specimens were irradiated in test reactors and even in power reactors [24, 25]. However, recent results from the United States "Heavy Section Steel Irradiation" (HSSI) program indicate that the margin between actual fracture toughness values and the predicted fracture toughness on the basis of the $\Delta T_{41J}$ shift shrinks to zero [26]. This does not directly affect the safety of a plant because of a variety of other conservative assumptions but points out the necessity of a thorough evaluation of data, in particular in the case of materials highly sensitive to neutron irradiation.

Other factors in the assessment of the plant lifetime are of equal or even greater importance. During the design phase, only the specified chemical composition of the steel is available to estimate the sensitivity of the material against
neutron irradiation. With actual data from quality assurance testing an adjustment can be made. With these data the change in transition temperature ($\Delta T_{41J}$) and the drop in upper shelf ($\Delta USE$) can be determined from design curves, as presented for example in the United States Regulatory Commission Regulatory Guide (US Reg. Guide) 1.99 or the German KTA 3203 (KTA, Regelwerk für kerntechnische Anlagen). These design curves have been evaluated on the basis of test reactor irradiation experiments in the past and have been updated recently with results from surveillance testing, supported by theoretical models accounting for metallurgical processes responsible for changes in the sub-microstructure.

At this time two diverse approaches exist. The US Code favours the combination of copper and nickel as being responsible for material degradation [13] whereas the trend curves in Germany [2] and in the ex-USSR [27] include copper and phosphorus. In the French approach copper, phosphorus and nickel is proposed [28]. In Japan an even complexer correlation between chemical composition and shift in transition temperature is being developed [29]. The contribution of nickel is still under discussion but has not been adopted in the Codes in general. Accepting that the effect of nickel is valid, this can be of special importance in considering the weld material because its nickel content is usually higher than that of the base material. In Fig 14 some typical design curves for $\Delta T_{41J}$, which were or are in use are compared.

The change in transition temperature and upper shelf energy due to neutron irradiation is important to judge the sensitivity of the material. More importantly, however, for the safety analysis are the absolute values of the reference temperature and the upper shelf energy. During operation of a reactor, plant specific data becomes available from surveillance testing. That data can replace or will replace the design life assumptions depending on the amount of actual
degradation in comparison with the data taken from the design curves.

The lay out of the surveillance program is therefore an essential point in the design phase (see chapter 8). To account for neutron spectrum and dose rate (neutron flux) effects, the location of the capsules in relation to the vessel wall has to be chosen adequately. A lead factor (ratio of neutron flux at capsule location and peak fluence at vessel wall) of 3-5 is recommended or required. This "close to wall irradiation" provides data which can be transferred reliably to assess the RPV wall material state (compare chapter 13).

6 Material limitations for pressurized thermal shock (PTS)

The vessel behavior under thermal shock loading is essentially controlled by the upper shelf toughness of the material with respect to ductile crack initiation and stable crack growth and by the nil ductility transition temperature ($RT_{NDT}$ adjusted) with respect to brittle crack initiation.

The theoretical basis for fracture mechanics analyses in the linear elastic and the elastic-plastic regime has been provided to determine the load paths during PTS events for individual plants. Efforts, however, were undertaken to evaluate material limitations from a more generalized point of view. A series of component failure interactions was investigated for different plants from which an upper limit of transition temperature could be established, the so called PTS-screening criterion [29]. For circumferential welds the adjusted reference temperature may not exceed 149 °C, for base material and axial welds, this value amounts to 132 °C.
In this case and when the upper shelf energy does not drop below 68 J, no individual safety analysis has to be performed. This screening criterion summarizes the theoretical linear elastic fracture mechanics models, the warm prestress effect and a validation of the fracture mechanics concepts for crack initiation and crack arrest by large scale experiments. In other Codes, besides the U.S. Code, this generalized screening criterion has not yet been adopted.

Extensive R & D work in the past was related to PTS loading situations as the most critical load paths for the RPV. In general, the underlying fracture mechanics concepts could be validated not only in the linear elastic regime but also in the elastic plastic regime using the J-integral. With respect to the definition of crack initiation toughness, however, different methods and opinions still exist which lead to remarkably different toughness values (see chapter 5). Under certain assumptions the stable crack growth behavior of components can be described. This has been demonstrated in PTS experiments with thick walled hollow cylinders [31, 32]. On the one hand the crack resistance curve (J as a function of Δa) of the material was determined with a compact tension specimen (CT 25 mm). On the other hand, the driving force J was calculated for the component under the specific test conditions (load, temperature, time) as shown in Fig 15. Under the applied test conditions stable crack extension occurred, the amount of which could be derived from the crack resistance curve of the CT-specimens. Studies with materials of low upper shelf toughness and high transition temperature comparable to materials degraded by neutron irradiation to validate the fracture mechanics concept, Fig 16, are still being continued.
7 Preventive measures to cope with in-service material degradation under severe loading conditions

In spite of efforts to design vessels to meet the harsh nuclear environment, additional steps were necessary to minimize material degradation and to mitigate loading conditions in the plant. Therefore measures to cope with neutron irradiation and severe loading situations have been carried out in the past successfully for different nuclear power plants, mainly for older plants the materials of which are particularly sensitive to neutron irradiation. Three separate courses of action were taken [33] which will be explained in more detail from the German point of view:

- reduction of neutron flux to reduce design life fluence
- enhanced non destructive examination to limit the flaw size to be postulated
- mitigation of thermal shock loading.

7.1 Flux reduction

Advanced computer codes and a more detailed modelling of the geometry have yielded a higher maximum flux at the RPV wall when compared with the computations performed in the mid sixties. This, coupled with increased safety requirements, has made it necessary to reduce the flux in order to reach the design lifetime. Firstly burned-up fuel elements were arranged at the outer core edges, but later on dummy fuel elements were inserted at specific locations, Fig 17. This mitigation method was also introduced in other reactors [34]. By this means it was possible to reduce the neutron flux and thus the design life fluence drastically, in some cases even without lowering the reactor power.
7.2 Non destructive examination

According to the Codes the stress calculation for emergency conditions must not consider the $1/4$ T flaw but a reduced flaw size which can be justified by the efficiency of the NDE techniques applied in the individual plant. Large efforts and progress have been made in recent years in the development of measuring equipment and data acquisition systems for flaw detection and sizing [35, 36]. The techniques have been verified and validated on a series of plates, forgings, small scale and full scale vessels. It is now extremely unlikely that a surface flaw of 5 mm depth will be missed. For the safety assessment a safety factor of two is required for the flaw size, therefore a flaw depth of 10 mm is commonly used in calculations. It was recognized that extensive NDE may be required also in the future. Therefore the design of the pressure vessel and the vessel internals must provide access to the critical areas of the RPV for non destructive testing equipment. In one case in Germany a special manipulative system had to be developed which was fed in the 60 mm gap between the thermal shield and the RPV to facilitate NDE of the core belt line region.

7.3 Mitigation of thermal shock loading

Calculations of the thermal shock load paths have clearly shown the influence of temperature of the water being injected during ECC, Fig 18. As one of the first steps to mitigate PTS impact on the RPV, the cooling water in some power plants was preheated to 30 °C and kept at that temperature constantly. In the future it is intended that the water will be preheated up to 50 °C. As a second step for plants with high design life (DL) fluence, additional modifications were made to alter the cooling water to be fed into the hot leg. This provides on the one hand a certain degree of mixing and thus a further increase in temperature.
On the other hand, the water does not pass the pressure vessel wall directly. Extensive studies [37] have demonstrated that even under hot leg water injection the core can be cooled sufficiently. This mitigation approach requires consideration in vessel design.

7.4 Annealing

Extensive studies have been performed with respect to annealing of a RPV to remove the effects of neutron irradiation on mechanical properties. Depending on the embrittlement mechanisms (see chapter 10) a high percentage of recovery occurs at 450 to 480 °C. Annealing of a RPV is only considered in case of highly sensitive materials and high DL fluences. Those materials, however, include the possibility of a remanent transition temperature shift after annealing. For the ex-USSR steel 15Kh12MFA used in WWER 440 reactors a remaining shift of 20 K is considered to which a safety margin of additional 20 K is added. Irradiation experiments with a high copper, high phosphorus, high nickel weld material have shown that high shift in transition temperature and a large drop in upper shelf energy occurs, Fig. 19. After annealing at 450 °C for 168 hours the upper shelf had completely recovered, but the remaining shift amounts to about 80 K. Fracture toughness data of this material give clear evidence that the shape of the Kic curve will not be maintained in the highly embrittled state as assumed in the Code, Fig. 20. This has already been indicated by other irradiated weld materials, Fig. 21 and specially heat treated base material to simulate irradiation embrittlement, Fig. 22 [38].

Annealing of RPVs has been mainly considered for reactors of the WWER 440 type with its high fluence. Ten WWER 440 reactors have been annealed by 1991 [39]. In several countries
feasibility studies for RPV annealing have been carried out on the basis of results of annealing behaviour studies. In Germany the licensing authorities have required a "stand by" full size annealing device for one particular PWR vessel in the late nineteen seventies [33]. At that time annealing was thought to be necessary for the particular reactor as a redundant measure. In the meantime, detailed fracture mechanics analyses in conjunction with other plant specific mitigation measures, such as flux reduction, preheating of cooling water and hot leg water injection have shown that annealing does not need to be considered for this particular vessel.

8 Summary

The generally increased safety requirements provided the impetus for a more detailed evaluation of the pressure vessel behaviour in some areas. Progress has been made in analysing transient conditions and to evaluate out the most severe load paths on a fracture mechanics basis. In this respect the expansion of the fracture mechanics analysis from the linear elastic regime into the elastic plastic regime was a decisive step towards describing component behaviour over the entire temperature regime. The better understanding of in-service degradation mechanisms and their quantification gave further elucidation of the conservatism in the safety analysis.

From the consequent research in this field together with feed-back from plant performance and surveillance results, implications can be derived for the design of new vessels (material optimization, increase in water gap and thus decrease in resulting neutron exposure) and for mitigation measure (ease of transient loading, reduction of neutron exposure) with regard to older plants. In this respect the pressurized thermal shock (PTS) event plays the most
important role. Associated with this loading condition the
defect size to be postulated and the reliable knowledge on
the material state are the crucial parameters on which the
life assessment is based.

Ageing parameters others than neutron irradiation, have not
considered in this chapter. Thermal ageing has minor effect
at LWR operating temperature and corrosion assisted crack
growth is an area which deserves separate consideration.
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Practical Experience of Annealing to Extend WWER Reactor Life Time 
11th International Conference on "Structural Mechanics in Reactor Technology (SMiRT) Session F12" 
Fig. 1: Elements of the DETERMINISTIC safety approach in the BASIS SAFETY CONCEPT

Fig. 2: Comparison of main RPV dimensions in the core belt line region

Fig. 3: Comparison of gross dimensions of different reactor pressure vessels

Fig. 4: Typical size of water gap and neutron flux at wet surface of BWR and PWR reactor pressure vessels

Fig. 5: Determination of reliable data on materials and stresses for life assessment

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Fig. 7: Fracture Toughness of the materials and load path for start up, normal operation and shut down

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**Fig. 10:** Stress intensity of an axial crack during resulting from transient as shown in Fig. 8

**Fig. 11:** Characteristic information needed for the safety assessment of an emergency core cooling event; circumferential flaw, pressure/temperature history according to Fig. 8
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   b) stress intensity depending on crack depth
   c) stress intensity as a function of time

**Fig. 12:** Determination of crack initiation and crack arrest events during ECC (schematically) on a fracture mechanics basis in the linear elastic regime

**Fig. 13:** Schematic demonstration of "fracture" and "no fracture" load paths as referred to the "warm prestress effect"

**Fig. 14:** Comparison of different internationally applied design curves to predict the shift in transition temperature as a function of neutron exposure
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Fig. 21: Fracture Toughness of high copper submerged arc weld in the unirradiated and irradiated state

Fig. 22: Fracture toughness of materials in conditions (chemical composition, forging and heat treatment) simulating the properties of irradiated material
BASIC PRINCIPLES

QUALITY
THROUGH OPTIMIZED
PRODUCTION, CONTROL
AND QUALIFICATION
- DESIGN
- MATERIAL
- MANUFACTURING

WORST CASE
- R & D WORK
- FAILURE
INVESTIGATION

VALIDATION / VERIFICATION
- CODES
- FM
- NDE

MULTIPLE PARTIES
TESTING
- INDEPENDENT
QUALITY
ASSURANCE

SURVEILLANCE
- IN-SERVICE
MONITORING AND
DOCUMENTATION
- IRRADIATION
SURVEILLANCE
- REPEATED NDE

INDEPENDENT REDUNDANCIES

BASIS SAFETY CONCEPT
EXCLUSION OF CATASTROPHIC FAILURE
INCREDIBILITY OF CATASTROPHIC FAILURE

FIGURE 1
<table>
<thead>
<tr>
<th>Reactor</th>
<th>Core Diameter</th>
<th>RPV Diameter</th>
<th>Pressure Vessel Diameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>FRG-2</td>
<td>( D_c = 1000 \text{ mm} )</td>
<td>( D_t = 1000 \text{ mm} )</td>
<td>( D_c = 1366 \text{ mm} )</td>
</tr>
<tr>
<td>KWO</td>
<td>( D_c = 2541 \text{ mm} )</td>
<td>( D_t = 2541 \text{ mm} )</td>
<td>( D_c = 1366 \text{ mm} )</td>
</tr>
<tr>
<td>KKS</td>
<td>( D_c = 3234 \text{ mm} )</td>
<td>( D_t = 3234 \text{ mm} )</td>
<td>( D_c = 1366 \text{ mm} )</td>
</tr>
<tr>
<td>GKN</td>
<td>( D_c = 3234 \text{ mm} )</td>
<td>( D_t = 3234 \text{ mm} )</td>
<td>( D_c = 1366 \text{ mm} )</td>
</tr>
<tr>
<td>Mülheim-Kär.</td>
<td>( D_c = 3710 \text{ mm} )</td>
<td>( D_t = 3710 \text{ mm} )</td>
<td>( D_c = 1366 \text{ mm} )</td>
</tr>
<tr>
<td>KWB-B</td>
<td>( D_c = 3450 \text{ mm} )</td>
<td>( D_t = 3450 \text{ mm} )</td>
<td>( D_c = 1366 \text{ mm} )</td>
</tr>
<tr>
<td>KRBB-A</td>
<td>( D_c = 3748 \text{ mm} )</td>
<td>( D_t = 3748 \text{ mm} )</td>
<td>( D_c = 1366 \text{ mm} )</td>
</tr>
</tbody>
</table>
after Kraftwerk Union

Gross Dimensions

MZFR
50 MWt-\(D_2O\)

CNA
300 MWt-\(D_2O\)

CNA 2
750 MWt-\(D_2O\)

KWO
300 MWt-\(H_2O\)

KCB
450 MWt-\(H_2O\)

KKS
600 MWt-\(H_2O\)

KWB
1200 MWt-\(H_2O\)

FIGURE 3
<table>
<thead>
<tr>
<th>REACTOR TYPE</th>
<th>POWER (MW)</th>
<th>WATER GAP (mm)</th>
<th>n-FLUX $cm^{-2} s^{-1}$</th>
<th>DL-FLUENCE $cm^{-2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>BWR</td>
<td>900</td>
<td>897</td>
<td>$1.2 \cdot 10^9$</td>
<td>$1.2 \cdot 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>1100</td>
<td>748</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1316</td>
<td>972</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PWR</td>
<td>357</td>
<td>336</td>
<td>$3.5 \cdot 10^{10}$</td>
<td>$3.5 \cdot 10^{19}$</td>
</tr>
<tr>
<td></td>
<td>672</td>
<td>430</td>
<td></td>
<td>$1.4 \cdot 10^9$</td>
</tr>
<tr>
<td></td>
<td>855</td>
<td>568</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1300</td>
<td>781</td>
<td>$5 \cdot 10^{18}$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1308</td>
<td>456</td>
<td>$2 \cdot 10^{10}$</td>
<td>$2 \cdot 10^{19}$</td>
</tr>
</tbody>
</table>

FIGURE 4
FIGURE 5
1: $T = (RT_{NDT} + 33K) - 110K$, $\sigma = 0.1\sigma_y$
2: $T = RT_{NDT}$, $\sigma = 0.2\sigma_y$
4: $T = RT_{NDT} + 33K$, $\sigma = \sigma_y$

**Figure 6**
FIGURE 7

Operating conditions
\[ K = 2K_{Ip} + K_{Ith} \]
FIGURE 8
FIGURE 9

ECC load path
\(\alpha/W = 0.60\)

STRESS INTENSITY vs. TEMPERATURE

MPa\(\sqrt{m}\)

\(K_{lc}\) (initial)

\(K_{lc}\) (adjusted)
FIGURE 10

ECC load path
a/W = 0.60

$K_{ic}$ (initial)
$K_{ic}$ (adjusted)

STRESS INTENSITY

MPa$\sqrt{m}$

TEMPERATURE

0
100
200
300 °C

0.06
0.20
Figure 11

Stress Intensity

Temperature

Stress Intensity

Time
FIGURE 12

- Crack arrest: $K_1 = K_{Ia}$
- Crack initiation: $K_1 = K_{Ic}$
- Final arrest: $K_1 > K_{Ic}$

Critical crack depth $a/W$ vs. time.

$K_{Ia}$ and $K_{Ic}$ are critical stress intensity factors for arrest and initiation, respectively.
FIGURE 13

"no fracture" load path

"fracture" load path
FIGURE 15

PTS TEST NKS3
initial crack length
\(a_0 = 62.8\) mm

\(J_R\) - curve
CT - specimen
test temp. 220 °C
\(J_{0.15} = 133\) N/mm

\(\Delta a\) (NKS3) \(= 35\) mm
**TENSILE PROPERTIES AT 20°C**

<table>
<thead>
<tr>
<th>Material</th>
<th>$\sigma_{\text{y}}$ MPa</th>
<th>$\sigma_{\text{u}}$ MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 MnMoNi55</td>
<td>440</td>
<td>600</td>
</tr>
<tr>
<td>NKS1</td>
<td>490</td>
<td>650</td>
</tr>
<tr>
<td>NKS2</td>
<td>560</td>
<td>720</td>
</tr>
<tr>
<td>NKS3</td>
<td>500</td>
<td>800</td>
</tr>
<tr>
<td>NKS4</td>
<td>1100</td>
<td>1180</td>
</tr>
<tr>
<td>22 NiMoCr37</td>
<td>440</td>
<td>600</td>
</tr>
<tr>
<td>NKS1</td>
<td>490</td>
<td>650</td>
</tr>
<tr>
<td>NKS2</td>
<td>560</td>
<td>720</td>
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<tr>
<td>NKS3</td>
<td>500</td>
<td>800</td>
</tr>
<tr>
<td>NKS4</td>
<td>1100</td>
<td>1180</td>
</tr>
<tr>
<td>17 MoV84</td>
<td>440</td>
<td>600</td>
</tr>
<tr>
<td>NKS1</td>
<td>490</td>
<td>650</td>
</tr>
<tr>
<td>NKS2</td>
<td>560</td>
<td>720</td>
</tr>
<tr>
<td>NKS3</td>
<td>500</td>
<td>800</td>
</tr>
<tr>
<td>NKS4</td>
<td>1100</td>
<td>1180</td>
</tr>
</tbody>
</table>

**EXPERIMENTAL BLUNT LINES**

1. **20 MnMoNi55 (KS 17, T = 80°C)**
   - USE = 200 J
   - $\sigma_{\text{y}} = 395$ MPa, $\sigma_{\text{u}} = 519$ MPa

2. **22 NiMoCr37 (KS 01, T = 65°C)**
   - USE = 90 J
   - $\sigma_{\text{y}} = 420$ MPa, $\sigma_{\text{u}} = 560$ MPa

3. **17 MoV84 mod. (KS 22, T = 300°C)**
   - USE = 90 J
   - $\sigma_{\text{y}} = 974$ MPa, $\sigma_{\text{u}} = 1070$ MPa

4. **22 NiMoCr37 mod. (KS07, T = 80°C)**
   - USE = 40 J
   - $\sigma_{\text{y}} = 620$ MPa, $\sigma_{\text{u}} = 744$ MPa

**FIGURE 16**
FIGURE 17

FLUENCE

TIME

DLT

initial design

after flux reduction program
FIGURE 18
FIGURE 19

SMA weld
Cu = 0.37 %
P = 0.017 %
Ni = 1.23 %

2.97 \times 10^{23} \text{ m}^{-2}
annealed 450^\circ\text{C}/168\text{ h}

unirradiated

2.99 \times 10^{23} \text{ m}^{-2}
as irradiated
FIGURE 20

KS01 WELD

acc. to KTA 3201.1
for RT_{NDT} = -12 °C

unirradiated

irradiated

unirradiated

irradiated

\Phi \quad \sigma_y \quad \sigma_u
\begin{align*}
2,0 \cdot 10^{19} & \quad 890 & \quad 935 \\
4,33 \cdot 10^{19} & \quad 925 & \quad 965
\end{align*}
FIGURE 21

SA weld (P390)
NiCrMo 1UP
0.27 % Cu

unirradiated

$K_{IR}$ for $RT_{NDT} = -12^\circ C$

(acc. to KWU)
FIGURE 22

The graph illustrates the fracture toughness $K_{IC}$ as a function of temperature. Different materials are represented by various symbols and lines:

- **22 NiMoCr 37**
- **KS 02**
- **KS 07** (modified)
- **KS 01**
- **17 MoV 84**

The graph shows $K_{IR}$ for $RT_{NOD} = 0^\circ C$. The temperature range is indicated from -200 to 400 degrees.
3. REACTOR PRESSURE VESSEL MATERIALS

K. Suzuki, The Japan Steel Works, Ltd., Muroran Plant

1 INTRODUCTION

The demands placed on reactor pressure vessel (RPV) steels are severe. They must be manufactured in required sizes and thicknesses, be of sufficient strength and toughness, show little deterioration under irradiation, allow the production of high quality welds and be compatible with the cladding. This Chapter refers to non-WER pressure vessels, but many of the guiding principles described here apply to that particular case.

Starting with carbon steel plates and forgings for conventional boiler drums in the dawn of commercial light water reactors (LWRs) followed by a few changes thereafter, SA533 and SA508 and similar grade steels have become well established [1]. Both grades are of the vacuum treated, quenched and tempered type of 600 N/mm² strength class, which is not the highest level in weldable structural steels.
The specific requirements for RPV steels are to give, even to large-size component materials of RPVs, higher values of the following properties:

- uniformity and isotropy of mechanical properties, including less mass effect in the mid-section
- fracture toughness
- internal defects
- weldability
- resistivity to neutron irradiation embrittlement

Some further items were added during the past two decades primarily for the purpose of easier execution of non-destructive examinations both pre-service and in-service. These are:

- fewer weld seams in RPV
- larger and more integral design of component materials

The requirements have been steadily realized.
With respect to the weld, improvements have been made by adequate selection of flux in combination with requirements on the chemical composition of the weld wire, welding parameters such as weld bead size and sequence to minimize the coarse grained areas in the heat affected zone (HAZ) [2].

2 HISTORICAL REVIEW [3]

Many of the earlier plants were constructed of so-called carbon or mild steels (usually in normalized and tempered, NT, condition), but several of these were prototypical units and applications have moved to the widely accepted low alloy manganese, molybdenum, nickel, quenched and tempered (QT) grades of higher strength. Early gas cooled reactors were also of carbon steel, but these were replaced in later versions by prestressed concrete vessels.

Most of the reactors operating at mid-1973 were constructed of the manganese-molybdenum steels in QT condition. In practice, the use of the low alloy steels has predominated. For example, all of the Japanese reactors of BWR and PWR types have A533-B Class 1 (QT) pressure vessels except Tsuruga which uses A302-B (QT) and JFQR-2 with A302-B in NT condition [4]. The sole Japanese gas cooled reactor (Tokai Nuclear Power Station) was contained in a vessel of a Japanese C-Mn steel. Typical composition data for the Japan Steel Works (JSW), Ltd. steel used is C-0.10%, Mn-1.30%, Si-0.25%, P-
0.014\%, and S-0.018\% (equivalent to the UK steel, BS NDIV originally planned for this reactor vessel). Special care was taken to minimize phosphorus and sulphur and to refine grain size, thereby enhancing notch toughness. In addition, an experimental programme was conducted to develop the best welding electrode material for joining plates of the Tokai vessel [5]. The relative content of manganese and silicon was determined for optimum toughness of welded structures.

The Agesta reactor of Sweden was contained in a carbon steel, equivalent to ASTM Type A212-B (a carbon-silicon-manganese steel), the Oskarshamn-1 vessel steel was equivalent to ASTM Type A302-B [6] (a manganese-molybdenum steel). The A212-B steel was also used in one of the early U.S. reactors, Indian Point-1, and a limited number of experimental reactors.

In the Federal Republic of Germany (F.R.G.) the steel designations match national conventions, but most water reactor steels were quite similar to U.S. grades A508-C1.1 for forging and A533-B for plate steel [7]. And the most RPVs were strongly dependent on the 22 NiMoCr 37 composition (similar to A508, C1.2 : a nickel-chromium-molybdenum steel). 22 NiMoCr 37 steel was used in F.R.G. until 1976 and fulfilled all requirements [8]. However, this type of steel exhibited some susceptibility to stress relief cracking and underclad cracking [9-11]. By this reason the use of 20 MnMoNi 55 steel (similar to A508, C1.3 : a manganese-molybdenum-nickel steel)
was increased.

Little basic difference in steel type exists among the vessels of reactors now operating throughout the world. However, certain metallurgical differences of crucial importance are identificable, but other considerations such as neutron flux and fluence, irradiation temperature, stress state etc. have equal or greater bearing on radiation embrittlement sensitivity. Relative to radiation embrittlement, however, it is important to realize that all steels used to date have the same basic weakness. They are susceptible to radiation hardening, increases in strength, and reduction to fracture toughness. Thus, it is appropriate, even necessary, to generalize initially in describing radiation embrittlement of vessel steels.

The current choice of material is of specific A-533B, Cl.1 alloys for plates and A-508, Cl.3 alloys for forgings (Table 1) [12]. These are very similar steels of the low alloy manganese-molybdenum type (A302-B) with nickel added. Significant differences have been developed between the old and new steels. Major differences are in improved strength and toughness for the new steels with a change of micro-structure from basically pearlitic to tempered bainitic to tempered martensitic or mixed bainitic-martensitic microstructure. Chemically, the new steel is purer, that is, it contains smaller amounts of residual or tramp elements, because steel-making procedures incorporating vacuum degassing steps are applied routinely in their production. These steps, producing significant
changes, are considered improvements from the view of radiation embrittlement, but the resulting implication is that the greatest concern must be addressed to the older plants, where our knowledge of the alloys used and the service conditions is least. Knowledge about welds is probably both the most critical and the most limited. Emphasis then must be placed on learning more about the old systems materials, especially welds, and improving the new (future) materials.

3 BASE MATERIAL

3.1 Chemical Composition [12]

Specifications for the structural steels which are extensively used for the LWR components in Germany and in the United States are listed in Table 1 [12].

Some basic investigations were conducted to confirm the effect of chemical composition on 20 MnMoNi 55 in addition to the basic studies for heavy forgings [10, 13]. Fig.1 shows the continuous cooling transformation curves (CCT curve) for this steel with 0.17% and 0.20% C. The differences in ferrite, pearlite and bainite transformation range are observed due to a little difference in C content. Also the effect of Mn, Ni, Mo on the hardenability was investigated, but the results do not indicate substantial differences in the CCT curve.
In order to maintain sufficient toughness of large forgings at lower temperature, it is important to provide fine grain as well as proper structure. There are some procedures to refine the grain size, and it is an usual procedure to add Al. During the quenching of large and massive forging, austenitizing requires considerably long time to obtain uniform heating. In this case, the presence of AlN is strongly effective in preventing grain growth.

V is known as a predominant element to enhance the tensile properties of material. However, the toughness and weld crack sensitivity of the material are also affected by the addition of this element. Thus no V should be added, even though the maximum content of 0.03 or 0.05% is allowed in the material specifications. It is said that Cu and P affect the irradiation damage which is evaluated by the shift in Charpy impact curve [14]. In response to this requirement, the target values of Cu and P content of 0.08% max. and 0.008% max. respectively are maintained.

Concerning weldability of the steel, liquation cracking and stress relief cracking in particular, crack promoting elements and threshold values for cracking occurrence were investigated. Cracking can be avoided by limiting the Mo content, the elements P, S, Cu, Sn, N, As, Co and Al as well [11].
3.2 Steel and Ingot Making [15]

3.2.1 Manufacturing Process of Large Ingots

(1) History of the production of large ingots

In Fig. 2, major changes in the history of the production of large ingots in JSW are summarized. It also shows the transition of maximum ingot size. Before Bochumer Verein-type vacuum casting facilities were installed in 1959, steel was melted by acid open hearth furnaces to minimize hydrogen pick up and the steel cast in air. After the installation, acid open hearth was replaced by basic open hearth and electric arc furnace (EAF), because hydrogen removal was made possible during vacuum casting. In order to obtain higher degree of vacuum during casting, a steam ejector was introduced in 1970. In 1973 a holding furnace was installed to replace the open hearth. According to the demand from industries, sizes of ingot became larger year after year. In 1969 a world largest 400 ton ingot was produced, and the record was soon renewed by 500 ton ingot in 1971, 570 ton ingot in 1980, then 600 ton ingot in 1985.

(2) Installation of ladle refining furnace (LRF)

In 1980, to meet higher requirements for the record 570 ton ingots of nuclear vessel application, vacuum facilities were installed to the holding furnace to convert it into a ladle refining furnace.
Fig. 3 shows a schematic outline of the furnace. It has one heating system and two vacuum systems so that two vessels of molten steel can be treated at a time. Combining vacuum treatment at the LRF with conventional tap degassing the "double degassing" process was developed for the production of large ingots up to 600 ton. Fig. 4 shows the production sequence of a 600 ton ingot. By this process the quality of products such as for nuclear applications was remarkably improved.

(3) General description of large forging ingot

Fig. 5 shows a sulphur print of the longitudinal section of a 75 ton ingot (left) and the description of segregations and solidification structures of the ingot (right). In a sulphur print solute-enriched portions are marked dark. In the region called "branched columnar zone", string-shaped A-segregates are observed and in the region called "equiaxed zone", V-segregates are observed. In these two regions, microporosities are easily formed. Non-metallic inclusions, especially oxides, are often observed in the region called "sedimentation zone" at the bottom of an ingot and this may lead to the rejection of the entire ingot. Distribution of C and of other alloying elements is not uniform. Generally speaking, these elements are rich in the top side of an ingot and poor in the sedimentation zone. In the following section, technical improvements to reduce these heterogeneities will be described.
3.2.2 Recent Improvement in Steelmaking and Ingotmaking

(1) Removal of P and S

Low S and P contents about 0.004–0.008% can be obtained by conventional EAF process, however, LRF process was introduced to obtain extremely low S and P contents in the liquid steel. Fig.6 shows a flow chart of the refining process by LRF. A high degree of desulphurization up to 90% can be obtained by using high basic slag and extremely low content of S less than 10ppm can be obtained. The lowest recorded S content is 2 ppm. Since P cannot be removed by LRF treatment, it is essential to cut off oxidizing slag from EAF to prevent rephosphorization. Fig.7 shows the changes of P content and S content when extremely low content of P+S<0.003% was obtained by EAF+LRF process.

(2) Control of Cu, As, Sn and Sb

Impurities such as Cu, As, Sn and Sb are kept sufficiently low by selecting grades of scrap.
(3) Degassing (H, O) and reduction of non-metallic inclusions

(a) Effect of LRF

Degassing is achieved very effectively by LRF process due to high vacuum and intensive stirring of molten steel. For the stirring, Ar gas is blown into molten steel through porous plug at the bottom of refining ladle. Energy density of Ar stirring \( i_M \) (Watt/ton) is expressed by:

\[
i_M = \frac{6.18QT_L}{M_L} \left\{ \ln\left(1+\frac{H}{148Pa}\right) + \left(1-\frac{T_0}{T_L}\right) \right\}
\]

Where

- \( Q \) is flow rate of Ar-gas (Nm\(^3\) /min),
- \( Pa \) is pressure at the surface of molten steel (atm),
- \( T_L \) is temperature of molten steel (K),
- \( T_0 \) is temperature of Ar gas before blowing (K),
- \( M_L \) is weight of molten steel (ton),
- \( H \) is bath depth (cm),

and \( t \) is treatment time (sec).

Fig. 8 shows the effect of \( i_M \cdot t \) on the degree of hydrogen removal. In this process a high degree of hydrogen removal, up to 80%, is obtained by increasing the "stirring" intensity \( i_M \cdot t \). Fig. 9 shows a change in hydrogen content during LRF process. When molten steel is cast by bottom pouring, about 0.5ppm of hydrogen pick-up should
be expected, whereas if it is cast by mold stream degassing, low hydrogen levels of 0.4 to 0.6ppm are obtained. Fig.10 shows the effect of \( \dot{M} \cdot t \) on the degree of deoxidation. At the region of low \( \dot{M} \cdot t \), degree of deoxidation increases with increasing \( \dot{M} \cdot t \), however, it decreases with further increase of \( \dot{M} \cdot t \). It is attributed to the erosion of brick or suspension of slag in the melt. By controlling the stirring intensity \( \dot{M} \cdot t \) around 80-100x10\(^3\) J/ton, low oxygen level of less than 20ppm is obtained.

(b) Effect of double degassing

As described before, the double degassing process (vacuum treatment at LRF plus mold stream degassing) is very effective for degassing of molten steel and was applied to the production of two 570 ton ingots of nuclear applications. Fig.11 shows the check analyses of hydrogen and oxygen of these ingots. Low hydrogen contents of less than 1ppm and oxygen contents around 10 to 20ppm are obtained in the body of ingots. From this low gas content the effect of double degassing is evident.

(4) Reduction of macrosegregates

For 0.7% carbon steel, critical condition for the formation of A-segregates is expressed by:

\[
\varepsilon \cdot R \leq 8.75
\]
Where

e is cooling rate in the radial direction of an ingot (°C/min)
and R is solidification rate in the radial direction of an ingot (mm/min).

By changing the constant term in the right side, the equation is applicable to other kinds of steel. If carbon content and silicon content are high, the constant is large and A-segregates are easily formed. Mo and Cr have the opposite effect. In general it is difficult to control e and R during the solidification of an actual ingot, therefore, chemical compositions are adjusted to minimize the formation of A-segregates. Since the driving force for the formation of A-segregates is considered to be the density difference between solute-enriched liquid and bulk liquid, the density difference \( \Delta \rho_L \) is calculated assuming solute enriched liquid is that at fraction solid 0.3 \( \Delta \rho_L \) varies with various steels and if \( \Delta \rho_L \) is large A-segregates are easily formed. The relation between \( e \cdot R^{1.1} \) and \( \Delta \rho_L \) is shown in Fig.12. If type of steel is determined, \( \Delta \rho_L \) is calculated and critical value \( e \cdot R^{1.1} \) is obtained from Fig.12 and then the position where A-segregates first starts to form is estimated.

V-segregates are formed due to rapid shrinking along the axis of an ingot at the final stage of solidification. V-segregates can be reduced by making the taper of ingot larger and height-to-diameter ratio H/D smaller.
(5) Reduction of microporosity

Microporosities are formed in the A-segregation zone and V-segregation zone. If sizes of pores are large, it may result in the rejection of the product. Therefore, it is very important to design the optimum shape of the ingot to reduce porosity. Based upon investigations on 3 - 220 ton ingots of carbon steel and low-alloy steel, critical conditions for the formation of microporosity were determined as shown in Fig. 13. Large porosity tends to form especially in the V-segregation zone.

The length of the porosity zone $V_y$ (mm) is expressed as a function of height-to-diameter ratio $H/D$ as shown in Fig. 14. In the figure, total weight of ingot is kept constant and $H/D$ is changed and it is known that $V_y$ decreases with decreasing $H/D$. Porosity along the axis of ingot can be reduced also by increasing the weight of the feeder head. In this case the rate of axial solidification becomes slower and the formation of large porosity are suppressed. Based upon the above mentioned studies, optimum ingot shapes were designed and satisfactory results obtained.

(6) Reduction of carbon segregation

Quantitative understanding of carbon segregation of ingots is very important. However, studies to date have yielded improvements, because carbon segregation is affected by many complicated factors. The equation given below shows the result of multiple regression
analysis on 75 to 570 ton Mn-Mo-Ni steel ingots (number of ingot: 81).

Segregation Rate \[ \frac{-(\%C)-(%C)_{ladle}}{(%C)_{ladle}} \times 100 \]

\[ = 38.5 + 0.132(Wt) - 72.8(F.H.R.) - 100(\Delta C \text{ multiple}) \]

Where

- \((\%C)\) is check carbon analysis in the centre of (ingot body/feeder head) boundary face (wt%),
- \((%C)_{ladle}\) is ladle analysis of carbon content (wt%),
- \((Wt)\) is total ingot weight (ton),
- \((F.H.R.)\) is feeder head ratio defined as (weight of feeder head) / (weight of ingot body)

and \((\Delta C \text{ multiple})\) is carbon content difference at multi-pouring practice defined as

\[ \sum_{i=1}^{\infty} \left( \frac{|(\%C)_i - (%C)_{ladle}| \times W_i}{\sum_{i=1}^{\infty} W_i} \right) \]

where \(W_i\) is the weight and \((\%C)_i\) is the carbon content of molten steel cast as "i"th time.

From the equation above, it is known that large feeder head ratio and large carbon content differences favour the minimizing of carbon segregation of ingots. Fig.15 shows the distribution of carbon content along the axis of a 140 ton and a 180 ton rotor ingot cast by multi-pouring method.

It is shown that these distributions are uniform compared with ingots cast by conventional methods. Fig.16 shows the distribution
of carbon content in the longitudinal section of 570 ton ingot for nuclear application. The carbon segregation is not as severe as in a "huge" ingot. It is considered that this results from the pouring process. If this ingot had been poured without a carbon content difference, the carbon segregation would have been about 10% (0.02%C) higher according to the estimation by the equation given above.
3.3 Forging Process [16]

Figs. 17 and 18 show the combined vessel flange and nozzle belt forging of KWU/1300MWe pressurized water reactor pressure vessel (PWRPV) made from a 400 ton ingot and the combined vessel flange and nozzle belt forging of WEC/157" PWRPV made from a 500 ton ingot (developed by COCKERILL) as compared with the conventional one, respectively. These integrated flange forgings were hot worked by a 10,000 ton forging press. Fig. 19 illustrates the forging processes for the flange forging made from a 500 ton ingot. Sufficient discard was made from each end of the ingot to insure that only sound metal enters the completed forging. After piercing of ingot core, repetition of enlarging and upsetting was performed to close possible porosity inside the ingot.

The forging ratio and repetition of enlarging and upsetting operations play an important role to improve the mechanical properties. Fig.20 shows the improved impact value due to repetition of forging. Therefore, from the results a minimum forging ratio of 1.5 should be required.

Anisotropy of forged material was intensively investigated by W. Coupette in 1940th [17]. Fig.21 shows the effect of forging ratio and anisotropy of 20 MnMoNi 55 steel. Compared with Coupette's data, anisotropy of forging at present time seems to be much smaller.
To estimate the mechanical properties due to differences in forging ratio, the use of the logarithmic strain concept is convenient. The logarithmic strains in three directions are defined in Fig. 22. From the results of mechanical tests for different components, the relation between logarithmic strain and mechanical properties, such as tensile properties and Charpy V-notch impact value, can be obtained, as shown in Fig. 23. Tensile strength, yield strength and elongation, at both room temperature and 350°C, are not so significantly related to logarithmic strain, but the reduction of area is increased with the increase of logarithmic strain. Charpy V-notch impact values are also increased with the increase of logarithmic strain. The integrated flange forgings made from a 400 ton and a 500 ton ingot respectively were manufactured under the above consideration. The logarithmic strains of integrated flange forgings were as given below:

a) Shell flange: \( \varepsilon_t = 0.96 \)
\[ \varepsilon_2 = -0.28 \]
\[ \varepsilon_1 = -0.68 \]

b) Mono-block vessel flange: \( \varepsilon_t = 0.81 \)
\[ \varepsilon_2 = -0.18 \]
\[ \varepsilon_1 = -0.63 \]

Therefore, anisotropy in three directions is expected to be minimized.
3.4 Heat Treatment [16]

Heat treatment technology for large forgings is based on the experimental investigations and experiences of actual operation in addition to heat treatment theory. The important points to be considered are the segregation in large ingots, mass effects, hydrogen induced defects, temper embrittlement and residual stresses.

Flake, ghost crack and fish eye etc. are well known hydrogen induced defects. Recently, the problems in large forgings due to hydrogen are remarkably decreased by the improvement of vacuum treatment technology.

However, it is still necessary to consider the prevention of hydrogen induced defects depending on the size and grade of materials. In general, the measures taken for the prevention of hydrogen induced defects are as follows.

- Slow cooling after forging or rolling
- Isothermal annealing (Pearlite transformation)
- Normalizing and tempering (Bainite transformation)

For large pieces of low alloy steel, normalizing and tempering technique is usually applied. Preliminary heat treatment after forging shown in Fig.24 is one example adopted in Germany for preventing hydrogen flake [18]. In this procedure, the bainite transformation is mainly performed after hot working of the forging.

At present, in JSW, the basic idea behind this heat treatment diagram is also being adopted for the preliminary heat treatment of large forgings such as integrated
flange forging made from 20 MnMoNi 55 steel under the consideration of CCT curve shown in Fig.1.

After preliminary heat treatment (normalizing and tempering), the forgings are machined to a simple cross-section for performing ultrasonic examination. After successful examination, the forgings are contour machined further for quenching and tempering in order to obtain a good quenching effect. The austenitizing temperature of 870 to 910 °C is selected to minimize the grain growth and a faster quenching operation is for a more complete transformation results.

3.5 Properties of Integrated Flange Forgings [16]

3.5.1 Metallurgical Homogeneity

Segregation is of vital importance with respect to welding and neutron irradiation. This is particularly true, if the carbon content is below 0.17%, the mechanical strength requirements are not satisfied, and if the carbon content is higher than 0.24%, the weldability is deteriorated due to hardening of the heat affected zone (HAZ).
Fig. 25 shows the carbon distribution on the integrated flange forgings made from a 400 ton and a 500 ingots. The carbon content, which contributes primarily to mechanical properties and weldability, was well controlled and the concentration was $0.20 \pm 0.02\%$ even for a 400 ton or a 500 ton ingot. This achievement was brought about by controlled ingot making technique.

3.5.2 Mechanical Properties

(1) Through-thickness mechanical properties

Distributions of mechanical properties through wall thickness are shown in Fig. 26 for both KWU's shell flange and COCKERILL's monoblock vessel flange. The curves show the improved uniformity of mechanical properties. Even at half-thickness, the tensile properties satisfy the specified values.

(2) Homogeneity of mechanical properties at various height locations

Fig. 27 shows the comparison of mechanical properties at top, middle and bottom of KWU's shell flange forging. No significant difference in height locations is observed for both strength and toughness values. Smaller differences in mechanical properties at various tangential locations ($120^\circ$ apart) is obtained. Thus, homogeneity
of mechanical properties is brought about by the homogeneity of chemical composition and an accurate and narrow range temperature control at quenching and tempering.

(3) Directionality of mechanical properties

Fig. 28 shows the directionality of tensile properties in three directions (tangential, axial and radial) for KWU's shell flange. There is no difference in both yield and tensile strength. Slight differences in elongation and reduction of area are found, but the difference is not remarkable. Fig. 29 shows the Charpy V-notch impact properties transition curves for tangential, axial and radial directions for KWU's shell flange and COCKERILL's mono-block vessel flange. Impact energy in all three directions is high enough, and any directionality of properties is not significant.

The homogeneity in mechanical properties described above is brought about by decreasing the micro-segregation and non-metallic inclusions. This was accomplished by the deeper understanding of steel making and using suitable forging techniques.
4 WELD

4.1 Structural Weld [2]

4.1.1 Weld Metal

(1) Requirement

Since the plates and forgings must be welded together, it is obvious that the mechanical property requirements of the welded region and the associated heat affected zone (HAZ) can be no less demanding than those of the base material itself, particularly as experience shows that the most likely location for flaws is in the weld and HAZ. The ASME requirements for weld mechanical properties and procedures are somewhat dispersed but appear mainly in Sections II, III (NB-2300 and NB-2400), and IX of the Code. They appear to be less specific than those for plates and forgings but there is a requirement that all weldments should conform to all of the minimum mechanical property specifications for the materials which are joined by welds. This is clearly a desirable requirement. Procedures and requirements for Charpy impact tests in particular are given mainly in the ASME Code Section III. Minimum tensile and notch toughness requirements which are specified in the ASME Codes are summarized in Table 2. Their extension to cover the deterioration of properties under irradiation is implemented in 10 CFR 50 Appendix G and in the ASME
Code Section I Appendix G. Section II (Part C) of the ASME Code includes general specifications for welding materials and methods while Section IX deals with qualifying standards for welding procedures as well as with the qualifications of the welders themselves.

(2) Chemical composition

Deposit compositions of manual metal arc welds, associated with nuclear vessels fabricated in Europe and the USA (Tables 3 and 4), show that consumables have been employed which are capable of alloying the deposit with Mn-Mo, Mn-Ni-Mo and Mn-Ni-Cr-Mo. Deposit strengths in the post-weld heat treated condition matching that of the base steel can be achieved by various combinations and levels of the elements carbon, manganese, nickel, chromium and molybdenum, and this explains the variety of deposit analyses that can be found in the literature. A similar situation is evident for submerged arc welds where wires alloyed with Mn-Mo, Mn-Ni-Mo or Mn-Ni-Cr-Mo have been used by fabricators to achieve the required deposit strengths (Tables 3 to 5). However, the choice of flux is a very important factor governing the deposit composition of submerged arc welds, influencing in particular the carbon, silicon and manganese levels and the impurity element levels of sulphur, phosphorus and oxygen. Fused fluxes of the calcium silicate type are favoured by many nuclear vessel fabricators. These tend to give lower carbon levels but higher silicon and oxygen levels in the deposit compared with
agglomerated fluxes [22], which are chemically more basic and favoured by some European fabricators, because the associated deposits generally have higher toughness. Most flux types will add small amounts of phosphorus but the basic fluxes are capable of lowering deposit sulphur levels, unlike the fused calcium silicate fluxes.

For the beltline regions, where it is necessary to limit the copper content to reduce the sensitivity to irradiation embrittlement, it is necessary to depart from the practice of using copper-coated electrodes. Hawthorne [21] has shown that copper contents below 0.1 wt% are readily achieved in sound welds providing the welding wires are protected from corrosion before use.

(3) Welding procedure

The main aims of evolving a satisfactory welding procedure are to obtain the required mechanical properties in the weld, namely strength and toughness, to produce a weld free from ultrasonic 'indications' which would require its repair and to avoid the existence of cracks which would also require repair but if remaining undetected could act as the nucleus for fracture processes.
(4) Defects in the weld metal

The welding process can lead to a variety of defects in the weld metal or adjacent HAZ of the parent material. Much is known about the mechanisms of formation of these defects and how to avoid or minimise their occurrence by control of material composition and fabrication procedure.

There are three forms of cracking that are potential problems in weld deposits made in low alloy steels; solidification cracking, reheat cracking and hydrogen induced cracking. There are no published reports of incidences in nuclear vessel fabrication of the first two forms, indicating that the consumables and procedures normally selected in European and American nuclear fabrication shops have adequate resistance to these types of cracking. The metallurgical factors controlling solidification and reheat cracking in weld deposits are reasonably well understood [23, 24].

Hydrogen induced cracking can occasionally be found in multi-pass deposits in situations where the welding procedure and shop floor storage and handling of consumables are not sufficiently controlled. The cracks occur typically in arrays and are often transverse to the line of the weld and inclined at about 45° to the weld surface. Sometimes referred to as chevron cracks, the individual cracks have a zigzag appearance and can be transgranular or intergranular with respect to the microstructure. They may be up to 40 mm in their
longest dimension, but are often confined to a single weld pass and, in these situations, are less than about 5 mm in length. The factors controlling this form of cracking are similar to those for hydrogen cracking in the HAZ, with cracking being more likely in situations where the weld metal hydrogen content is high, the restraint is high and the weld deposit microstructure is susceptible, this normally means that the deposit is of high hardness [25-28]. However, cracking can occur in weld metals at significantly lower hardnesses than would be associated with cracking in the HAZ. The relationship between weld hardness, microstructure and susceptibility to cracking is the subject of current research but there is sufficient knowledge at present time to specify adequate procedures for fabrication in order to avoid cracking.

For nuclear vessels, weld metal hydrogen induced cracking must be regarded as a potential problem in submerged arc and manual metal arc weld deposits. For welds of the former type, the type of submerged arc flux used is an important factor to be considered, since there is evidence that agglomerated flux types have been associated with a greater tendency to weld metal hydrogen cracking than fused fluxes [29]. Crack-free welds can be produced with either fused or agglomerated fluxes but the latter type needs more careful shop floor control. As stated the choice of a fused flux, to provide a good resistance to weld metal hydrogen cracking, brings with it the penalty of a generally lower weld metal notch toughness,
due principally to the higher non-metallic inclusion content of such deposits when compared with those made with agglomerated fluxes which are more basic. It must also be noted that the level of hydrogen in the weld metal is partly determined by wire cleanliness and thus wire quality as well as flux quality is important.

4.1.2 Heat Affected Zone

(1) Defects in the weld metal

As for the weld metal, there are three forms of cracking that are potential problems in the HAZ, namely liquation cracking, reheat cracking and hydrogen induced cracking.

(a) Liquation cracking

Liquation or hot cracking is a mode of intergranular cracking occurring at elevated temperature in the initial welding thermal cycle, i.e. before post-weld heat treatment. It is associated with weak grain boundary zones of reduced melting point material containing enhanced concentrations of impurities particularly sulphur, and occurs preferentially in localised regions of positive segregation [30, 31]. Liquation cracking has been observed in the HAZ of 22 NiMoCr 37 (similar to SA-508 Class 2) and in 20 MnMoNi 55 (similar to SA-508 Class 3 and SA-533B Class 1) but none has been
reported for weld metal itself [31-34]. In examination of 120 weldments from test plates, production weld prolongations and actual components, about 30% contained liquation cracks with dimensions typically of a few grain diameters (i.e.< 1 mm).

The control of liquation cracking is primarily a question of control of bulk and local purity, because other parameters such as welding technique and heat input appear to be of second order significance [30]. Restrictions on copper, tin, phosphorus, sulphur and arsenic bulk impurity content to the levels given in Table 6 have been proposed [33] together with a 'threshold cracking criterion' such that cracking is likely to occur if two or more elements exceed these values. Whilst this criterion successfully characterises the cracking susceptibility of the weldments examined, it must be conceded that local regions of segregation may occasionally lead to small isolated liquation cracks in material satisfying these bulk compositional requirements. Liquation cracking susceptibility as a phenomenon generic to weldable steels, is known to decrease with increasing manganese-sulphur ratios [35]. Although all modern PWR pressure vessel steels, whether of the SA-533B/SA-508 Class 3 or SA-508 Class 2 type, have high manganese-sulphur ratios, the former typically contains 1.6-1.8 times the manganese of the latter and therefore for a fixed sulphur impurity content it may be anticipated to be less susceptible to this mode of cracking.
(b) Reheat cracking

Of the embrittlement and cracking phenomena resulting from the fabrication of nuclear pressure vessels, reheat cracking is considered the most difficult to solve. Furthermore, in spite of extensive research of this phenomenon in PWR pressure vessel [9, 30, 33-49] and other steels, a complete understanding of all controlling parameters has not yet been achieved. However, there is general agreement in qualitative terms of the mechanism involved and susceptibility of individual classes of steels; for example SA-533B Class 1 and SA-508 Class 3 although not immune are less susceptible to this mode of cracking than SA-508 Class 2.

Reheat cracks vary in size from a single grain (\(\sim 20\ \mu m\)) to a significant fraction of the weldment (10-1m) [34, 35]. Cracking occurs preferentially in regions of alloying and impurity element segregation. Microcracks are restricted to the coarse-grained unrefined regions of the HAZ whereas macrocracks link coarse-grained regions interconnected by partially refined regions of HAZ microstructure. The positions and types of cracking found in the extensive investigations by Kussmaul and co-workers [30, 32-34] are summarised in Fig. 30.

Reheat cracking is a high temperature grain boundary fracture phenomenon occurring at temperatures below about 700°C. The occurrence is more likely in materials with coarse austenite grain
size and relatively high levels of the impurities together with a high hardness before reheating or stress relieving and a resistance to softening at elevated temperatures. Thus the extent of cracking in heat affected zone is reduced by the presence of aluminium nitride which induces a fine grain size and prevents grain growth by minimising the heat input into the weld to reduce the size of the HAZ, and by restricting levels of those elements which form fine dispersions of stable carbides and hence are responsible for resistance to softening during the tempering heat treatment. For the latter purpose it is important to achieve very low levels of vanadium, zirconium and niobium.

Reheat cracking results when the relaxation strain exceeds the local creep ductility of the material. It occurs during postweld heat treatment (PWHT) when the welded structure is heated slowly from room temperature or the post-welding temperature (up to 300 °C) to a temperature between 550 and 650 °C, held at this temperature for several hours and slowly cooled to minimise further residual stresses. Cracking can occur during heat-up or holding when the instantaneous conditions of residual stress, hardness, accumulated strain, microstructure and interfacial segregation of impurity elements are consistent with the requirements of a particular failure mechanism.

The creep processes occurring during relief of residual stresses are sensitive to small changes in alloy and impurity element composition and to microstructure [31, 33, 34, 41, 44, 46,
Whilst there is general agreement that SA-533B Class 1 and SA-508 Class 3 are less susceptible to reheat cracking than SA-508 Class 2 and that impurity elements are deleterious, there is only a broad quantitative consensus on the relative effects of different impurities. From studies on simulated HAZs in experimental steels containing single impurities, Brear and King [47] recommend that individual elements should not exceed the limits given in Table 7 and the combined impurity element (wt.%) should be

\[ P + 0.81 \text{ As} + 1.18 \text{ Sn} + 1.49 \text{ Sb} + 0.12 \text{ Cu} + 0.195 \text{ S} < 0.03 \]

in order to avoid reheat cracking. Specimens such as those used in this study, with simulated coarse-grained microstructure across the whole cross-section, in general, yield pessimistic results in comparison with those obtained from actual weldments. The results also refer to specific test conditions and it is not clear how they relate quantitatively to the occurrence of reheat cracking in service. Kussmaul et al. report an increasing tendency for cracking with increasing contents of phosphorus, sulphur, copper, arsenic, aluminium, nitrogen, molybdenum and cobalt [33]. They also propose a 'threshold value' criterion such that reheat cracking occurs if two or more elements exceed the limits given in Table 6. More recently [38], examination of reheat cracking in commercial casts of SA-533B Class 1 and SA-508 Class 2 steels has indicated that increasing the chromium content is deleterious. Similarly the importance of chromium and other strong carbide-forming elements such as molybdenum, vanadium, titanium and niobium in prompting
reheat cracking in other low alloy steels has been recognised [50, 51].

(c) Hydrogen cracking

Hydrogen cracking is a potential source for defects associated with the HAZ of structural weld. This is a brittle cracking mechanism occurring below about 200°C. The phenomenon is associated particularly with high strength steels in the as-welded condition where hydrogen has been introduced during welding and high levels of stress remain [53]. Although no failures or large defects in PWR plant have been reported to be caused by hydrogen cracking, recent work has demonstrated that regions of alloy segregation in PWR pressure vessels and other steels are more susceptible to hydrogen cracking than the matrix [49, 54-56]. Liquation cracks, also occurring in segregated material, are ideally suited to act as stress concentrators for subsequent hydrogen cracking.

The critical concentration of hydrogen below which crack initiation will not occur is not known, although values as low as 1.3 ppm have been suggested [56]. A low hydrogen content in plates and forgings is achieved by vacuum degassing prior to casting (2-3 ppm) together with one or more heat treatments during fabrication (1 ppm). On welding, the local hydrogen concentration of the weldment will increase. With good welding practice, as specified in procedures for fabrication of nuclear vessels, a concentration of about 5 ppm
can be expected. Immediately after welding the HAZ is susceptible
to hydrogen cracking and therefore should be maintained at an
elevated temperature until sufficient hydrogen has diffused
away. Tests have shown that no cracking occurs in SA-508 Class 3
weldments provided that either a 200 °C preheat and post-heat
temperature is employed or a lower preheat temperature combined with
a post-heating cycle after welding is used [37]. Further reductions
in hydrogen concentration will occur during subsequent stress relief
annealing.

4.2 Cladding

(1) Material and welding procedure [2]

The inner surface of the vessel is clad with a corrosion resistant
layer by continuously melting cladding material onto the vessel
surface to produce a fusion weld. The method is mechanised for the
single curvature surfaces in order to provide a layer of constant
thickness. Certain regions of double curvature, however, must be
clad manually. Two types of feed material are used; a type 309/308
austenitic stainless steel which is used for cladding the ferritic
steel of the main pressure vessel and Inconel overlay which is used
on penetrations, core support pads and on the faces of the nozzles.
Detailed of established methods of clad welding, electrode and flux
compositions and operating conditions are given in the review of
overlay welding by Gooch [57]. A recent process has been developed
reportedly capable of depositing high quality austenitic cladding up to 300 mm with [58].

(2) Defects associated with cladding [2]

There are several differences between structural welding and cladding which are relevant to the mechanisms of formation of welding defects. In the case of cladding the deposit is mainly austenitic rather than ferritic, and therefore possesses a higher coefficient of thermal expansion and greater high temperature strength, both factors influencing residual stress formation and stress relaxation behaviour. Also heat input is high during cladding thus promoting residual stresses and large heat affected zones. However, with the exception of the inside of nozzles the cladding process is a low-restraint weld configuration and therefore less severe with respect to long range residual stress formation. Hydrogen has a higher solubility but lower diffusivity in austenitic than in ferritic steels. Consequently, the austenitic cladding can retain hydrogen which may subsequently pass into the base material. Most reported defects associated with the cladding of PWR pressure vessels are related to the HAZ in the ferritic steel below the cladding. However, defects can occur both in the cladding itself and along the cladding/ferrite interface. Gooch reports three instances of cracking in the cladding during fabrication due to failure to achieve the desirable composition and microstructure [57].
(a) Underclad reheat cracking [2]

A potential problem associated with cladding of reactor pressure vessels is the formation of underclad cracks. The first report of defects in the HAZ beneath austenitic cladding in nuclear plant was in 1970 [59]. The Welding Research Council undertook a comprehensive review of the phenomenon and Vinckier and Pense reported this work in 1974 [9]. Cracks were found exclusively along prior austenite grain boundaries with sizes varying from a minimum of 0.2 mm in depth and length to a maximum of 10 mm length and 3 mm depth. In more recent reviews [31, 44] the maximum depth is reported as 4 mm. The cracks exist in a region which is somewhat difficult to examine by conventional ultrasonic testing techniques because of the proximity of the cladding and the surface. Cracks have been revealed by stripping the cladding and using surface crack detection methods. Metallographic examination showed that cracks were in the coarse-grained region of the HAZ which had been fully austenitised by the first cladding deposit and then heated to just below the austenitisation temperature, i.e. 600-700°C, by the subsequent adjacent cladding deposit. The susceptible region is under the highest residual tensile stress immediately after welding. The direction of cracking was usually between 45 and 90° to the direction of welding. Fig. 31 illustrates the position of the cracks.
Vinckier and Pense concluded that pressure vessel steels manufactured to the different specifications have different susceptibilities to underclad cracking. Out of 96 reports showing 26 cases of underclad cracking, 25 were in SA-508 Class 2, one in SA-508 Class 3 and no cases were reported for SA-533B Class 1. High heat input during cladding resulted in underclad cracking in SA-508 Class 2 but not in the other steels. This pattern of behaviour was confirmed by other reviews in the period 1974-1978 [31, 44, 60, 61]. The cracking reported was all attributed to reheat cracking occasionally augmented by liquation cracking.

Figs. 32 and 33 show reheat cracking susceptible areas and methods to avoid reheat cracking by refining heat affected coarse grain zones [62].

(b) Underclad hydrogen cracking [54]

Hydrogen cracking is one of the most important problems for the integrity of steel structures and many studies on this subject have been extensively carried out until now. The dominant factors for the hydrogen cracking are summarized as follows:

- existence of diffusible hydrogen
- existence of tensile stress or strain
- existence of hydrogen embrittlement susceptible microstructure
- low temperature conditions (below 150°C)
The hydrogen crack initiates when all four factors are simultaneously satisfied. In the case of welding between low alloy ferritic metals, the weld joint becomes quite susceptible to the hydrogen cracking and almost all the studies have been conducted on this subject. On the other hand, weld joints of ferritic metal and austenitic weld metal are considered to be resistant to hydrogen cracking because of the high hydrogen solubility, low hydrogen diffusion rate in the weld metal and sufficient capacity of relaxation of welding induced strain. Nevertheless, the hydrogen cracks in the HAZ under austenitic stainless steel overlay were recently reported on the tube sheet forging of a steam generator in a light water reactor [55, 63]. The references pointed out that the cracks mainly initiate in the zones of segregation.

The existence of susceptible microstructure to hydrogen embrittlement is also an important factor influencing the initiation of hydrogen cracks.

Four vital conditions to initiate the hydrogen cracking have been examined independently for the heat affected zone under the austenitic stainless steel overlay. From the results, it is concluded that:

- 2 to 4 ppm hydrogen diffuses from the austenitic weld metal into the base metal by the $\gamma$-transformation of the base metal due to the welding
- heat affected zone is quite hydrogen embrittlement susceptible and the embrittlement is
especially noticeable at room temperature. The remarkable hydrogen embrittlement occurs at hydrogen content of 1.5 ppm and higher.

- segregation, which is difficult to avoid in large forgings at the present time, is the most susceptible to hydrogen embrittlement.
- maximum magnitude of residual stress amount to 500 MPa and the magnitude is sufficient to initiate the hydrogen cracking.

From the above facts, it is quantitatively proved that hydrogen cracking occurs even in the weld joint between the austenitic weld metal and ferritic metal.

The preventive measures against hydrogen cracking are summarized as follows. On the assumption that the materials and design of component are not changed for this purpose, the existence of hydrogen embrittlement susceptible microstructure and restraint condition of weld joint cannot be avoided. On the other hand, the large decreases in residual stresses under the weld overlay cannot be expected by the conventional soaking treatments. Hence, the countermeasures against the hydrogen cracking becomes as follows.

- elimination of hydrogen in the heat affected zone
- to avoid the low temperature conditions under the hydrogen absorbed conditions of heat affected zone

Therefore, it is recommended that the preheating should be maintained at least during 1st and 2nd layer welding and until post
weld heat treatment or soaking treatment. The soaking treatment at about 250°C is undoubtedly effective to avoid the hydrogen cracking. By these countermeasures, hydrogen cracking in the HAZ of heavy forging with stainless steel overlay clad can be completely avoided.

4.3 Mechanical Properties of Weld Metal and Heat Affected Zone (HAZ)[2]

Tensile and Charpy V-notch impact properties for the European weld metals are comparable with the base metal data. It would appear that adequate low temperature notch toughness and comparable tensile ductility can be achieved in weld metals even when the weld metal yield strength is approximately 10-20% higher than the nominal mean value of 470 MPa for SA-533 Grade B Class 1/SA-508 Class 3 base materials. Data for base metals, weld metals and HAZs which were tested in the EPRI programmes [20] are collated in Tables 8, 4 and 9 to 12. Data from Japan which provide some indication of the improvement in upper shelf notch toughness and also in the ductile-brittle transition temperature that can be obtained using narrow gap welding processes [64, 65].

From the results examined for both weld metals and HAZs, which are rather limited in some cases, it can be concluded that it is possible to achieve mechanical properties in weld metals and heat affected zones
that are at least as good as those of plates and forgings. It is believed that the weld metal rather than the HAZ will govern acceptance. By correct selection of welding consumables/parameters, weld metal properties can be achieved which are well in excess of the minimum property values specified by the ASME Codes. The upper shelf notch toughness of weld metals should also be reasonably high, but cannot be guaranteed to match the high notch toughness values of the very high quality steel now available from a number of sources. Recent developments in the production of plates and forgings, and to a lesser extent welds, have resulted in an improvement in properties, beyond the minimum values required by the ASME Codes, and therefore beyond the material properties upon which previous PWR vessel production has been based.

5 Advanced Design due to Optimized Material

The design of the RPV for the light water reactor (LWR) tends to minimize the weld seams, which reduces the period of in-service inspection (ISI) together with easier performance of ISI. This tendency required the more integrated and larger parts for nuclear steam supply system (NSSS) components.

It was said that weld seams can be reduced to 70 percent for boiling water reactor pressure vessels (BWRPVs) and 25 percent for pressurized water reactor pressure vessels (PWRPVs) when compared to
conventional designs, by the use of the large forgings and plates available at present in the world [66]. Typical layouts for these designs of nuclear pressure vessels are as shown in Fig.34 for the PWR and Fig.35 for BWR. The seamless forged shells for the BWRPV and PWRPV, as well as the vessel flange integral with nozzle belt shell in PWRPV, are significantly advantageous from the standpoint of design, fabrication and inspection.

The numerous seamless forged shells in BWRPV and PWRPV have already been realized as shown in Figs.36 and 37 [67], respectively, using the advanced technology for the manufacture of heavy steel forgings.

One-piece forged shell flanges weighing 165 tons for KWU type 4-loop PWRPV made from 400 ton ingots shown in Fig.17 have been successfully developed [13]. Furthermore, on the basis of much manufacturing experience of one-piece shell flange for KWU, monoblock vessel flange of WEC type 157" (3988 mm) PWRPV, combined vessel flange with nozzle belt shell and on which the set-on type main coolant nozzles are welded as shown in Fig.18, was manufactured successfully using a 500 ton ingot [68-71].
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<table>
<thead>
<tr>
<th>Material Specification</th>
<th>Chemical composition (wt., %)</th>
<th>Mechanical properties</th>
</tr>
</thead>
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<tr>
<td></td>
<td>C</td>
<td>Si</td>
</tr>
<tr>
<td>USA (ASME)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plate</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SA533B, Cl.1</td>
<td>≤ 0.25</td>
<td>0.15/1.15</td>
</tr>
<tr>
<td>Forging</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SA508, Cl.2</td>
<td>≤ 0.27</td>
<td>0.15/0.70</td>
</tr>
<tr>
<td>SA508, Cl.3</td>
<td>≤ 0.25</td>
<td>0.15/1.20</td>
</tr>
<tr>
<td>Germany (KTA)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plate &amp; Forging</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20MnMoNi55 (W.Nr.1.6311)</td>
<td>0.17/0.23</td>
<td>0.15/0.30</td>
</tr>
</tbody>
</table>

Notes:
2) 350°C for KTA
3) ASME SA533, P ≤ 0.012
4) ASME SA508, Supplementary Requirements P ≤ 0.012
5) KTA 3201.1, 20MnMoNi55
P: to be aimed at lower value
Cu ≤ 0.010, S ≤ 0.015, V ≤ 0.05
Cu ≤ 0.10
Cu ≤ 0.010

Appendix X1.1

S9.1(a)
### TABLE 2
Minimum Mechanical Properties Specified in the ASME Codes (2)

<table>
<thead>
<tr>
<th>Property</th>
<th>A533B 1 Plates 20°C</th>
<th>A533B 1 Plates 350°C</th>
<th>A508 3 Forgings 20°C</th>
<th>A508 3 Forgings 350°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Yield stress (MPa)</td>
<td>345</td>
<td>285(*)</td>
<td>345</td>
<td>285</td>
</tr>
<tr>
<td>Ultimate tensile stress (MPa)</td>
<td>552</td>
<td>527</td>
<td>550</td>
<td>-</td>
</tr>
<tr>
<td>El (in 50 mm) %</td>
<td>18</td>
<td>-</td>
<td>18</td>
<td>-</td>
</tr>
<tr>
<td>R of A %</td>
<td>-</td>
<td>-</td>
<td>38</td>
<td>-</td>
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</tbody>
</table>

**Charpy Impact**

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy (J)</td>
<td>68 J at RT&lt;sub&gt;NDT&lt;/sub&gt; + 33°C</td>
</tr>
<tr>
<td>Lateral expansion (mm)</td>
<td>0.89 mm at RT&lt;sub&gt;NDT&lt;/sub&gt; + 33°C</td>
</tr>
<tr>
<td>Minimum ave. value (+) of three specimens</td>
<td>×</td>
</tr>
<tr>
<td>Minimum value of one specimen</td>
<td>×</td>
</tr>
<tr>
<td></td>
<td>41 J at 4.4°C</td>
</tr>
<tr>
<td></td>
<td>34 J at 4.4°C</td>
</tr>
</tbody>
</table>

(**) non-mandatory

(+) not more than one specimen from a set may fall below this value

× to be specified by purchaser
### TABLE 3
Chemical Composition and Mechanical Property Values of Weld Metal in Simulated Heat-Treatment Condition

<table>
<thead>
<tr>
<th>Weld metal type</th>
<th>Chemical composition (wt.% min-max value)</th>
<th>Tensile Strength</th>
<th>Charpy Impact Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cr</td>
<td>Mo</td>
<td>Ni</td>
</tr>
<tr>
<td>E-1</td>
<td>.05</td>
<td>.40</td>
<td>1.60</td>
</tr>
<tr>
<td>E-2</td>
<td>.07</td>
<td>.42</td>
<td>1.50</td>
</tr>
<tr>
<td>UP-1</td>
<td>.05</td>
<td>.21</td>
<td>1.30</td>
</tr>
<tr>
<td>E-3</td>
<td>.04</td>
<td>.32</td>
<td>1.28</td>
</tr>
<tr>
<td>UP-2</td>
<td>.05</td>
<td>.16</td>
<td>1.70</td>
</tr>
<tr>
<td>E-4</td>
<td>.02</td>
<td>.21</td>
<td>1.09</td>
</tr>
<tr>
<td>UP-3</td>
<td>.05</td>
<td>.42</td>
<td>1.68</td>
</tr>
<tr>
<td>E-5</td>
<td>.09</td>
<td>.17</td>
<td>1.35</td>
</tr>
<tr>
<td>UP-4</td>
<td>.05</td>
<td>.14</td>
<td>1.35</td>
</tr>
<tr>
<td>UP-5</td>
<td>.07</td>
<td>.23</td>
<td>1.91</td>
</tr>
</tbody>
</table>

Abbreviations: X = mean value; s = standard deviation; n = number of specimens
<table>
<thead>
<tr>
<th>Heat</th>
<th>C</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Si</th>
<th>Ni</th>
<th>Cr</th>
<th>Mo</th>
<th>Y</th>
<th>Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>MMA Welds</td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>P</td>
<td>0.100</td>
<td>1.000</td>
<td>0.005</td>
<td>0.010</td>
<td>0.390</td>
<td>0.880</td>
<td>0.010</td>
<td>0.290</td>
<td>0.004</td>
<td>0.020</td>
</tr>
<tr>
<td>Q</td>
<td>0.100</td>
<td>1.110</td>
<td>0.007</td>
<td>0.010</td>
<td>0.400</td>
<td>1.060</td>
<td>0.010</td>
<td>0.340</td>
<td>0.006</td>
<td>0.020</td>
</tr>
<tr>
<td>R</td>
<td>0.100</td>
<td>1.100</td>
<td>0.006</td>
<td>0.010</td>
<td>0.400</td>
<td>1.000</td>
<td>0.010</td>
<td>0.330</td>
<td>0.005</td>
<td>0.020</td>
</tr>
<tr>
<td>S</td>
<td>0.09</td>
<td>1.03</td>
<td>0.005</td>
<td>0.01</td>
<td>0.39</td>
<td>0.95</td>
<td>0.01</td>
<td>0.32</td>
<td>0.006</td>
<td>0.020</td>
</tr>
<tr>
<td>T</td>
<td>0.04</td>
<td>1.02</td>
<td>0.017</td>
<td>0.022</td>
<td>0.49</td>
<td>0.95</td>
<td>0.01</td>
<td>0.53</td>
<td>0.014</td>
<td>0.020</td>
</tr>
<tr>
<td>U</td>
<td>0.050</td>
<td>0.150</td>
<td>0.016</td>
<td>0.024</td>
<td>0.520</td>
<td>0.940</td>
<td>0.010</td>
<td>0.540</td>
<td>0.014</td>
<td>0.030</td>
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<tr>
<td>S/A Welds</td>
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<tr>
<td>Y</td>
<td>0.150</td>
<td>1.38</td>
<td>0.008</td>
<td>0.009</td>
<td>0.16</td>
<td>0.13</td>
<td>0.04</td>
<td>0.60</td>
<td>0.007</td>
<td>0.040</td>
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<td>H</td>
<td>0.14</td>
<td>1.19</td>
<td>0.01</td>
<td>0.009</td>
<td>0.19</td>
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<td>0.09</td>
<td>0.54</td>
<td>0.005</td>
<td>0.120</td>
</tr>
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<td>X</td>
<td>0.150</td>
<td>1.280</td>
<td>0.011</td>
<td>0.010</td>
<td>0.200</td>
<td>0.190</td>
<td>0.080</td>
<td>0.540</td>
<td>0.005</td>
<td>0.110</td>
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<td>0.13</td>
<td>1.25</td>
<td>0.011</td>
<td>0.010</td>
<td>0.18</td>
<td>0.10</td>
<td>0.09</td>
<td>0.53</td>
<td>0.005</td>
<td>0.200</td>
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<td>Base</td>
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<tr>
<td>B</td>
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<td>1.41</td>
<td>0.008</td>
<td>0.014</td>
<td>0.260</td>
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<td>0.11</td>
<td>0.49</td>
<td>0.003</td>
<td>0.120</td>
</tr>
<tr>
<td>L</td>
<td>0.21</td>
<td>1.34</td>
<td>0.012</td>
<td>0.019</td>
<td>0.230</td>
<td>0.44</td>
<td>0.07</td>
<td>0.53</td>
<td>0.004</td>
<td>0.100</td>
</tr>
<tr>
<td>N</td>
<td>0.24</td>
<td>1.30</td>
<td>0.009</td>
<td>0.013</td>
<td>0.240</td>
<td>0.46</td>
<td>0.11</td>
<td>0.53</td>
<td>0.002</td>
<td>0.080</td>
</tr>
</tbody>
</table>
TABLE 5

Chemical composition of filler metal as specified, as received and as deposited, together with that of the base metal (SA533B) (21)

<table>
<thead>
<tr>
<th>Chemical composition, wt.%</th>
<th>C</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Si</th>
<th>Ni</th>
<th>Cr</th>
<th>Mo</th>
<th>Cu</th>
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<tr>
<td>specification(^a)</td>
<td>.15</td>
<td>1.80</td>
<td>LAP</td>
<td>LAP</td>
<td>0.10</td>
<td>.55</td>
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<td>.45</td>
<td>LAP</td>
<td>.02</td>
<td>.05</td>
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<td></td>
<td>.20</td>
<td>2.10</td>
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<td>0.010</td>
<td>(max)</td>
<td>(max)</td>
<td>(max)</td>
<td>(max)</td>
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<td>(max)</td>
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<td>(max)</td>
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<td><strong>Filler metal:</strong></td>
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<td>.008</td>
<td>.014</td>
<td>.10</td>
<td>.71</td>
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<td>.02</td>
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<td>Lukens</td>
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<td><strong>Weld deposit(^b):</strong></td>
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<td>.015</td>
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<td>.02</td>
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<tr>
<td><strong>Base metal:</strong></td>
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<td>1.22</td>
<td>.008</td>
<td>.008</td>
<td>.19</td>
<td>.58</td>
<td>.06</td>
<td>.50</td>
<td>.03</td>
<td>.02</td>
<td>.015</td>
<td>&lt;.03</td>
<td>&lt;.02</td>
<td>&lt;.01</td>
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</tbody>
</table>

\(^a\) LAP = low as possible

\(^b\) Carbon content of weld deposit to be 0.12-0.18
### TABLE 6

Compositional Upper Limit for Elements which Promote HAZ Cracking

(wt.%) (After Kussmaul) (33)

<table>
<thead>
<tr>
<th>Trace and impurity elements</th>
<th>Carbide Formers</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>Sn</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Liquation Cracking</td>
<td>0.13</td>
</tr>
<tr>
<td>Stress Relief Cracking</td>
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</table>

### TABLE 7

Compositional Upper Limit for Elements which Promote Reheat Cracking

(wt.%) (After Brear & King) (47)

<table>
<thead>
<tr>
<th>P</th>
<th>As</th>
<th>Sn</th>
<th>Sb</th>
<th>Cu</th>
<th>S</th>
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<tbody>
<tr>
<td>0.012</td>
<td>0.015</td>
<td>0.010</td>
<td>0.008</td>
<td>0.10</td>
<td>0.015</td>
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<td>EPRI Heat</td>
<td>Type of Weld</td>
<td>Thickness (mm)</td>
<td>Base Material</td>
<td>Electrode</td>
<td>Flux</td>
</tr>
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<td>-----------</td>
<td>--------------</td>
<td>----------------</td>
<td>---------------</td>
<td>-----------</td>
<td>------</td>
</tr>
<tr>
<td>U (L)*</td>
<td>MMA</td>
<td>0.127</td>
<td>A533B Cl 1</td>
<td>E8018-MN</td>
<td>-</td>
</tr>
<tr>
<td>T (N)</td>
<td></td>
<td></td>
<td></td>
<td>E8018-NM</td>
<td>-</td>
</tr>
<tr>
<td>P</td>
<td></td>
<td>0.152</td>
<td></td>
<td>E8018-C3</td>
<td>-</td>
</tr>
<tr>
<td>Q</td>
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<td></td>
<td></td>
<td>-</td>
</tr>
<tr>
<td>R</td>
<td></td>
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<td></td>
<td></td>
<td>-</td>
</tr>
<tr>
<td>S</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>-</td>
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<tr>
<td>(B)</td>
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<td>-</td>
</tr>
<tr>
<td>G (A)</td>
<td>MMA</td>
<td></td>
<td>A508 - 2</td>
<td>E8015-C3</td>
<td>-</td>
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<tr>
<td>I (C)</td>
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<td></td>
<td></td>
<td>E8018-C3</td>
<td>-</td>
</tr>
<tr>
<td>K (E)</td>
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<td>H (B)</td>
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<tr>
<td>J (D)</td>
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<tr>
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<tr>
<td>P (E)</td>
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<td>A508 - 2</td>
<td>Mn Mo Ni</td>
<td>Linde 80</td>
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<td>O (D)</td>
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<td>L (A)</td>
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<td>-</td>
</tr>
<tr>
<td>N (C)</td>
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<td>M (B)</td>
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<td>-</td>
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* Reference Code in parenthesis identifies heat code for HAZ tests.
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<th>Tensile Property</th>
<th>Material</th>
<th>AS3301 base (T)</th>
<th>MMA weld in AS3301 (L)</th>
<th>M/M weld in AS3301 (T)</th>
<th>S/A weld in AS3301 (L)</th>
<th>S/A(WZ in AS3301 (L)</th>
</tr>
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<tr>
<td>C_{y} (MPa)</td>
<td>24</td>
<td>5 440 33.6 5</td>
<td>526 69.5 5 507 47</td>
<td>200 402 26.7 443 20.6 434 30.1</td>
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</tr>
<tr>
<td>$\sigma_{u}$ (MPa)</td>
<td>24</td>
<td>5 595 23.5 5</td>
<td>611 52.9 5 619 36.2</td>
<td>200 585 21.2 567 31.1 560 32.5</td>
<td></td>
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</tr>
<tr>
<td>El (%)</td>
<td>24</td>
<td>5 27.4 1.0 5</td>
<td>27.0 1.0 5 25.3 1.7</td>
<td>200 24.3 0.5 22.9 1.6 21.0 1.7</td>
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<td></td>
</tr>
<tr>
<td>R of A (%)</td>
<td>24</td>
<td>5 440 33.6 5</td>
<td>526 69.5 5 507 47</td>
<td>200 402 26.7 443 20.6 434 30.1</td>
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<td></td>
</tr>
<tr>
<td>UTS (MPa)</td>
<td>24</td>
<td>5 595 23.5 5</td>
<td>611 52.9 5 619 36.2</td>
<td>200 585 21.2 567 31.1 560 32.5</td>
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<tr>
<td>El (%)</td>
<td>24</td>
<td>5 27.4 1.0 5</td>
<td>27.0 1.0 5 25.3 1.7</td>
<td>200 24.3 0.5 22.9 1.6 21.0 1.7</td>
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<tr>
<td>R of A (%)</td>
<td>24</td>
<td>5 440 33.6 5</td>
<td>526 69.5 5 507 47</td>
<td>200 402 26.7 443 20.6 434 30.1</td>
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<tr>
<td>UTS (MPa)</td>
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<td>611 52.9 5 619 36.2</td>
<td>200 585 21.2 567 31.1 560 32.5</td>
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<tr>
<td>El (%)</td>
<td>24</td>
<td>5 27.4 1.0 5</td>
<td>27.0 1.0 5 25.3 1.7</td>
<td>200 24.3 0.5 22.9 1.6 21.0 1.7</td>
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</tr>
<tr>
<td>R of A (%)</td>
<td>24</td>
<td>5 440 33.6 5</td>
<td>526 69.5 5 507 47</td>
<td>200 402 26.7 443 20.6 434 30.1</td>
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</table>
### TABLE 10
Impact Properties of EPRI Base Metals and Welds (20)

<table>
<thead>
<tr>
<th>Material</th>
<th>A533B1</th>
<th>MMA weld metal</th>
<th>S/A weld metal</th>
<th>MMA HAZ</th>
<th>SA/HAZ</th>
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</thead>
<tbody>
<tr>
<td>Transition Temp.</td>
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</tr>
<tr>
<td>L</td>
<td>13</td>
<td>18</td>
<td>6 -16</td>
<td>11.8</td>
<td>3 -32</td>
</tr>
<tr>
<td>T</td>
<td>13</td>
<td>17</td>
<td>5 -20</td>
<td>10.7</td>
<td>-</td>
</tr>
<tr>
<td>Upper Shelf Energy</td>
<td>L</td>
<td>13 202</td>
<td>27</td>
<td>5 248</td>
<td>62.3</td>
</tr>
<tr>
<td>Energy (J)</td>
<td>T</td>
<td>13 142</td>
<td>27.6</td>
<td>5 224</td>
<td>53.7</td>
</tr>
<tr>
<td>NDTT (°C)</td>
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<td></td>
<td></td>
<td></td>
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<tr>
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<td>13</td>
<td>-24</td>
<td>9.5</td>
<td>5 -53</td>
<td>6.6</td>
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<tr>
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<td></td>
<td>4 -57</td>
<td>0</td>
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<tr>
<td>RT&lt;sub&gt;NDT&lt;/sub&gt; (°C)</td>
<td>13</td>
<td>-11</td>
<td>10.0</td>
<td>6 -48</td>
<td>14.5</td>
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<table>
<thead>
<tr>
<th>Material</th>
<th>A508-2</th>
<th>MMA weld metal</th>
<th>S/A weld metal</th>
<th>MMA HAZ</th>
<th>S/A HAZ</th>
</tr>
</thead>
<tbody>
<tr>
<td>To (°C)</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>L</td>
<td>5</td>
<td>7.6</td>
<td>3.9</td>
<td>5 -33</td>
<td>3.9</td>
</tr>
<tr>
<td>T</td>
<td>5</td>
<td>9</td>
<td>6.2</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Upper Shelf Energy</td>
<td>L</td>
<td>5 219</td>
<td>34.7</td>
<td>5 189</td>
<td>10.1</td>
</tr>
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<td>Energy (J)</td>
<td>T</td>
<td>5 194</td>
<td>35.7</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>NDTT (°C)</td>
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<td></td>
<td></td>
</tr>
<tr>
<td></td>
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<td>5</td>
<td>5 -61</td>
<td>12.6</td>
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<tr>
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<td>-3.6</td>
<td>5</td>
<td>5 -58</td>
<td>12.5</td>
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(Notation: See Table 3.18)
### TABLE II
Charpy Impact and RT\text{\textsubscript{NDT}} Data for Welds in A533B Cl1

<table>
<thead>
<tr>
<th>Plate Thickness</th>
<th>Weld Prep.</th>
<th>Electrode /Flux</th>
<th>Heat Input (MJ/m)</th>
<th>Heat Code</th>
<th>RT\text{\textsubscript{NDT}} (°C)</th>
<th>\text{To} (°C)</th>
<th>\text{CV US} (J)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Manual Metal Arc Weld Metal</td>
<td></td>
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</tr>
<tr>
<td>0.127</td>
<td>Y groove</td>
<td>E8018-MN</td>
<td>3.5</td>
<td>U*</td>
<td>-40</td>
<td>-16(L)</td>
<td>138(L)</td>
</tr>
<tr>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td>-43(L)</td>
<td>164*(L)</td>
</tr>
<tr>
<td>0.127</td>
<td>Y groove (R)</td>
<td>E8018-NM</td>
<td>3.5</td>
<td>L*</td>
<td>-46*</td>
<td>-1(T)</td>
<td>146(T)</td>
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<tr>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>-30(L)</td>
<td>168*(L)</td>
</tr>
<tr>
<td>0.152</td>
<td>Straight wall groove</td>
<td>E8018-C3</td>
<td>4.5</td>
<td>T</td>
<td>-23</td>
<td>+5(L)</td>
<td>275(L)</td>
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<td>-24(T)</td>
<td>216(T)</td>
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<td>0.152</td>
<td>Straight wall</td>
<td>E8018-C3</td>
<td>4.5</td>
<td>N*</td>
<td>-40*</td>
<td>-17(L)</td>
<td>289(L)</td>
</tr>
<tr>
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<td>212(T)</td>
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<td>Straight wall</td>
<td>E8018-C3</td>
<td>4.5</td>
<td>P</td>
<td>-57</td>
<td>-14(L)</td>
<td>274(L)</td>
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<td>-28(T)</td>
<td>263(T)</td>
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<tr>
<td>0.178</td>
<td>Straight wall</td>
<td>E8018-C3</td>
<td>4.5</td>
<td>Q</td>
<td>-62</td>
<td>-30(L)</td>
<td>266(L)</td>
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<td>-24(T)</td>
<td>285(T)</td>
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<td>Double U groove</td>
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<td>B*</td>
<td>-40*</td>
<td>-23(L)</td>
<td>205*(L)</td>
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<td>Submerged Arc Weld Metal</td>
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<tr>
<td>0.178</td>
<td>Straight wall</td>
<td>MIL.8-4/</td>
<td>2.8</td>
<td>Y</td>
<td>-57</td>
<td>-29(L)</td>
<td>206(L)</td>
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<td>-46(T)</td>
<td>218(T)</td>
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<tr>
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<td>Double U groove</td>
<td>MIL.8-4/</td>
<td>4.0</td>
<td>B*</td>
<td>-40*</td>
<td>-14(L)</td>
<td>181*(L)</td>
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<tr>
<td>0.300</td>
<td>Double U (F)</td>
<td>MIL.8-4/</td>
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<td>W</td>
<td>-57</td>
<td>-25(L)</td>
<td>163(L)</td>
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<td>-34(T)</td>
<td>178(T)</td>
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<td>Double U (F)</td>
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<td>-24(L)</td>
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<td>-18(L)</td>
<td>160(L)</td>
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<td>Linde 0091</td>
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<td></td>
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<td>-22(T)</td>
<td>167(T)</td>
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* - Heat affected zone  
(F) - Full cylindrical restraint  
(R) - Extensive repairs during welding  
\text{To} - Transition temperature defined by Tanh curve fit
<table>
<thead>
<tr>
<th>Electrode / Flux</th>
<th>Heat Input (MJ/m)</th>
<th>Weld Metal</th>
<th>HAZ</th>
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</tr>
<tr>
<td></td>
<td>°C</td>
<td>°C</td>
<td>°C</td>
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<td>Manual Metal Arc</td>
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<td>I</td>
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<td>K</td>
<td>-62</td>
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<td>-40</td>
</tr>
<tr>
<td></td>
<td>2.3</td>
<td>J</td>
<td>-51</td>
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<td>Submerged Arc</td>
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<td>O</td>
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To = Transition temperature defined by Tanh curve fit
Chemical composition, wt. %

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<tr>
<td>2</td>
<td>0.20</td>
<td>0.21</td>
<td>1.46</td>
<td>0.010</td>
<td>0.007</td>
<td>0.78</td>
<td>0.03</td>
<td>0.10</td>
<td>0.50</td>
</tr>
</tbody>
</table>

Austenitizing conditions: 890°C for 15 min.

Fig. 1 Continuous cooling transformation diagram for 20 MnMoNi 55 steel
<table>
<thead>
<tr>
<th>Year</th>
<th>'56</th>
<th>'58</th>
<th>'60</th>
<th>'62</th>
<th>'64</th>
<th>'66</th>
<th>'68</th>
<th>'70</th>
<th>'72</th>
<th>'74</th>
<th>'76</th>
<th>'78</th>
<th>'80</th>
<th>'82</th>
<th>'84</th>
<th>'86</th>
<th>'88</th>
<th>'90</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melting</td>
<td>Open hearth (acidic &amp; basic) &amp; electric arc furnace (10ton, 25ton)</td>
<td>Open hearth (basic) &amp; electric arc furnace (10ton, 25ton, 100ton)</td>
<td></td>
<td></td>
<td></td>
<td>Electric arc furnace (25ton, 100ton, 120ton)</td>
<td>Holding furnace</td>
<td>Ladle refining furnace</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pouring</td>
<td>Air pouring</td>
<td>Vacuum pouring (mechanical pump)</td>
<td></td>
<td></td>
<td></td>
<td>Vacuum pouring (steam ejector)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ingot size</td>
<td>140ton</td>
<td>220ton</td>
<td>250ton</td>
<td>400ton</td>
<td>500ton</td>
<td>570ton</td>
<td>600ton</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Fig. 2 Transition of steel and ingot making methods, and of size of ingots in these 35 years in JSW
FIG. 3 PRINCIPAL FUNCTIONS OF LADLE REFINING FURNACE
Fig. 4 Production Sequence of 600 Ton Ingot
Chemical Compositions (%)

<table>
<thead>
<tr>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Ni</th>
<th>Cr</th>
<th>Mo</th>
<th>V</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.31</td>
<td>0.35</td>
<td>0.55</td>
<td>0.018</td>
<td>0.019</td>
<td>1.3</td>
<td>1.09</td>
<td>0.042</td>
<td>0.07</td>
</tr>
</tbody>
</table>

Acid Open Hearth Furnace Air Cast 75T Ingot

A: Chill zone
B: Columnar zone
C: Branched columnar zone (A-segregation zone)
D: Equiaxed zone (V-segregation zone)
E: Sedimental zone (Negative segregation zone)
F: Positive segregation zone

Fig. 5 SULPHUR PRINT AND THE CLASSIFICATION OF SEGREGATIONS AND SOLIDIFICATION STRUCTURE OF LONITUDINAL SECTION OF 75 TON INGOT
**FIG. 6** FLOW CHART OF LADLE REFINING PROCESS

**FIG. 7** PROCESS FOR THE PRODUCTION OF EXTREMELY LOW P AND S STEEL BY EAF + LRF PROCESS
FIG. 8  EFFECT OF STIRRING ENERGY ON THE DEGREE OF HYDROGEN REMOVAL
FIG. 9 CHANGE IN HYDROGEN CONTENT DURING LRF PROCESS

FIG. 10 EFFECT OF $\dot{E}_M \cdot t$ ON THE DEGREE OF DEOXIDATION
FIG. 11 DISTRIBUTION OF HYDROGEN AND OXYGEN CONTENTS IN 570 TON INGOTS PRODUCED BY DOUBLE-DEGASSING PROCESS FOR NUCLEAR APPLICATION
FIG. 12 EFFECT OF DENSITY DIFFERENCE OF LIQUID ON CRITICAL $\varepsilon$ $\cdot$ R$^{1.1}$
FIG. 13 CRITICAL CONDITIONS FOR THE FORMATION OF MICROPOROSITIES IN A-SEGREGATION ZONE AND V-SEGREGATION ZONE
FIG. 14  EFFECT OF HEIGHT-TO-DIAMETER RATIO, H/D, ON THE LENGTH OF MICROPOROSITY ZONE
FIG. 15 DISTRIBUTION OF CARBON CONTENT ALONG THE AXIS OF INGOT

FIG. 16 DISTRIBUTION OF CARBON CONTENT IN 570 TON Mn-Mo-Ni STEEL INGOT
(a) One-piece forged ring

(b) Four pieces of bent forging, electro-slag welded to form a ring

Fig. 17 Combined vessel flange and nozzle belt forging of PWRPV, KWU/1300 MWe. Weight is 165 ton with 8 nozzle necks welded.

(a) Combined vessel flange and nozzle belt design: set-on type nozzles (developed by COCKERILL)

(b) Conventional design: vessel flange and nozzle belt separated, set-in type nozzles

Fig. 18 WEC/157" PWRPV, combined vessel flange and nozzle belt forging as compared with conventional one.
<table>
<thead>
<tr>
<th>Process</th>
<th>Sketch</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flame cutting</td>
<td><img src="image1" alt="Flame cutting Sketch" /></td>
</tr>
<tr>
<td>Upsetting</td>
<td><img src="image2" alt="Upsetting Sketch" /></td>
</tr>
<tr>
<td>Piercing by punch</td>
<td><img src="image3" alt="Piercing by Punch Sketch" /></td>
</tr>
<tr>
<td>Enlarging</td>
<td><img src="image4" alt="Enlarging Sketch" /></td>
</tr>
<tr>
<td>Repetition of enlarging and upsetting</td>
<td>Several heatings are required</td>
</tr>
<tr>
<td>Enlarging</td>
<td><img src="image5" alt="Enlarging Sketch" /></td>
</tr>
<tr>
<td>Finish forging</td>
<td><img src="image6" alt="Finish Forging Sketch" /></td>
</tr>
</tbody>
</table>

**Fig. 19** Forging process
Forging procedure for each test block

As cast condition

Solid forging

Upsetting

(Dimensions are the same as cast condition)

<table>
<thead>
<tr>
<th>Test Block No.</th>
<th>D₀</th>
<th>d₀</th>
<th>Forging Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>400</td>
<td>—</td>
<td>As cast</td>
</tr>
<tr>
<td>2</td>
<td>400</td>
<td>350</td>
<td>1.3</td>
</tr>
<tr>
<td>3</td>
<td>400</td>
<td>325</td>
<td>1.5</td>
</tr>
<tr>
<td>4</td>
<td>400</td>
<td>280</td>
<td>2.0</td>
</tr>
<tr>
<td>5</td>
<td>400</td>
<td>250</td>
<td>2.5</td>
</tr>
</tbody>
</table>

Fig. 20 Variation of Charpy impact value due to repetition of forging
(Material ; 20 MnMoNi 55)
Fig. 21 Effect of forging ratio and anisotropy of material

(Compared with complete data)

Matrix without segregation

Mn : 0.50%, P : 0.05%, S : 0.03%
C : 0.20%

Material: JAN data
Complete data: --
Transverse to RD:
Complete data: --
Transverse to RD, JIS's data:
Complete data: --
Matrix working direction (MD)'

Impact value: kg-m

Reduction of area; %

Tensile strength; kg/mm²
a) For enlarging and mandrel forging

Logarithmic strain: 
\[ \varepsilon_t = \ln \frac{D_m}{dm} \] in tangential direction
\[ \varepsilon_a = \ln \frac{L}{l} \] in axial direction
\[ \varepsilon_r = \ln \frac{T}{t} \] in radial direction
and \[ \varepsilon_t + \varepsilon_a + \varepsilon_r = 0 \] as the volume of the forging is constant.

b) For solid forging and upsetting

Logarithmic strain: 
\[ \varepsilon_t = \ln \frac{D}{d} \] in tangential direction
\[ \varepsilon_a = \ln \frac{L}{l} \] in axial direction
\[ \varepsilon_r = \ln \frac{D}{d} \] in radial direction
and \[ \varepsilon_t = \varepsilon_r = -1/2 \varepsilon_a \]

Fig. 22 Logarithmic strains in three directions
Fig. 23 Relation between logarithmic strain and mechanical properties (Material: 20 MnMoNi 55)

**Note:**
- St: Tangential for Shell
- Sa: Axial for RPV
- Sr: Radial for pressurizer

**Legend:**
- Pr: Radial
- Sr: Tangential
- Nt: Radial
- Na: Tangential
- Pt: Radial
- Nozzle
- Axial
- Tangential
Fig. 24 Preliminary heat treatment after forging for preventing hydrogen flake.
Fig. 25 Carbon distribution on integrated flange forgings made from a 400 ton and a 500 ton ingot.
(a) Through-thickness mechanical properties for KwU's shell flange made from a 400 ton ingot

(b) Through-thickness mechanical properties for monoblock vessel flange made from a 500 ton ingot

Fig. 26 Through-thickness mechanical properties for integrated flange forging
<table>
<thead>
<tr>
<th>Properties</th>
<th>Test at Location</th>
<th>Room temp.</th>
<th>350°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Yield strength</td>
<td>Top</td>
<td>(455)</td>
<td>(40.1)</td>
</tr>
<tr>
<td></td>
<td>Middle</td>
<td>(48.7)</td>
<td>(41.8)</td>
</tr>
<tr>
<td></td>
<td>Bottom</td>
<td>(45.5)</td>
<td>(40.1)</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>Top</td>
<td>(61.3)</td>
<td>(57.1)</td>
</tr>
<tr>
<td></td>
<td>Middle</td>
<td>(64.5)</td>
<td>(59.0)</td>
</tr>
<tr>
<td></td>
<td>Bottom</td>
<td>(61.4)</td>
<td>(56.0)</td>
</tr>
<tr>
<td>Elongation (%)</td>
<td>Top</td>
<td>(24.2)</td>
<td>(19.5)</td>
</tr>
<tr>
<td></td>
<td>Middle</td>
<td>(22.4)</td>
<td>(22.0)</td>
</tr>
<tr>
<td></td>
<td>Bottom</td>
<td>(25.0)</td>
<td>(20.0)</td>
</tr>
<tr>
<td>Reduction of area</td>
<td>Top</td>
<td>(75.5)</td>
<td>(70.8)</td>
</tr>
<tr>
<td></td>
<td>Middle</td>
<td>(71.9)</td>
<td>(74.0)</td>
</tr>
<tr>
<td></td>
<td>Bottom</td>
<td>(73.0)</td>
<td>(71.9)</td>
</tr>
<tr>
<td>Impact value</td>
<td>Top</td>
<td></td>
<td>(17.2)</td>
</tr>
<tr>
<td>at 0°C (kg/m²)</td>
<td>Middle</td>
<td>(22.8)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Bottom</td>
<td>(23.3)</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 27 Mechanical properties at various height locations for KWU's shell flange

<table>
<thead>
<tr>
<th>Properties</th>
<th>Test at Direction</th>
<th>Room temp.</th>
<th>350°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Yield strength</td>
<td>Tangential</td>
<td>(42.2)</td>
<td>(55.7)</td>
</tr>
<tr>
<td></td>
<td>Axial</td>
<td>(42.2)</td>
<td>(55.3)</td>
</tr>
<tr>
<td></td>
<td>Radial</td>
<td>(45.9)</td>
<td>(56.7)</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>Tangential</td>
<td>(59.2)</td>
<td>(54.9)</td>
</tr>
<tr>
<td></td>
<td>Axial</td>
<td>(58.7)</td>
<td>(54.8)</td>
</tr>
<tr>
<td></td>
<td>Radial</td>
<td>(59.5)</td>
<td>(55.4)</td>
</tr>
<tr>
<td>Elongation (%)</td>
<td>Tangential</td>
<td>(24.5)</td>
<td>(25.4)</td>
</tr>
<tr>
<td></td>
<td>Axial</td>
<td>(24.9)</td>
<td>(23.0)</td>
</tr>
<tr>
<td></td>
<td>Radial</td>
<td>(24.2)</td>
<td>(19.6)</td>
</tr>
<tr>
<td>Reduction of area</td>
<td>Tangential</td>
<td>(70.5)</td>
<td>(69.7)</td>
</tr>
<tr>
<td></td>
<td>Axial</td>
<td>(69.2)</td>
<td>(60.9)</td>
</tr>
<tr>
<td></td>
<td>Radial</td>
<td>(64.0)</td>
<td>(59.7)</td>
</tr>
</tbody>
</table>

Fig. 28 Directionality of tensile properties for KWU's shell flange
Fig. 29  Directionality of impact value for integrated flange forgings
Welding direction

HAZ (Coarse grained and refined)

Liquation cracking — Reheat cracking

Micro-cracks and cracks in mm range

Macro-cracks HAZ plane

Fig. 30: CRACKING IN WELD HAZ IN MULTI-PASS STRUCTURAL WELDS (AFTER KUSSMAUL)
**FIGURE 31. LOCATION OF REHEAT UNDERCLAD CRACKS WITH RESPECT TO CLADDING DEPOSITS (AFTER VINCKIER AND PENSE (63))**
Heat affected coarse grain area

HAZ (Ac1)

Reheat cracking susceptible area

(a) One layer cladding

Reheat cracking susceptible area

Refined HAZ by 2nd layer cladding

(b) Two layer cladding: area not refined by 2nd layer remained (old procedure)

Refined HAZ by 2nd layer cladding

(c) Two layer cladding: new procedure for the starting bead of first layer cladding

Fig. 32 Location of reheat UCCs and their remedy: a schematic drawing.
FIG. 33 Location of new type reheat UCG found in two-layer cladding: Old procedure (b), and its remedy by new procedure (c).
Fig. 34 Development in material layout of a PWRPV/173-182
Left—conventional design
Right—advanced design

Fig. 35 Development in material layout of a BWRRV/251
Left—conventional design
Right—advanced design
Fig. 36  1350MWe A-BWRPV
Chapter 4

WWER REACTOR PRESSURE VESSEL DESIGN

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1. INTRODUCTION

The General design of water cooled and moderated reactors in USSR concentrated from the very beginning on PWR type reactors, the so-called "WWER" type (vodo-vodianyi energeticheskiy reactor = water-water power reactor) where light water was used for better cooling and moderation in the reactor core. This type of reactor had four development steps, main parameter which are shown in Table I.
Table I. Main parameters of WWER type reactors

<table>
<thead>
<tr>
<th>parameter</th>
<th>WWER-210</th>
<th>WWER-365</th>
<th>WWER-440</th>
<th>WWER-1000</th>
</tr>
</thead>
<tbody>
<tr>
<td>electric output, MW</td>
<td>210</td>
<td>365</td>
<td>440</td>
<td>1000</td>
</tr>
<tr>
<td>working pressure, MPa</td>
<td>10.0</td>
<td>10.5</td>
<td>12.5</td>
<td>15.7</td>
</tr>
<tr>
<td>reactor inlet water temperature, °C</td>
<td>252</td>
<td>252</td>
<td>268</td>
<td>289</td>
</tr>
<tr>
<td>reactor outlet water temperature, °C</td>
<td>273</td>
<td>280</td>
<td>301</td>
<td>322</td>
</tr>
<tr>
<td>number of loops</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>4</td>
</tr>
<tr>
<td>maximum outer diameter of reactor vessel, m</td>
<td>4.4</td>
<td>4.4</td>
<td>4.27</td>
<td>4.535</td>
</tr>
<tr>
<td>reactor vessel height, m</td>
<td>11.34</td>
<td>11.80</td>
<td>11.80</td>
<td>10.85</td>
</tr>
</tbody>
</table>

2. DESIGN CONCEPTS

Reactor pressure vessels are thick-walled cylindrical vessels with spherical domes, as it is shown in Fig.1. Reactor pressure vessels of WWER type reactors have some significant features that are different from Western design that derive from their overall
specification. For example, the reactor pressure vessel (as well as all other components) must be transportable on land, i.e. by rail or by road.

This requirement led to some design consequences, mainly (see Table I and II):

- decreased vessel diameter which results in a smaller water reflector thickness and thus higher neutron fluxes at the reactor vessel, which leads to the requirement for a PV material which has a high resistance against irradiation embrittlement,

- decreased mass of vessel, that led to the requirement for higher strength properties of materials to the decreased vessel wall thickness,

- two nozzle rings, an upper one for outlet nozzles, and a lower one for inlet nozzles. This feature result in the presence of a weldment between the upper and lower nozzle rings also a partition of temperatures in the vessel. Also there is no temperature gradient in circumferential direction in any nozzle ring. This feature also resultsin a larger vessel length,

- vessels are made only from forgings, (i.e. from cylindrical rings) and plates forged into domes. The spherical parts of the vessel’s bottom and cover are stamped from forged
plates; in the ex-USSR these forged domes are made from a plate, welded from two smaller ones by electroslag welding. In the ČSFR the forged are made from one large plate prepared by a development of one ingot. No axial weld is allowed in vessels,

- inlet and outlet nozzles are not welded to the nozzle ring, but they are machined from a thicker forged ring for the older WWER-440 vessels, but forged in from a thick forged ring for WWER-1000 vessels,

- materials for the pressure vessels of a different type than there are used elsewhere, see Table III,

- not all the pressure vessels were wholly clad in austenitic stainless steel,

- all austenitic steels, used for cladding, internals and for primary piping that are in contact with the water coolant, are stabilized.

Table II. Main parameters of reactor pressure vessels

<table>
<thead>
<tr>
<th>parameter</th>
<th>WWER-440</th>
<th>WWER-1000</th>
</tr>
</thead>
<tbody>
<tr>
<td>230 Model</td>
<td>213 Model</td>
<td>320 Model</td>
</tr>
<tr>
<td>Parameter</td>
<td>Value 1</td>
<td>Value 2</td>
</tr>
<tr>
<td>------------------------------------------------</td>
<td>----------</td>
<td>----------</td>
</tr>
<tr>
<td>mass, t</td>
<td>215</td>
<td>320</td>
</tr>
<tr>
<td>length, m</td>
<td>11.800</td>
<td>11.000</td>
</tr>
<tr>
<td>outer diameter, m</td>
<td></td>
<td></td>
</tr>
<tr>
<td>in cylindrical part</td>
<td>3.840</td>
<td>4.535</td>
</tr>
<tr>
<td>in nozzle region</td>
<td>3.980</td>
<td>4.660</td>
</tr>
<tr>
<td>wall thickness (without cladding), m</td>
<td></td>
<td></td>
</tr>
<tr>
<td>in cylindrical part</td>
<td>0.140</td>
<td>0.193</td>
</tr>
<tr>
<td>in nozzle ring</td>
<td>0.190</td>
<td>0.285</td>
</tr>
<tr>
<td>working pressure, MPa</td>
<td>12.26</td>
<td>17.65</td>
</tr>
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<td>design pressure, MPa</td>
<td>13.7</td>
<td>15.7</td>
</tr>
<tr>
<td>hydrotest pressure, MPa</td>
<td>17.1</td>
<td>19.2</td>
</tr>
<tr>
<td>design wall temperature, °C</td>
<td>325</td>
<td>350</td>
</tr>
<tr>
<td>vessel lifetime, y</td>
<td>30</td>
<td>40</td>
</tr>
<tr>
<td>vessel irradiation temperature, °C</td>
<td>268</td>
<td>268</td>
</tr>
<tr>
<td>total thickness of steel</td>
<td></td>
<td></td>
</tr>
<tr>
<td>in shielding between core</td>
<td></td>
<td></td>
</tr>
<tr>
<td>and RPV, m</td>
<td>0.100</td>
<td>0.103</td>
</tr>
<tr>
<td>total thickness of water</td>
<td></td>
<td></td>
</tr>
<tr>
<td>in shielding between core</td>
<td></td>
<td></td>
</tr>
<tr>
<td>and RPV, m</td>
<td>0.240</td>
<td>0.228</td>
</tr>
<tr>
<td>maximum neutron fluence during design lifetime, m² (E &gt;0.5 MeV)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>- base metal</td>
<td>2.3×10²⁴</td>
<td>2.6×10²⁴</td>
</tr>
<tr>
<td>- weld metal</td>
<td>1.6×10²⁴</td>
<td>1.8×10²⁴</td>
</tr>
</tbody>
</table>
maximum neutron fluence during design lifetime, m^2 (E >1 MeV)
- base metal  1.4x10^{24}  1.6x10^{24}  3.7x10^{23}
- weld metal  1.0x10^{24}  1.1x10^{24}  3.4x10^{23}

cover mass, t
50  90

number of nozzles
37+18  61+30

3. REACTOR PRESSURE VESSEL MATERIALS

Table III. RPV materials

<table>
<thead>
<tr>
<th>reactor type</th>
<th>V-230</th>
<th>V-213</th>
<th>V-320</th>
</tr>
</thead>
<tbody>
<tr>
<td>base materials:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>- cylindrical rings</td>
<td>15Kh2MFA</td>
<td>15Kh2FAA</td>
<td>15Kh2NMFAA</td>
</tr>
<tr>
<td>- other parts of vessel</td>
<td>15Kh2MFA</td>
<td>15Kh2MFA</td>
<td>15Kh2NMFA</td>
</tr>
<tr>
<td>- cover</td>
<td>18Kh2MFA</td>
<td>18Kh2MFA</td>
<td>15Kh2NMFA</td>
</tr>
<tr>
<td>- free flange</td>
<td>25Kh3MFA</td>
<td>25Kh2MFA</td>
<td>-</td>
</tr>
<tr>
<td>- stud bolts and nuts</td>
<td>25Kh1MF</td>
<td>38KhN3MFA</td>
<td>38KhN3MFA</td>
</tr>
</tbody>
</table>

welding materials:
- automatic submerged arc Sv-15KhMFT Sv-15KhMFT Sv-12Kh2N2M
  + AN-42  + AN-42M  + FC-16A
In USSR a special high strength heat and radiation resistance steel of Cr-Mo-V type (marked 48-TS) was developed for WWER reactor vessels from the very beginning. This steel was designated according to its chemical composition as 15Kh2MFA. Chemical composition of this steel and its heat treatment was chosen to ensure the required values of strength, ductility and toughness as well as satisfactory weldability. The choice of Cr-Mo-V type of steel also ensures an increased resistance against temper embrittlement during vessel fabrication (which requires multiple high temperature tempering) and against thermal ageing and irradiation embrittlement during operation.

The mechanical properties of steels are determined to a large extent by their structure which depends in turn on their alloy composition and heat treatment. The requirements for high strength properties and high toughness in thick-walled materials, are achieved by quenching from temperatures about 980-1000 °C followed by high tempering at temperatures about 680-700 °C. The principal task when selecting steel alloy composition is to provide through-thickness properties and precipitation of thermodynamically stable carbide phase during tempering. The carbide phase should be fine dispersed and uniformly distributed over the body of ferrite to fix a fragmented dislocation sub-structure. Chromium and nickel alloying is the most
significant factor in producing properties of reactor steel matrix. Optimum proportions of carbon, molybdenum and vanadium determine to a great extent the heat and tempering resistance of steel and such impurities as phosphorus, copper, tin and arsenic in a combination with nickel and manganese play the main role in thermal and irradiation embrittlement.

Thus, steel with low carbon content (0.13-0.18wt%) was used for vessel of thicknesses up to 400 mm, that has to be welded. Steels with medium carbon content (0.18-0.21wt%) were used for vessel heads. And finally, steels with with higher carbon content (0.22-0.27wt%) for free flanges with semi-product thicknesses up to 600 mm but which were not welded.

To ensure appropriate structure and properties in heat-affected zones of welding joints, resistance against "hot" and "cold" cracks, as well as underclad cracks, it is necessary to provide high preheating for welding at a temperature range of 350-400 °C.

A need for improvement of properties of steel arose during the design of WWER-1000 reactor pressure vessel. Thus, a new alloy steel of the Cr-Ni-Mo-V type was investigated. Nickel is one of alloying elements that increases the strength of ferrite without decreasing its toughness, and provides better quenchability. On the other hand, a concentration of more than 1.5 wt% of nickel in Cr-Mo-V steels is not favourable, as it increases susceptibility of steel to temper, thermal, and irradiation embrittlement.
A new type of steel of 15Kh2NMFA for WWER-1000 vessels was developed and brought into use. Alloying of reactor steel with a chromium content within the range 1.8 to 2.7 wt% and decreasing the vanadium content to about 0.15 wt% also improved resistance to cracking in the heat affected zones of welded joints during heat treatment. This allows a simplification of reactor vessel welding procedure because of a decrease in preheating temperature before welding (150 to 200 °C) as well as in the temperature for intermediate and final tempering (620 and 650 °C, respectively).

Table IV. Chemical composition of materials (wt%)

<table>
<thead>
<tr>
<th>material</th>
<th>C</th>
<th>Mn</th>
<th>Si</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Ni</th>
<th>Mo</th>
<th>V</th>
</tr>
</thead>
<tbody>
<tr>
<td>15Kh2MFA</td>
<td>0.13</td>
<td>0.30</td>
<td>0.17</td>
<td>max.</td>
<td>max.</td>
<td>2.50</td>
<td>max.</td>
<td>0.60</td>
<td>0.25</td>
</tr>
<tr>
<td></td>
<td>0.18</td>
<td>0.60</td>
<td>0.37</td>
<td>0.025</td>
<td>0.025</td>
<td>3.00</td>
<td>0.40</td>
<td>0.80</td>
<td>0.35</td>
</tr>
<tr>
<td>18Kh2MFA</td>
<td>0.15</td>
<td>0.30</td>
<td>0.17</td>
<td>max.</td>
<td>max.</td>
<td>2.50</td>
<td>max.</td>
<td>0.60</td>
<td>0.25</td>
</tr>
<tr>
<td></td>
<td>0.21</td>
<td>0.60</td>
<td>0.37</td>
<td>0.025</td>
<td>0.025</td>
<td>3.00</td>
<td>0.40</td>
<td>0.80</td>
<td>0.35</td>
</tr>
<tr>
<td>25Kh3MFA</td>
<td>0.22</td>
<td>0.30</td>
<td>0.17</td>
<td>max.</td>
<td>max.</td>
<td>2.80</td>
<td>max.</td>
<td>0.60</td>
<td>0.25</td>
</tr>
<tr>
<td></td>
<td>0.25</td>
<td>0.60</td>
<td>0.37</td>
<td>0.025</td>
<td>0.025</td>
<td>3.30</td>
<td>0.40</td>
<td>0.80</td>
<td>0.35</td>
</tr>
<tr>
<td>15K2NMFA</td>
<td>0.13</td>
<td>0.30</td>
<td>0.17</td>
<td>max.</td>
<td>max.</td>
<td>1.80</td>
<td>1.00</td>
<td>0.50</td>
<td>max.</td>
</tr>
<tr>
<td></td>
<td>0.18</td>
<td>0.60</td>
<td>0.37</td>
<td>0.020</td>
<td>0.020</td>
<td>2.30</td>
<td>1.50</td>
<td>0.70</td>
<td>0.10</td>
</tr>
<tr>
<td>Sv-10KhMFT</td>
<td>0.04</td>
<td>0.60</td>
<td>0.20</td>
<td>max.</td>
<td>max.</td>
<td>1.20</td>
<td>max.</td>
<td>0.35</td>
<td>0.10</td>
</tr>
<tr>
<td>+AN-42</td>
<td>0.12</td>
<td>1.30</td>
<td>0.60</td>
<td>0.042</td>
<td>0.035</td>
<td>1.80</td>
<td>0.30</td>
<td>0.70</td>
<td>0.35</td>
</tr>
<tr>
<td>Sv-10KhMFT</td>
<td>0.04</td>
<td>0.60</td>
<td>0.20</td>
<td>max.</td>
<td>max.</td>
<td>1.20</td>
<td>max.</td>
<td>0.35</td>
<td>0.10</td>
</tr>
<tr>
<td>+AN-42M</td>
<td>0.12</td>
<td>1.30</td>
<td>0.60</td>
<td>0.012</td>
<td>0.015</td>
<td>1.80</td>
<td>0.30</td>
<td>0.70</td>
<td>0.35</td>
</tr>
<tr>
<td>Sv-12Kh2N2MA</td>
<td>0.05</td>
<td>0.50</td>
<td>0.15</td>
<td>max.</td>
<td>max.</td>
<td>1.40</td>
<td>1.20</td>
<td>0.45</td>
<td>-</td>
</tr>
</tbody>
</table>
Guaranteed mechanical properties of materials for both types of reactor pressure vessels, i.e. for WWER-440 and WWER-1000 units are given in Table V.

<table>
<thead>
<tr>
<th>Material</th>
<th>Rp0.2</th>
<th>Rm</th>
<th>A5</th>
<th>Z</th>
<th>Rp0.2</th>
<th>Rm</th>
<th>A5</th>
<th>Z</th>
<th>Tk0</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MPa</td>
<td>MPa</td>
<td>%</td>
<td>%</td>
<td>MPa</td>
<td>MPa</td>
<td>%</td>
<td>%</td>
<td>°C</td>
</tr>
<tr>
<td>+FC-16</td>
<td>0.12</td>
<td>1.00</td>
<td>0.45</td>
<td>0.025</td>
<td>0.020</td>
<td>2.10</td>
<td>1.900.75</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sv-12Kh2N2MA</td>
<td>0.05</td>
<td>0.50</td>
<td>0.15</td>
<td>max.</td>
<td>max.</td>
<td>1.40</td>
<td>1.20</td>
<td>0.45</td>
<td></td>
</tr>
<tr>
<td>+FC-16A</td>
<td>0.12</td>
<td>1.00</td>
<td>0.45</td>
<td>0.012</td>
<td>0.015</td>
<td>2.10</td>
<td>1.900.75</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sv-13Kh2MFT</td>
<td>0.11</td>
<td>0.40</td>
<td>0.17</td>
<td>max.</td>
<td>max.</td>
<td>1.40</td>
<td>-</td>
<td>0.40</td>
<td>0.17</td>
</tr>
<tr>
<td>+OF-6</td>
<td>0.16</td>
<td>0.70</td>
<td>0.35</td>
<td>0.030</td>
<td>0.030</td>
<td>2.50</td>
<td>0.80</td>
<td>0.35</td>
<td></td>
</tr>
<tr>
<td>A 533-B,Cl.1</td>
<td>max.</td>
<td>1.15</td>
<td>0.15</td>
<td>max.</td>
<td>max.</td>
<td>-</td>
<td>0.40</td>
<td>0.45</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.25</td>
<td>1.30</td>
<td>0.30</td>
<td>0.035</td>
<td>0.035</td>
<td>0.70</td>
<td>0.60</td>
<td></td>
<td></td>
</tr>
<tr>
<td>A 508,Cl.30</td>
<td>0.15</td>
<td>1.20</td>
<td>0.15</td>
<td>max.</td>
<td>max.</td>
<td>-</td>
<td>0.40</td>
<td>0.45</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.25</td>
<td>1.50</td>
<td>0.35</td>
<td>0.025</td>
<td>0.025</td>
<td>0.80</td>
<td>0.60</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table V. Guaranteed mechanical properties of materials
Long term properties of materials such as resistance to thermal ageing and irradiation embrittlement determine reactor pressure vessel lifetime. Initial studies showed that Cr-Mo-V (15Kh2MFA) steel had excellent resistance to both types of ageing. Thus no special requirements for steels and their welded joints were prescribed. As this type of steel needed to be of a higher quality than was prescribed for chemical and thermal vessels, a grade designated -A was specified. See Table II.

Only later, in the early 1980s, when the deteriorating influence of some residual elements (copper, phosphorus etc.) was established, supplementary requirements for materials composition and properties were included in the specification. Materials manufactured to these additional requirements are designated -AA, i.e. having a high quality. These requirements are summarized in Table VI. Only such materials are now used for cylindrical shells.
(base materials and welded joints) that are adjacent to the reactor core region (beltline).

Table VI. Specification for materials for active core shells (wt %)

<table>
<thead>
<tr>
<th>material</th>
<th>P</th>
<th>S</th>
<th>Cu</th>
<th>As</th>
<th>Sb</th>
<th>Sn</th>
<th>P+Sb+Sn</th>
<th>Co</th>
</tr>
</thead>
<tbody>
<tr>
<td>15Kh2MFAA</td>
<td>0.012</td>
<td>0.015</td>
<td>0.08</td>
<td>0.010</td>
<td>0.005</td>
<td>0.005</td>
<td>0.015</td>
<td>0.020</td>
</tr>
<tr>
<td>15Kh2NMFAA</td>
<td>0.010</td>
<td>0.012</td>
<td>0.08</td>
<td>0.010</td>
<td>0.005</td>
<td>0.005</td>
<td>0.015</td>
<td>0.020</td>
</tr>
<tr>
<td>A 533-B,Cl.1</td>
<td>0.012</td>
<td>0.015</td>
<td>0.10</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Irradiation embrittlement of reactor pressure vessel materials can be characterized by a shift of critical temperature of brittleness $T_{k0}$ (similar to a Charpy-V transition temperature, and is roughly equal to $R T_{NDT}$ for given materials) and according to Refs [1] and [2] can be described by the following relation:

$$\Delta T_F = A_F \times (F \times 10^{22})^{1/3}$$

where $A_F$ is the irradiation embrittlement coefficient

$F$ is the fast neutron fluence, n.m$^2$ (E > 0.5 MeV)

Values of this irradiation embrittlement coefficient are given in Soviet Code [2] and are summarized in Table VII:
Table VII. Values of irradiation embrittlement coefficients ($A_F$)

<table>
<thead>
<tr>
<th>Material</th>
<th>Irradiation temperature</th>
<th>Irradiation embrittlement coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>°C</td>
<td>$A_F$</td>
</tr>
<tr>
<td>15Kh2MFA</td>
<td></td>
<td></td>
</tr>
<tr>
<td>- base metal</td>
<td>270</td>
<td>18</td>
</tr>
<tr>
<td>- A/S weld metal</td>
<td>270</td>
<td>800(P+0.07 Cu)</td>
</tr>
<tr>
<td>15Kh2MFAA</td>
<td></td>
<td></td>
</tr>
<tr>
<td>- base metal</td>
<td>270</td>
<td>12</td>
</tr>
<tr>
<td>- A/S weld metal</td>
<td>270</td>
<td>15</td>
</tr>
<tr>
<td>15Kh2NMFAA</td>
<td></td>
<td></td>
</tr>
<tr>
<td>- base metal</td>
<td>290</td>
<td>23</td>
</tr>
<tr>
<td>- A/S weld metal</td>
<td>290</td>
<td>20</td>
</tr>
</tbody>
</table>

From Table VII and Fig. 2, 3 and 4 it is clearly seen that

- steel of 15Kh2MFA type has higher resistance to irradiation embrittlement with respect to ASTM A 533-B type of steel (even its irradiation temperature is by about 20 °C lower than suggestions for Reg.Guide 1.99, Rev. 1 as well as Rev. 2 [2]) that is probably caused by the absence of nickel as an alloying element and by a presence of vanadium that
creates very stable carbides (see Figs. 2 and 3),
- 15Kh2NMFA steel has a similar resistance to irradiation embrittlement to that of ASTM A 533-B type. The relatively high content of nickel (more than 1%) reduces advantages of the previous type of steel (see Fig. 4 - comparison with data from Reg. Guide 1.99, Rev. 2).

4. SAFETY CONCEPTS

Reactor pressure vessels for the first WWER-440 reactors were designed according to Ref [1] that used the "temperature approach" for assuring their resistance to brittle fracture. A "Critical temperature of brittleness", \( T_k \), was used as the main parameter and a simplified Fracture Analysis Diagram as the principal approach for assuring against brittle fracture.

\( T_k \) is determined from Charpy impact notch toughness testing - the criterial values depend on the yield strength of material; second criterion is 50% of ductile fracture appearance at temperature equal to \( T_k + 30 \, ^\circ C \).

\( T_k \) during operation was defined as:

\[ T_k = T_{k0} + \Delta T_f + \Delta T_T + \Delta T_N \]

where:
- \( T_{k0} \) is the initial critical temperature of brittleness
- \( \Delta T_f \) is the shift in \( T_{k0} \) due to irradiation
\( \Delta T_T \) is the shift in \( T_{k0} \) due to thermal ageing
\( \Delta T_N \) is the shift in \( T_{k0} \) due to fatigue damage.

Safety concepts was then defined by the formula:

\[
T > T_i + \Delta T_S
\]

where: \( \Delta T_S \) is the safety factor (given from FAD) and equal to + 30 °C

Later, the new generation of WWER-440 and all WWER-1000 reactors were designed according to Ref [2], which used the "Linear Elastic Fracture Mechanics" approach for assuring resistance to brittle fracture. In this calculations only the initiation approach is used, thus, static fracture toughness, \( K_{IC} \), is used. No initiation of a crack is allowed for a "calculated (postulated) defect". Depth of this defect is equal to one quarter of vessel wall thickness with a length which is equal to six times of the depth.

Resistance to brittle fracture is considered to be assured if the following relation is met for this calculated defect:

\[
K_i < [K_{IC}]_i
\]

where: \( K_i \) is the stress intensity factor of a calculated defect
\( [K_{IC}]_i \) is the allowable values of stress intensity factor

Index "i" demonstrates that allowable stress intensity factors
have different values for different operating conditions:

- **i=1** - normal operating conditions safety factors
  \[ n_K = 2, \Delta T = +30 \, ^\circ C \]

- **i=2** - operational events and hydrotests
  \[ n_K = 1.5, \Delta T = +30 \, ^\circ C \]

- **i=3** - accident (emergency cooling) conditions
  \[ n_K = 1, \Delta T = 0 \, ^\circ C \]

Allowable stress intensity values, \([K_{IC}]_i\), are then received as a lower bound curve of two curves, calculated from the initial one, using shown safety factors. Temperature dependence of \([K_{IC}]_i\), was set up as a lower boundary curve of all accessible fracture toughness values of materials for RPVs using mathematical statistics methods.

General dependencies for all materials now permitted allowed for WWER RPVs are given below [2]:

\[
[K_{IC}]_1 = 13 + 18 \exp(0.02(T-T_k))
\]
\[
[K_{IC}]_2 = 17 + 24 \exp(0.018(T-T_k))
\]
\[
[K_{IC}]_3 = 26 + 36 \exp(0.02(T-T_k))
\]

which means that the reference temperature is the critical temperature of brittleness, \(T_k\), defined earlier.

Calculation of stress intensity factor of calculated defect is carried out according to formulae given in [2]:

\[
K_i(a) = \frac{1}{Q}(\sigma_p M_p + \sigma_q M_q)(\pi a)^{1/3}
\]

where: \(K_i(a)\) is the factor taking into account stress
concentrations in defect neighbourhood

\[ Q \]

is the defect shape factor

\[ \sigma_p \]

is the tensile stress component

\[ \sigma_q \]

is the bending stress component

\[ M_p, M_q \]

are coefficients

\[ a \]

is the defect depth

This formula is used for loading due to internal pressure, external bending as well as by temperature gradient during stationary and/or non-stationary conditions.

The calculated defect is the same for all calculated events, i.e. for normal conditions, hydrotests and for emergency cooling regimes.

5. MEASURES TAKEN FOR DESIGN LIFETIME ASSURANCE

In the early 1980s a difference was revealed between the indices of the irradiation embrittlement accepted in Code [2] and the results from surveillance specimens withdrawn from some WWER-440 reactors. On the basis of all data available on the radiation embrittlement of 15Kh2MFA steel and its weld metal - and with regard for comprehensive studies on the effect of impurity content - particularly the effect of copper and phosphorus, on the radiation embrittlement, empirical relationships were established of radiation embrittlement factors, \( A_F \), versus impurity content (see Table VII).
Analysis of radiation embrittlement factors for specific vessels carried out using the relationship given above showed that the value of $A_F$ for the base metal did not impose limitations on the reactor vessel radiation life. Performing the analysis for the weld metal became complicated because for "old" reactor vessels the impurity elements concentration in the weld was specified and recorded. Statistical data from the manufacturer were used to estimate phosphorus content - this was related to the time of vessels fabrication. Concentration of each element in the weld metal was determined as a sum of the content of the particular element in welding wire and its increase or decrease on interaction of the wire with flux during welding. Copper content in weld metal was assumed equal to the copper content in the welding wire. Due to the absence of data on initial values of critical temperature of brittleness, $T_{k0}$, a mathematical model was developed which related $T_{k0}$ to the chemical composition.

The radiation embrittlement factors $A_F$ were calculated from the data on copper and phosphorus content in weld metals. The calculations showed that values of $A_F$ for weld metal of "old" (i.e. mostly for V-230 type) reactor vessels are between 32 and 43 while the design value was only 13. This fact required the repeated analysis of assurance of brittle fracture resistance. On the basis of these results, measures were recommended to assure operational safety of reactor vessels during design service lifetime.

These measures include:
correction of schedules of pressure and temperature variations under the conditions of reactor heating-up and cool-down, correction of permissible temperatures for pressure test versus time of reactor operation. This correction is performed in order that the vessel wall temperature is above that permitted with regard to the real transition temperature shift due to irradiation,

- reduction of temperature effects on the reactor vessel cylinder part under accident conditions - which is achieved by means of heating-up water stored in tanks of emergency core cooling system and in tanks of the emergency supply, both to minimum of 55 °C, and by means of changes to inlet nozzle of the pipelines from emergency feed pumps from "cold" leg to "hot" legs,

- exclusion of over cooling the primary circuit following a break in the steamlines by installing fast-response valves between steamlines of steam generators and main steam header and introducing additional interlockings,

- installation of dummy fuel assemblies (usually up to 36) to peripheral core fuel assemblies. Neutron flux to the reactor vessel wall reduces and results in a decrease of transition temperature shift of $T_{k0}$ due to irradiation,

- assurance by periodical in-service inspection of
vessels, primarily of the welds located in areas of intensive irradiation (i.e. welding joint 0.1.4).

Implementation of measures specifically recommended for each NPP unit allows the ensurance of safe operation of WWER-440 reactor vessels during the design service lifetime.

To increase knowledge about irradiation embrittlement of welding joints, additional activities have been proposed and mostly realized, specifically:

- accurate definition and knowledge of chemical composition, especially of copper and phosphorus contents of weld metal by removal of specimens from the inner surface in the case of reactors without austenitic cladding, or from the outer surface in case of those reactors with cladding,

- measurement of the neutron flux (fluence) on the outer pressure vessel wall (in cavity) and related to full size benchmark experiments in zero-power reactors with modelling of the active core, shielding and pressure vessel wall,

- measurement of hardness of irradiated vessel metals, the results of which are compared with empirical relationships and transformed into yield strength,
- taking samples from the inside of the reactor pressure vessel wall, making and testing small impact specimens correlated to standard Charpy-V tests,

- thermal annealing of the irradiated part of vessels to restore their initial mechanical properties, mainly the initial toughness of vessel materials.

6. SUMMARY

Reactor pressure vessels of the WWER type reactors are characterized by some differences when compared with Western types, mainly by:

- the requirements for transportation by land resulted in reduced vessel diameter, which caused a reduction in the dimension of the water gap and thus a higher neutron fluence on the reactor vessel wall,

- reactor vessels are manufactured from different steels,

- "old" models of WWER-440 reactors (mainly of V-230 type) showed a substantially greater increase in irradiation embrittlement due to high impurity content in comparison with the design model, thus special analysis together with recommendations for necessary corrective measures have been
performed to ensure design reactor lifetime.

REFERENCES


ILLUSTRATIONS:

Fig.1. Schematic diagram of WWER-440 (left) and WWER-1000 (right) reactor pressure vessels.

Fig.2. Comparison of Soviet Code [2] and Reg.Guide 1.99, Rev.1 for 15Kh2MFA type of steel and its welding joint (BM - base metal, WM - weld metal)

Fig.3. Comparison of Soviet Code [2] and Reg.Guide 1.99, Rev.2 for 15Kh2MFA type of steel

Fig.4. Comparison of Soviet Code [2] and Reg.Guide 1.99, Rev.2 for 15Kh2NMFA type of steel
5 Determination of Reliable Material Properties

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1 Introduction

The pressure retaining boundary of the primary circuit is subjected to complex loadings resulting from normal operation and exceptionally, from emergency conditions. To assure sufficient safety margins assuming that the structure contains flaws reliable material properties must be available. The safety margin can then be determined quantitatively when these parameters are considered in conjunction with the calculated stresses, strains and stress intensity values.

For the assessment of the safety margin, the ASME boiler and pressure vessel Code in the US and the KTA rules in Germany present normalized lower bound fracture toughness values for brittle crack initiation (static and dynamic) and crack arrest of commonly used reactor pressure vessel materials. In addition, methods are given which show how component specific lower bound fracture toughness curves can be derived from the normalized curves. These are based on material acceptance test data and surveillance results with respect to the design life time (DLT). The fracture mechanics requirements of these Codes, however, are limited to the linear elastic fracture mechanics regime and do not cover the elastic-plastic fracture mechanics regime. For all considerations the Charpy impact test and the data derived therefrom play a central role although this data do not directly allow the quantification of
the safety margin. Therefore, experiments with a variety of reactor pressure vessel steels of different quality, including irradiated materials, have been performed to prove the reliability of the implementation of the Charpy impact test into a quantitative fracture mechanics concept.

Additional consideration has been given in applying those results with particular regard to the size and geometry of the specimens and their transferability to complex structures. This considerations required a detailed understanding of the parameters affecting material failure as well as the development of experimental and analytical methods to describe the loading situation resulting from transients.

Much experimental effort has been undertaken to demonstrate the transferability of those results to the component in service by investigations on large scale specimens and model vessels.

While the changes in material as a Function of time is described in other chapters, the following part deals only with the applicability and transferability of results of given materials. It focuses on research work and on the validation of the underlying principles of the Code with regard to lower bound fracture toughness properties and analytical methods.

2 Evaluation of Fracture Mechanics Properties

Fracture mechanics data are needed for the whole temperature range of the operational loading regime. Because of the transition in fracture behaviour from brittle to ductile fracture, different theories in the linear elastic (LEFM) and the elastic-plastic (EPFM) regime have been developed and reliable fracture mechanics parameters must therefore be determined in both regimes. The lower bound fracture toughness curves of the Code are only valid for the LEFM regime.
2.1 Assessment of Reliable Fracture Mechanics Properties (LEFM)

The linear elastic fracture mechanics parameter is the fracture toughness ($K_{lc}$). It characterizes the onset of brittle fracture and can therefore be regarded as a crack initiation value. The experimental determination of $K_{lc}$ values is described e.g. in the American Test Standard ASTM E 399 [1], and the British Standard BS 5447 [2]. Both standards are similar. The results are only valid in the range that satisfies the requirements of linear elastic (plain strain) conditions. In principle, those data should be size independent and their transferability to the real component should be expected. Tests were performed with either compact tension (CT) specimens or three point bend (TPB) specimens. For both types of specimens all other dimensions are related to the specimen thickness which is the most important parameter with respect to the constraints necessary to provide plain strain conditions. From experience a validity criterion has been defined with regard to the thickness requirement of the specimens

$$B \geq \omega \left(\frac{K_{lc}}{\sigma_y}\right)^2$$

$B$ specimen thickness  
$K_{lc}$ fracture toughness value  
$\sigma_y$ yield strength  
$\omega$ the validity criterion (material dependent)

According to the standards, $\omega$ is required to be $\geq 2.5$, however, this value is still being considered because only limited information from large specimen testing is available. Data from testing large specimens indicated that even in the range where the validity criterion was fulfilled a further decrease in $K_{lc}$ values occurs with increasing specimen thickness as shown in Fig. 1. According to these test results lower bound values for $K_{lc}$ are yet reached at relatively high values of $\omega$. This general problem is under international
discussion and is being reflected in the work of the appropriate ASTM Committee which has suggested using a value of $\omega \geq 4$ [3].

The fracture toughness of a material depends strongly on the temperature. A lower bound fracture toughness curve $K_{lc}$ as a function of temperature relative to the Reference Nil Ductility Transition Temperature $R_{NDT}$ is given in Fig. 2 for a high strength fine grained reactor pressure vessel (RPV) steel [4, 5, 6]. As a consequence of this behavior tests can be performed with small specimens at low temperature whereas large specimens are needed at elevated temperature.

2.2 Assessment of Fracture Mechanics Characteristics Based on Charpy-V Notch Impact and Drop Weight Tests

The reference fracture toughness curves as discussed in section 2.1 above and used in the fracture mechanics approach is adjusted onto an absolute temperature scale by using the Reference Nil Ductility Transition Temperature $R_{NDT}$. This Reference Temperature is obtained from the Charpy-V notch impact test and the drop weight test [7], as discussed in Chapter 2. The determination of $R_{NDT}$ and thus the adjustment of the lower bound fracture toughness curve $K_{IR}$ includes data scatter deriving from both the Charpy and the drop weight test.

Because of the significance of those data in the safety analysis, results obtained with two different reactor pressure vessel steels (22 NiCrMo 37 with 90 J upper shelf energy and 20 MnMoNi 55 with 200 J upper shelf energy), tested in 10 different laboratories in Germany were compared [8]. Eighteen specimens were machined and tested in a temperature range of -100°C to 350°C by each participant. The mean values of the energy for the high upper shelf energy material (KS 13) are plotted against the temperature in Fig. 3. The upper part of the figure shows the standard deviation assuming a Gaussian
normal distribution. The corresponding data for the lateral expansion also necessary to determine the $RT_{NDT}$ is presented in Fig. 4. The test results include scatter deriving form the test machine and from the material. Similar behaviour was observed in the material with a low upper shelf value of 90 J.

Since the data for the determination of the Reference Nil Ductility Transition Temperature $RT_{NDT}$ are taken as the lower bound of three specimens tested at each temperature or from the lowest envelope of the energy-temperature curve, this procedure covers part of the scatter in a conservative way.

Ten laboratories participated in the drop-weight round robin test also each participant prepared its own specimens. The test results are shown in Fig. 5. The overall span of values was 30 K with a mean value at -25°C assuming a normal distribution. One of the main sources for the scatter was the individual evaluation of the sensitive criterion "break" or "no break" of the specimens. Discrepancies were observed between the evaluations of the fracture surface of the specimens and from the appearance of completely fractured specimens after heat tinting, especially in cases when the crack ran very closely to the edges. A more precise definition of the evaluation technique in Standards is desirable.

2.3 Elastic Plastic Fracture Mechanics Properties

Apart from the crack tip opening displacement (CTOD), the $J$-integral can be used to determine fracture mechanics parameters in the range of elastic-plastic material behaviour.
From the crack resistance curve ($J_R$-curve)

$$J = f (\Delta a)$$

$J$  J-integral
$\Delta a$  stable crack growth

characteristic values can be determined from individual points on the $J_R$ curve. In the past, different procedures were developed to assess fracture toughness data.

$J_{IC}$  according to ASTM E 813-88 [9]
$J_1, J_{0.2}, J_{0.2/h1}$  EGF P 1-90 [10]

The various criteria are defined in the corresponding standards. They make use of different algorithms to evaluate the J-integral from the load-line elongation results measured on the specimen and for the approximation of the $J_R$ curve from $J/\Delta a$ data pairs describing the crack resistance curve from which reliable crack initiation data are derived. An additional method has been introduced as a supplement to the existing standards to determine crack initiation values which are considered to be intrinsic material properties (physical initiation values) and thus can be transferred to complex components.

The characteristic data evaluated according to different methods are compared in Fig. 6. It is obvious that the data which are used to describe crack initiation differ quite markedly. In the example given the values vary from 85 N/mm to 167 N/mm.

With respect to transferability, it can be demonstrated that the physical crack initiation value $J_i$ [12] is a reliable property to use. The evaluation is based on a special fit of the $J/\Delta a$ data pairs and the intersection with an
experimentally determined blunting line derived from the "stretched zone" $\Delta a_1$. This is the region of extensive plastic deformation (crack tip blunting) developed before the onset of stable crack growth. The basic principle of this evaluation method is as follows: the stretched zone is completely formed during the blunting process and represents a "steady state volume" of highly deformed material which is maintained during the process of stable crack extension. Tests with different amounts of stable crack growth confirm the assumption of a constancy in the size of the stretched zone. After completion of the test, the stretched zone $\Delta a_1$ can be measured on the fracture surface e.g. by means of the scanning electron microscope (SEM) and can be separated from stable crack growth $\Delta a$ according to its different appearance. $J_i$ is derived from the $J_R$ curve as the intersection of the vertical line at $\Delta a_1$ with the $J_R$ curve, resulting in

$$J(\Delta a_1) = J_i$$

The $J$ value obtained from this equation is called crack initiation parameter $J_i$, Fig. 7. For a reliable determination of the $J_i$ value it is important that the difference between the approximation function and the $J/\Delta a$ points is minimized especially in the region of small crack growth. These features are neglected in the standards.

Nevertheless this procedure is comparable with the Japanese standard [11] and also with EGF P 1-90 [10]. The major difference can on the one hand be referred to the selection of the $J/\Delta a$ data points used to determine the crack resistance curve and on the other hand to the kind of polynomial approximation of the $J_R$ curve.

For a comparison with $K_{IC}$ data obtained in the linear elastic regime, $J$ values can be converted into $K_I$ values according to
\[ K_{IJ} = \frac{E \cdot J_I}{1-v^2} \]

- \( E \) Young's modulus
- \( v \) Transverse contraction coefficient
- \( K_{IJ} \) indicates that data was derived from J-integral test procedure

The \( J_I \) evaluation method is not only applicable in the plastic regime as, it is the Charpy upper shelf region, but also in the transition temperature regime. Scatter of data has, however to be taken into account.

When applying the different evaluation methods as seen in Fig. 6, sufficient safety margin against fracture is observed regardless of the evaluation method used - because of the large amount of crack growth before the occurrence of the fracture. The quantification of this margin in a real component is the aim of such a fracture mechanics analysis, additionally focusing on those parameters which give the best agreement with the failure behaviour of large and complex loaded structures.

The first step in investigating the transferability of fracture mechanics properties is a detailed analysis of the deformation behaviour of the individual laboratory specimen. To visualize the equivalence of the different crack initiation values, the data points are marked in different diagrams, Fig. 8, which are

a) load / crack opening displacement (COD)

b) load / stable crack extension (\( \Delta a \))

c) J-Integral / COD, and

d) J-Integral / \( \Delta a \)

as obtained on a typical RPV steel 20 MnMoNi 55 with a 20\% side grooved CT specimen (CT-25 mm) tested in the upper shelf
Charpy regime at 80°C. The position in the load / COD diagram indicates the margin from first crack initiation to maximum load carrying capability. This comparison shows clearly that the $J_1$ value lies on the curve before maximum load while the "pseudo" properties ($J_{0.2}$, $J_{0.2/b}$, $J_{IC}$), related to crack initiation, fall together with maximum load or even in the dropping branch of the load curve after maximum load has been reached in the case of the $J_{IC}$ value being determined according to ASTM E 813-88.

With respect to the necessary safety margin in the design of a component, material characteristics which are strongly influenced by the testing and evaluation procedure cannot be accepted. Therefore of all $J$ values, only the $J_1$ value, which can be considered to be an intrinsic material property describing the physical crack initiation should be applied. Similarly, $\delta_1$, the CTOD at initiation, yields a critical material property under given conditions, like temperature and loading rate.

2.4 Determination of Dynamic Fracture Mechanics Parameters

The lower limit fracture toughness curve ($K_{IA}$ or $K_{IR}$) specified in the Codes [4, 5, 6, 13] is based on crack arrest toughness ($K_{IA}$) and on dynamic fracture toughness values ($K_{ID}$). However the loading rate at which the dynamic fracture toughness data should be determined has not been defined. Tests can either be performed with CT-specimens in servo-hydraulic machines and rotary disc impact machines or with 3 point-bend specimens in pendulum and drop weight impact machines. Typical load rates are applied up to $\dot{K} = 10^6$ MPa$\sqrt{m}$/s. Only few test standards exist for the determination of dynamic fracture mechanics parameters. As a first step towards the standardization, ASTM have specified loading rates of $\dot{K} > 2.75$ MPa$\sqrt{m}$/s [1] in the linear elastic
Similar loading rates, ranging from $2.5 < K < 3200 \text{ MPa}\sqrt{\text{m}}/\text{s}$, are required in the British Standard [14]. According to those Standards tests can also be performed at higher loading rates if reliable techniques are available to measure load and load-line displacement during the dynamic test. This derives from tests on CT-specimens [15]. In the elastic plastic regime the requirements are similar to those specified in ASTM E 813 [9], but extended to a higher loading rate. Inertial forces are not considered in this evaluation of the J-integral which is equivalent to quasi-static loading conditions.

In recent studies, dynamic tests were performed at impact velocities from 0.04 to 20 m/s corresponding to loading rates in terms of $K_Y$ of $1$ to $1.5 \cdot 10^6 \text{ MPa}\sqrt{\text{m}}/\text{s}$ [16] with CT-specimens of 15 mm thickness containing 20% side grooves. The validity of the test results was evaluated on the basis of the existing standards [1, 9, 14] using the dynamic yield strength of the material determined with smooth round tensile bars under equivalent strain rate as calculated for the edge of the plastic zone in the CT specimens.

In Fig. 9 dynamic fracture toughness data of a RPV material 20 MnMoNi 55 (KS 17) with high upper shelf toughness (USE = 200 J) are compared with data obtained from quasi static testing. All tests were performed using CT specimens. The dynamic fracture toughness data are at the lower end of the quasi-static values or even lower. The fracture toughness decreases further with increasing load rate which has the global effect of a shift in fracture toughness to higher temperature. In the linear elastic regime, the dynamic testing yields higher fracture toughness data compared with the $K_{Ia}$-curve. This provides validation of the $K_{Ia}$ curve as a lower bound in this regime.
2.5 Evaluation of Crack Arrest Parameters

The crack arrest toughness is the plain strain elastic stress intensity at the arrest point. It can be determined by means of modified compact specimens, wide plates and rotating discs. The use of transversely wedge loaded compact specimens has proven to be the simplest and most favoured method, Fig. 10. The crack in the specimen is usually initiated in a brittle weld introduced at the location of the notch. In high strength material the notch can be produced directly by means of electric discharge machining (EDM) of the specimens, which simplifies considerably the specimen preparation. The test requirement and the evaluation the $K_{Ia}$ value is described in ASTM E 1221-88 [17]. The crack arrest toughness is calculated on the basis of the crack tip opening displacement and the crack length at arrest for the given specimen dimensions. However, the $K_{Ia}$ values are only valid if plain strain conditions and linear elastic material behaviour during the phase of crack extension is assured.

In Fig. 11, results of static ($K_{Ic}$) and dynamic ($K_{Id}$) fracture toughness values are compared with crack arrest toughness ($K_{Ia}$) and Charpy-V-notch energy as a function of temperature for the high toughness material 20 MnMoNi 55. According to the validity criteria the fracture toughness values do not encompass the temperature range of the entire Charpy curve but only that up to the transition temperature regime. In the lower shelf regime of Charpy energy the $K_{Id}$ values lie invariably below the $K_{Ic}$ and $K_{IJ}$ values, respectively. The $K_{Ia}$ values are in the main below the scatter band of each $K_{Ic}$, $K_{IJ}$ and $K_{Id}$. Due to the wide scatter of the results, a definite correlation has not be established between the different toughness values.
A summary of the results discussed above is shown in Fig. 12. Where the lower limit curves of the $K_{Ic}$ or $K_{IJ}$ scatter bands taken from figure 11 and additionally tested materials, together with the $K_{IR}$ reference curve, the lower limit for $K_{Id}$ and $K_{Ia}$ values and the $K_{Ic}$ curve as the lower limit for initiation values are related to the Nil Ductility Transition Temperature $T_{NDT}$. This representation clearly demonstrates that within the validity range of linear-elastic fracture mechanics the lower limit curves of all materials fall above the $K_{Ic}$ curve. Thus the $K_{Ic}$ curve can be used as a conservative approximation. In the upper transition range and on the upper shelf, only the $K_{IJ}$ values of 15 MnNi 6 3 exceed the limit of 220 MPa $\sqrt{\pi}$ specified in the Codes [5, 6, 13].

For the "other materials" it can, therefore, be concluded that for the upper shelf the $K_{Ic}$ curve provides a non-conservative estimation of the actual fracture toughness derived from the initiation value.

A comparison of $K_{Id}$ and $K_{Ia}$ values with the $K_{IR}$ reference curve is shown in Fig. 13. All values, except those of the low upper shelf energy of 40 J material 22 NiMoCr 3 7 (modified), manufactured for research purpose only, exceed the $K_{IR}$ curve. This confirms the description of the $K_{IR}$ curve as the lower envelope of the fracture toughness values (static and dynamic) - at least on the lower shelf and in the transition regime of the $C_v$-T curve.

Contrarily the lower limit curve of the $K_{Ia}$ band of the low shelf material 22 NiMoCr 3 7 (modified) increases slightly with rising temperature and intersects the $K_{IR}$ curve at approx. 40 K above $T_{NDT}$ (= 70°C). This makes the conservative nature of the reference curve doubtful. Similar results are available for a second high-strength, low-toughness model material [18]. The results do not allow conclusion to be drawn about crack arrest behaviour on the upper shelf.
3 Transferability of Fracture Mechanics Properties from Small Scale Specimens to Components

The standard test conditions and evaluation procedures (compare with section 2 above) provide fracture toughness data which is transferable to any structure - regardless of size and dimensions - as long as the validity criteria are fulfilled. This has been proven in the linear elastic fracture mechanics regime for $K_{lc}$ [16], with the exception of the validity limitation depending on the size criteria for plain strain conditions (compare 2.1 and 3.2 above).

For the elastic-plastic fracture mechanics regime it has to be demonstrated that the physical initiation process does indeed begin at the level of $J_1$ which was found to be size independent in case of CT specimens. Also a correlation has to be established between crack resistance, as given by the $J_R$ curve, and the specimen geometry and size.

3.1 Evaluation of Fracture Mechanics Characteristics on Large Scale Specimens

A large worldwide effort has been undertaken in order to develop transferability criteria in the elastic-plastic fracture mechanics regime. One of those projects was the research program "Integrity of Components" in Germany [19]. Tests were carried out with large scale specimens made of fine grained structural steel of different quality covering high and extremely low Charpy upper shelf toughness. The crack initiation values and the crack resistance curves were compared with those from small scale specimens. The variety of specimens tested is shown in Fig. 14, it comprises CT specimens with a thickness up to 200 mm, single edge cracked tensile (SECT) specimens, double edge cracked tensile (DECT) specimens, centre cracked tensile (CCT) and three point bend (TPB) specimens with a width (B) ranging from 100 to
600 mm and a thickness (W) of up to 200 mm. The $J_R$ curves experimentally determined with different large scale specimens are shown in Fig. 15a and b in comparison with the $J_R$ curves evaluated from CT-specimens. The specimens in Fig. 15a were made of a fine grained structural steel 22 NiMoCr 3 7 (KS 01) with an upper shelf energy of about 90 J, the specimens in Fig. 15b were made of the modified low toughness material 22 NiMoCr 3 7 (KS 07) with a Charpy upper shelf energy of 40 J. The same crack initiation value $J_i$ could be evaluated on all specimens as defined in section 2.3 above (converted to $K_{ij}$) within a reasonable scatter band as usual for this type of test. Moreover, the data corresponded with that derived from small scale specimens (CT-25 mm, 20% side grooved). This clearly demonstrates $J_i$ to be a material property, not depending on size and geometry and thus a basis for application to large and complex structures and components. A biaxial stress state exists on traction free surfaces as it is at the crack tip. In case of compressive stresses resulting e.g. from internal pressure, yielding at the crack tip is promoted by the triaxial stress state – with one compressive principal stress component.

With regard to crack resistance (process of stable crack growth), however, there is a strong size and geometry effect [20] which, additionally, seems to depend on the material toughness. Tests performed at the onset of upper shelf show consistence in crack initiation $J_i$ but not in the course of the crack resistance curve. This is due to the distinct differences in the degree of multiaxiality of the stress state as they are associated with the selected specimen geometries and sizes. The degree of multiaxialty can be quantified by the equation

$$ q = \frac{1}{\sqrt{3}} \frac{\sigma_e}{\sigma_m} $$
q is the degree of multiaxiality [21]  
\( \sigma_e \) equivalent stress according to von Mises  
\( \sigma_m \) mean stress \( \frac{1}{3} (\sigma_1 + \sigma_2 + \sigma_3) \)

The lower the value of \( q \), the higher the degree of multiaxiality. For the hydrostatic stress state (where \( q = 0 \)) no plastic deformation (except void formation) can occur even in high toughness material.

The degree of multiaxiality is not constant across the specimen ligament and depends on the relative crack length \( (a/W) \) as shown for a DENT specimen in Fig. 16. In case of a modest crack length \( (a/W = 0.5) \), \( q \) is a minimum in front of the crack tip and increases across the ligament. For deeply cracked structures \( (a/W = 0.8) \) \( q \) remains nearly constant across the ligament at a relatively low level - which indicates a high degree of multiaxiality.

The strong influence of geometric parameters on crack resistance can also be evaluated from tests performed on pipes in Japan [22] and on CCT specimens in the USA [23]. The pipe tests (circumferential slit, \( a/W = 1 \)) were carried out under additional bending loads with systems of different stiffness. Although detailed fracture mechanics analyses are not yet available, the test results demonstrate the general influence of the geometry on the crack resistance curve \( J_R \), Fig. 17. Under extreme conditions spontaneous fracture can occur in the component without any detectable stable crack growth as in the case of double edge notched tensile specimens with a crack length ratio of \( a/W = 0.8 \), Fig. 18.

The influence of material toughness on both the absolute value and the variation of \( q \) across the ligament can be demonstrated on materials with different Charpy upper shelf energy. After a certain amount of plastic deformation material separation occurs initially in the area where \( q \) reaches the critical value. In structures with \( q \) values close to critical value \( q_c \)
in a large part of the ligament, stable crack growth cannot be expected. This was the case for the DENT specimen (compare Fig. 18) where unstable crack growth (spontaneous fracture) occurred at the crack initiation value $J_1$. Structures of high toughness material in terms of Charpy upper shelf energy can mitigate the multiaxiality of the stress field by a greater amount of plastic deformation or void formation. This results in higher $q$ values and thus reduces the risk of unstable crack extension causing brittle failure.

The $q$-value indicates whether or not stable crack extension is possible at given temperature. However, it provides, at present, no basis to transform $J_R$ curves of a CT specimen to other specimen geometries or structures.

3.2 Application of Fracture Mechanics Concepts for Thermal Shock Experiments

In case of loss of cooling accidents (LOCA) water is injected into the RPV at low temperature to cool the nuclear core. This leads to transient conditions causing high thermal stresses in addition to the stresses resulting from internal pressure (pressurized thermal shock, PTS). In order to describe the behaviour of the RPV under those conditions much experimental and theoretical research work has been carried out to validate the fracture mechanics concepts. The investigations not only focused on crack initiation and crack extension but also on crack arrest. They included wide plates with thermal gradients and superimposed axial load, nozzles in large vessels and hollow cylinders under internal pressure and thermal gradients.

Wide plate tests were performed with materials having a toughness gradient across the plate thickness to simulate the effect of neutron irradiation [24]. The wide plates contained surface fatigue cracks in the less tough material and were subjected to asymmetric thermal shock loading. The external
load was applied as axial tension or 3 point bending. The aim of the tests was to demonstrate that even in materials, the toughness of which corresponds to the state at the end of design life time (DLT), brittle crack extension did not occur under PTS loading conditions. The test conditions were typical of the cylindrical wall and the nozzle corner region in the upper shelf toughness and the ductile/brittle transition regime.

Investigations on different sized hollow cylinders were carried out Fig. 19. Within the Heavy Section Steel Technology Program (HSST) tests with regard to thermal shock loading were performed at the Oak Ridge National Laboratory (USA) already in 1973 [25].

The first series of tests were loaded by thermal stresses only without internal pressure (Thermal Shock Experiments, TSE 1-6). These conditions were achieved by submerging an open hollow cylinder into liquid nitrogen after it had been heated homogeneously up to 100°C. To build up sufficient thermal stresses the cylinders contained long axial flaws with one exception (TSE-2) which was prepared with a semi-elliptical flaw. All failures occurred in the linear elastic fracture mechanics regime. Crack initiation and crack arrest was in accordance with the predictions derived from lower bound fracture toughness values of small scale specimens. The tests gave no indication that dynamic fracture processes played any role. On the basis of the lower bound fracture toughness curves, presented in the ASME Code, all test results could be described in a conservative way, Fig. 20.

In the Federal Republic of Germany, thermal shock experiments were carried out with thick walled hollow cylinders (emergency cooling simulation programme NKS) at MPA Stuttgart and on a full size vessel at the HDR plant (decommissioned superheated steam reactor). The hollow cylinders were fabricated from different steel qualities with Charpy upper shelf energy.
ranging from 200 J to 40 J. The low shelf material had a nil ductility transition temperature - evaluated from Charpy energy/temperature curve - of about 250°C. Internal pressure and external axial load was applied on the hollow cylinder (O.D. = 800 mm I.D. = 400 mm) while the inner surface was cooled down rapidly from 300°C to room temperature. The loading conditions during the transient for the test specimen which contained an inner surface circumferential crack could be described by means of the J-Integral - as shown in the left part of Fig. 21. After a cooling time of about 7 min, the maximum J-value of about 430 N/mm was reached in this experiment (NKS-3). The stress and deformation analysis was carried out axial-symmetrically taking non-linear material behaviour into account. Calculations have shown that the degree of multiaxiality in the test specimen was of the same order as the one in the CT specimen. Therefore the $J_R$ curve of a corresponding temperature which is plotted in the right part of Fig. 21 can be used for stable crack growth assessment. On the fracture surface a stable crack extension of 3.6 mm could be measured, Fig. 22, which is in good agreement with the calculated value of 3.5 mm as shown in Fig. 20 and thus validates the fracture mechanics concept and the applied FE analysis [26].

In reality, the cooling of the vessel during LOCA is not symmetrical for the vessel but occurs locally being concentrated at the nozzle corner and the cylindrical wall in the form of strip-cooling. The phenomena associated with this loading condition was investigated at the HDR pressure vessel by introducing "guided" cooling. FE analyses have indicated a more severe situation for a circumferential crack than for an axial one. Therefore circumferential cracks were produced in the RPV wall by milling, with subsequent fatiguing by cyclic thermal shock loading. In addition, cracks were produced in the nozzle corner region by cyclic thermal shock and then subjected to rapid cooling under internal pressure (PTS). The maximum crack depth for both locations, cylindrical wall and
nozzle corner, was approximately 15 mm. Within the
time range investigated the J-integral increased steadily with
time for the cracks in the cylindrical wall, but dropped after
having reached the maximum in a short time for the nozzle
corner crack, Fig. 23. The principal differences in loading
condition result from axi-symmetric cooling in the nozzle area
and non-symmetric cooling in the cylindrical wall [27].

According to the $J_i$ values represented by the scatter bands
for different circumferential specimen orientations
converesponding to the crack growth directions (T-S nozzle
crack, L-S crack in cylinder) plotted in Fig. 23, crack
initiation and a certain amount of stable crack growth should
have occurred for the crack in the cylindrical wall.
Fractographic investigations, however, have not given any
indication of stable crack growth. Only in a few areas could
signs of incipient stretched zones at the fatigue crack tip be
identified. The discrepancy between prediction of stable crack
growth by the calculation and the absence of crack extension
in the test must be referred to the extensive crack branching
and thus to a relief in stress intensity, Fig. 24 [27].

Comparable thermal shock experiments have been conducted at
the European Joint Research Center (JRC) in Ispra, Italy on
nozzle corner cracks [28]. In the United Kingdom thermal shock
experiments were performed on thick walled hollow cylinders
spinning at a high revolution rate with a longitudinal crack
along the whole length of the inner surface [29].

From the results of the wide-plate experiments it can be
concluded that the transferability of data is valid even for
complex structures and loading conditions. The application of
the crack resistance curve to other specimens and structures,
is, however, only possible when the stress states are
comparable.
3.3 Crack Arrest Behaviour

The main feature of crack arrest studies with CT-specimens, as described in section 2.5 above, is that the crack grows into a decreasing K-field. Arrest in an increasing K-field is only possible when the crack initiates in a low toughness material and extends towards a region of higher toughness. This situation corresponds to a neutron irradiated vessel, assuming a crack is initiated at the inner surface and penetrates towards the outside. The necessary toughness gradients can be obtained in the test specimen either through a temperature gradient across a specimen section or by the use of a composite specimen [24, 30]. Most commonly used are tests where the toughness gradient is generated by a stationary thermal gradient in a single edge cracked wide plate specimen and the loading is applied by axial tension. Crack arrest toughness values of different materials determined under different test conditions, e.g. thermal shock (TSE) and pressurized thermal shock experiments (PTSE) on vessels in USA [25] and in France (FTSE) [31], wide plate tests in Japan (ESSO) [24], in USA (WP-1, WP-2) and Germany (GP-1) were found to be all enveloped by the crack arrest curve from the Codes, Fig. 25.

High crack arrest toughness values of up to 400 MPa\(\sqrt{m}\) result from the American wide plate tests (WP) and the Japanese ESSO tests. The crack arrest values derived from the thermal shock vessel experiments are generally lower than those from wide plates but they are still higher than the upper limit according to the Codes. The lowest crack arrest toughness values were obtained in the French thermal shock experiments (FTSE) which were carried out at a temperature of 20 K above \(RT_{NDT}\). The results of the German wide plate experiments and the rotating disc experiment fall within the scatter band of all the other results [30]. Contrary to the other results, a kind of upper bound toughness tending towards 200 MPa\(\sqrt{m}\) can be observed from the wide plate test GP 1 (see Fig. 25). The test
material KS 22 is a low toughness material (USE = 40 J) with high transition temperature (-250°C) which was produced for research purposes. Similar results on the same materials were also obtained from CT specimens, Fig. 26 [18].

At present, there is no ready explanation for the extremely high arrest values determined for some of the wide plate tests which are higher than the crack initiation values of materials with high upper shelf Charpy energy. Conclusions on the transferability of crack arrest results in the elastic-plastic regime cannot yet be drawn.

3.4 Summary of Transferability

To summarize the present state on fracture mechanics characteristics. Lower limit curves for $K_{IC}$ and $K_{IJ}$ derived from $J_1$ have been compared with the Code $K_{IC}$, $K_{IA}$ and $K_{IR}$ curves. Within the linear-elastic regime, the Code curves cover the experimental results for all the investigated materials in a conservative way. However, in the Charpy upper transition range and the upper shelf regime the experimentally determined data intersect the $K_{IC}$, $K_{IA}$ and $K_{IR}$ curves depending on the toughness of the material. It therefore has to be concluded, that the $K_{IC}$ curve is not conservative for all materials with respect to crack initiation on the upper shelf.

4 Correlation of Fracture Mechanics Properties with Charpy V-Notch Energy

In general fracture toughness data are not available for actual material of a reactor pressure vessel. From surveillance programmes, however, Charpy-V notch energy data are determined with the aim of using that data for a quantitative safety analysis based on fracture mechanics.
considerations. To evaluate the effects of size and multiaxiality a correlation between Charpy transition regime and Nil Ductility Transition Temperature can be derived on an empirical basis. In the upper shelf regime to obtain a correlation between the dynamically absorbed energy to fracture and the quasi-statically evaluated J initiation value \((J_i)\) is much more difficult.

Regardless of any physical solution, a correlation between Charpy energy and initiation fracture toughness has been established on an empirical basis. From statistical evaluation of 200 different melts of 22 different ferritic materials a tangent hyperbolic \((\tanh)\) fit was used to convert Charpy data into fracture toughness data covering the complete temperature range \([32]\). The evaluation of a A 508 Cl 2 steel shows good agreement with measured fracture toughness values up to the transition regime within a confidence limit of 95%, Fig. 27. Major differences occur in the upper shelf regime. More correlations between static and dynamic fracture toughness values and Charpy test results are summarized in Ref. \([33]\).

From experimentally determined \(J_i\) values and the corresponding upper shelf Charpy energy \((USE)\), a statistical correlation between \(J_i\) and USE has been established, Fig. 28 \([34]\). All experimental \(J_i\) data falls beyond the curve \(J_i - 2\sigma\) with a probability of 97.73%. In addition another correlation is shown in this figure, which takes besides the Charpy energy the mean flow stress \(\sigma_{fl}\) and stable crack extension into account \([35]\). This curve is similar to the one mentioned above and falls between the two curves \(J_i - \sigma\) and \(J_i - 2\sigma\). On the basis of this correlation a complete \(J_R\) curve can be derived in the elastic-plastic fracture mechanics regime from Charpy upper shelf energy. In the correlation the two discrete data for \(\Delta a = 0.1\) and 0.2 mm were selected (see Fig. 25) because \(\Delta a = 0.1\) mm matches quite well with the stretched zone of a highly tough material whereas \(\Delta a = 0.2\) mm represents the region of the
technical material characteristics on the $J_R$ curve according to ASTM E 813-88 and EGF P 1-90.

5 Summary

Since the transferability of the $J_R$ curves is not allowed in general as described a comparison of calculated $J_R$ curves with experimental data is not considered here.

The fracture mechanics limit curves given in the main Codes for initiation $K_{ic}$, crack arrest $K_{ia}$ and the reference curve $K_{IR}$, covering crack arrest as well as dynamic fracture mechanics data, are conservative when compared with all test results, if linear elastic (plain strain) conditions can be assumed. In the transition regime, and on the upper shelf - depending on the material involved - these curves exceed the actual fracture toughness. Thus, predictions based on these curves can be non-conservative in these toughness regimes.

The effective crack initiation values $J_i$ are independent of the size and geometry of the specimen or component at given temperatures and loading rates and, therefore, are material properties transferable to components. However, this does not apply to the crack resistance curves which strongly depend on the degree of multiaxiality of the stress state. With increasing material toughness not only the effective crack initiation value $J_i$ but also the tearing resistance against stable crack growth will increase as well. This holds also for complex loading conditions (pressurized thermal shock) and complex component shape (e.g. nozzle corner crack).


[4] ASME Boiler and Pressure Vessel Code, Section XI, Division 1, Appendix A, 1988


Fig. 1: Experimentally determined $K_{lc}$ values evaluated according to ASTM 399 in comparison with different validity criteria for thickness.

Fig. 2: Lower bound fracture toughness curves as a function of temperature relative to the "Nil Ductility Reference Temperature" $RT_{NDT}$.

Fig. 3: Mean curve, standard deviation and mean deviation of Charpy-V notch test results performed in different laboratories.

Fig. 4: Mean curve, standard deviation and mean deviation of lateral expansion from tests shown in Fig. 3.

Fig. 5: Nil Ductility Transition Temperature (NDT) determined from drop-weight tests in different laboratories.

Fig. 6: Evaluation of stable crack initiation values according to different standards and methods; material 20 MnMoNi 5 5, USE = 200 J.

Fig. 7: Method to determine crack initiation parameter $J_I$.

Fig. 8: Comparison of different crack initiation values on the load traces:
   a) load / COD,
   b) load / $\Delta a$,
   c) $J$ / COD,
   d) $J$ / $\Delta a$.

Fig. 9: Comparison of fracture toughness data obtained from quasi-static and dynamic tests.

Fig. 10: Schematic view of a wedge loaded CT specimen to determine crack arrest toughness data.

Fig. 11: Static and dynamic fracture toughness ($K_{IC}$, $K_{IJ}$, $K_{Id}$), crack arrest toughness ($K_{Ia}$) and Charpy-V notch energy ($C_V$) as a function of temperature for RPV Steel 20 MnMoNi 5 5 (KS 17).
Fig. 12: Lower bound fracture toughness curves of different materials relative to NDT-temperature in comparison with the corresponding reference curves ($K_{lc}$ and $K_{IR}$).

Fig. 13: Dynamic fracture toughness ($K_{Id}$) crack arrest toughness ($K_{Ia}$) relative to NDT-temperature for different materials.

Fig. 14: Fracture mechanics specimens tested for investigation of transferability criteria.

Fig. 15a: $J_R$ curves of large scale specimens of different size and geometry; material 22 NiMoCr 3 7 (USE 90 J), specimen dimensions:
- DECT: $B = 300, W = 100$
- SECT: $B = 300, W = 200$

Fig. 15b: $J_R$ curves of large scale specimens of different size and geometry; material 22 NiMoCr 3 7 (modified, USE 40 J),
- CCT: $B = 200, W = 300$
- TPB: $R = 500, W = 200$
- SECT: $B = 300, W = 200$
- DECT: $B = 250, W = 70$

Fig. 16: Change of multiaxiality $q$ across the ligament of double edge notched tensile (DENT) specimens with different crack length $a/W$.

Fig. 17: $J_R$ curves determined in pipe tests with different crack geometries and stiffness of the test set up.

Fig. 18: $J_R$ curves of low upper shelf double edge notched tension (DENT) specimens with different crack length.

Fig. 19: Test specimen, model vessel and full size vessel used for pressurized thermal shock (PTS) experiments.

Fig. 20: Fracture toughness values derived from thermal shock experiments in comparison with the ASME $K_{lc}$ and $K_{Ia}$ curve.
**Fig. 21:** Calculated J values (left hand side) during pressurized thermal shock (NKS-3) experiment in comparison with a $J_R$ curve determined with a CT 10 specimen (right hand side) to derive amount of stable crack growth during experiment.

**Fig. 22:** Crack depth in PTS specimen NKS-3 before and after test and average stable crack extension derived.

**Fig. 23:** Calculated J-values as a function of time during the HDR thermal shock experiment.

**Fig. 24:** Crack configuration in the cylindrical wall of the HDR vessel.

**Fig. 25:** Crack arrest toughness data determined with large scale specimens in comparison with the ASME $K_{Ia}$ curve.

**Fig. 26:** Crack arrest toughness data as a function of temperature for materials with different upper shelf energy in comparison with the ASME Reference Curve ($K_{IR}$)

(20 MnMoNi 5 5 USE = 200 J; 22 NiMoCr 3 7 modified USE = 40 J; 17 MoV 8 4, USE = 40 J)

**Fig. 27:** Fracture toughness data converted from Charpy-V notch energy in comparison with experimental $K_{IC}$ data of a A 508 Cl 2 steel.

**Fig. 28:** Correlation of crack initiation values ($J_1$) with Charpy upper shelf energy.
limits of validity for CT-specimen

\[ B \geq \omega \left( \frac{K_{lc}}{R_0} \right)^2 \]

- 20 MnMoNi 55
- \( T_{NDT} = -20^\circ C \)
- \( T = -100^\circ C \)

- experimental values

- ASTM E 399: \( \omega \geq 2.5 \)

\( \omega - \text{values} \)
- \( \omega = 2.5 \)
- \( \omega = 5 \)
- \( \omega = 10 \)
- \( \omega = 20 \)
- \( \omega = 30 \)

FIGURE 1
FIGURE 2

Graph showing the fracture toughness of KTA 3201.2, ASME XI, KTA 3201.2, ASME II, and KIR KTA 3201.2, ASME III materials.
FIGURE 3

round robin test KS13
notched bar impact energy
mean value curve (8 participants)

- ductile fracture
- mixed fracture
- brittle fracture

standard deviation
mean deviation

temperature

$C_v$

200 150 100 50 0

-100 -50 0 RT 50 100 150 200 250 300 350 °C
FIGURE 4

- standard deviation
- mean deviation

lateral extension

round robin test KS13
lateral extension
mean value curve (8 participants)
• ductile fracture
• mixed fracture
• brittle fracture

temperature

0,4
0
0,2
0,1
0
-0,1
-0,2
-0,3
-0,4

mm
MATERIAL KS13

A - L = PARTICIPANT

FIGURE 5
CT-specimen KS01 BM62

$J - \Delta a$

FIGURE 6

$J_1$

$J_{ic}$

$J_{0.2/\text{bl}}$

$J_{0.2}$

ASTM E813-88

EGF-P1-90

EGF-P1-90, JSME S001-81
FIGURE 8
FIGURE 10
lower bound KS 07 A/B, WB 36, KS 17, 15 MnNi 6 3 compared to ASME reference curves

FIGURE 12
Kla/Kld KS 07 A/B, WB 36, KS 17, 15 MnNi 6 3 compared to ASME reference curves

FIGURE 13
FIGURE 15 a

- J - integral / N/mm
- crack extension Δ a / mm

exp.blunting lines
DECT α/W = 0.5
SECT α/W = 0.5
CT 25
20%sg.
CT 150
CT 100
J_i
\[ J_{\text{integral}} / \text{N/mm} \]

\[ J_{0.2/\text{bl}} \]

\[ J_i \]

\[ \Delta a / \text{mm} \]

FIGURE 15 b
FIGURE 16
FIGURE 17

J - integral / MN/mm

crack growth $\Delta a_m$ / mm

- 239 -
CT25, 20\% sq

DENT $\alpha/W = 0.25$

DENT $\alpha/W = 0.8$

unstable crack extension

$J_t$, notched

$K_S07$

$USE = 40 \, \text{J}$

FIGURE 18
Wall thickness $s$

$s = 148\text{mm}$

long. crack

$s = 200\text{mm}$

circ. f. crack

![Diagram showing ORNL, MPA, and HDR with wall thicknesses and dimensions.](image-url)
FIGURE 20
average measured crack extension (3.6 mm)

average initial crack depth 62.8 mm

ultrasonic crack depth measurement

O = before test
△ = after test

fractographic crack depth measurement

● = before test
■ = after test
crack in HDR-RPV

![Graph showing J-integral over time with phases labeled L-T, L-S 110°C and T-L, T-S 110°C, and different crack locations marked on the graph.]

FIGURE 23
**Typical Branching of the Cracks**

**Section A from B9 S1**

**Section from HDR trepan B9**

**Section of a Crack in the RPV-Cylinder**

**FIGURE 24**
FIGURE 25
FIGURE 26

- Crack arrest toughness $K_a / \text{ksi} \sqrt{\text{in}}$
- $T - T_{NDT} / ^\circ C$
- $K_{IR}, \text{ASME III}$

Symbols:
- □ = 20 MnMoNi 5.5
- ○ = 22 NiMoCr 3.7 (mod.)
- Δ = 17 MoV 8.4
TANH-CORRELATION

O - mean values
+ - 99% confidence limit

--- ASME XI ($K_{lc}$ curve)
$T_{NDT} = 120 \, ^{\circ}C$

● measured values
\[ /34/ = f(C_v) \]
\[ /35/ = f(C_v, \Delta \theta, \sigma_{II}) \]

![Graph showing notched bar impact energy vs. notched bar impact energy (upper shelf)](image)
Chapter 6

NEUTRON EXPOSURE

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1. GENERAL ASPECTS

In the evaluation of neutron embrittlement of light water reactor pressure vessel (LWR-PV) steel, it is necessary to know the neutron exposure of the steel in order to interpret the resulting changes in mechanical properties. In this chapter we discuss LWR-PV neutron transport calculations and dosimetry methods, and how they are combined to evaluate neutron exposure. First, we review the history of the development of these approaches.

1.1 History

In early work on the neutron embrittlement of pressure vessel steels [1], there were very large uncertainties in the evaluation of neutron exposure. It became clear [2] that the best experimental approach for reducing the uncertainties was activation dosimetry (sometimes referred to as radiometric dosimetry). In this approach a set of foils or wires of different chemical elements or isotopes is irradiated. Each one undergoes activation in response to a different range of the neutron energy spectrum [3-5]. The spectrum can then be evaluated from the measured activities of the foils, if the activation cross sections are known.

Activation dosimetry by itself, however, was not able to give a very detailed picture of the neutron spectrum, principally because of the lack of availability of a large enough set of suitable monitors that responded to different parts of the neutron energy range. Nor could activation dosimetry provide a knowledge of neutron exposure at locations other than the dosimetry locations.
Fortunately the problem could also be approached theoretically by solving the energy-dependent Boltzmann transport equation. To do this in a practicable manner, the development of new calculational methods, high speed computer hardware, and complex computer codes was necessary. Evaluated neutron scattering and absorption cross sections were also needed. As these developments proceeded in the early sixties, it became possible to combine the results of neutron transport calculations and dosimetry measurements for the same reactor location and achieve the "best fit" neutron flux and spectrum by iteration [6]. The transport calculations could then supply results for other locations. However, different workers used different cross section libraries [7], making it difficult to compare results from one reactor to another, and there were still significant uncertainties in the results.

In 1966 the Cross Section Evaluation Working Group (CSEWG) was formed in the U.S. in order to coordinate the evaluation of nuclear data, originally for the fast breeder reactor program, and later, for a variety of purposes. This group has produced six versions of the Evaluated Nuclear Data File (ENDF/B) to date, the most recent being ENDF/B-VI. Dosimetry files have been included since ENDF/B-IV [8]. Evaluated cross section files have also been developed in Europe, Japan, and the USSR, and are called JEF, JENDL, and BROND, respectively [9]. A major advance was the inclusion of error files, beginning in the late seventies. Extensive cross section sensitivity and error studies on parameters related to LWR-PV damage analysis became feasible [10]. These efforts have led to great improvements in the cross section information available, and to more standardization between laboratories.

In 1971, the Interlaboratory Liquid Metal Fast Breeder Reactor Reaction Rate (ILRR) program was initiated by the U.S. Atomic Energy Commission to develop a capability to determine consistent and reliable experimental values for reaction rates in various well-established and permanent neutron fields [11]. Cooperation was established with the U.S. National Bureau of Standards and the Centre d'Etude de l'Energie Nucléaire-Studiecentrum Voor Kernenergie in Mol, Belgium. This program provided excellent quality control for dosimetry in breeder reactor development.

In 1975, recognizing the need for further improvement and for international cooperation in the dosimetry field, the American Society for Testing and Materials (ASTM) Committee E-10 (now called Nuclear Technology and Applications), together with the EURATOM Working Group on Reactor Dosimetry, began sponsoring symposia on reactor dosimetry. Seven of these symposia have now been held, and
the proceedings have been published [12-18]. These proceedings detail the progress that has been made internationally in reactor dosimetry research and development.

In 1977 the U.S. Nuclear Regulatory Commission established the Light Water Reactor Pressure Vessel Surveillance Dosimetry Improvement Program. Its purposes are "to improve, maintain, and standardize neutron dosimetry, damage correlation, and the associated reactor analysis procedures used for predicting the integrated effect of neutron exposure to LWR pressure vessels [19]." Cooperative links have been established between U.S. and European laboratories. Benchmark neutron facilities have been set up, including PCA, VENUS, and NESDIP [16-17], in which radial neutron penetration, variation of the reactor core fission neutron source, and the reactor cavity environment were studied, respectively, and comparative experiments have been performed.

In 1980 the IAEA Nuclear Data Section organized an international calculational exercise to determine the state of the art of data treatment in reactor neutron metrology. It has been conducted in three rounds, called REAL-80, REAL-84, and REAL-88. REAL stands for "Reaction Rate Estimates, Evaluated by Adjustment Analysis in Leading Laboratories." This exercise has been helpful in revealing problems in unfolding calculations [20].

ASTM Committee E-10 has developed standard methods for pressure vessel dosimetry, contained in the Annual Book of ASTM Standards [21]. Standard methods have also been developed in Germany, as DIN 25456 [22].

In recent years the Electric Power Research Institute in the U.S. has sponsored the development of the LEPRICON computer code system, which takes careful account of uncertainties in cross sections, dosimetry measurements, and transport calculations in terms of covariances and bias factors [23].

As a result of these efforts, it is now possible to evaluate integrated neutron exposure (fluence of neutrons having energy greater than 1 MeV) with 1σ uncertainties as low as 10% for power reactors [24] and in the range of 5 to 10% for mockup experiments. Maintenance of uncertainties at levels this low depends on continued attention to benchmarking, standardization of methods, "round robin" radiation counting comparisons, the complementary use of transport calculations and
dosimetry, and rigorous accounting for uncertainties in all aspects of the process of evaluating neutron exposure.

1.2 Definitions of Important Concepts

In light water reactors the neutrons originate primarily from the fission of $^{235}\text{U}$ by thermal neutrons, although thermal fission of $^{239}\text{Pu}$ becomes significant as fuel burn-up proceeds. The energy spectrum of the neutrons produced in $^{235}\text{U}$ thermal fission has been described by Watt [25] and by Cranberg [26]. About 2.4 neutrons are produced per $^{235}\text{U}$ fission, compared to about 2.8 for $^{239}\text{Pu}$. Their average energy is about 2 MeV for both nuclides.

The flux density or simply "flux" of neutrons is defined as the number of neutrons per unit time that pass in all directions through an imaginary sphere of unit cross sectional area centered on the point in question. Flux is represented by the symbol $\phi$, and its units are neutrons/m$^2$-s.

The fluence, $\Phi$, expressed in neutrons/m$^2$, is the time integral of the flux. The energy spectrum of the neutrons is most usefully plotted in terms of the flux per unit lethargy.

Lethargy($u$) is defined by

$$u = \ln \left( \frac{E_{\text{max}}}{E} \right)$$

where $E_{\text{max}}$ is usually taken to be 10 MeV. By plotting the flux per unit lethargy on the ordinate versus the neutron energy on the abscissa of a log-log plot, one obtains a graph in which the number of neutrons in each part of the spectrum is proportional to the area under the curve, and the detailed shape of the spectrum can be seen.

The wide energy spectrum of the neutrons in a reactor results from interaction of the original fission neutrons with nuclei of the materials in the reactor. The principal interactions in an LWR are elastic scattering ($n,n$), inelastic scattering ($n,n'\gamma$), radiative capture ($n,\gamma$), and fission ($n,f$), the first two yielding a neutron of lower energy than that of the incident neutron.
The rate of neutron interactions is expressed in terms of the cross section (\(\sigma\)) for the particular reaction:

\[
\frac{dn^*}{dt} = N\phi\sigma
\]

where \(\frac{dn^*}{dt}\) is the number of interactions occurring per unit volume and per unit time, \(N\) is the number of target nuclei per unit volume, \(\phi\) is the neutron flux, and \(\sigma\) is normally expressed in barns (= 10^{-28} m^2).

The total cross section is the sum of the partial reaction cross sections for all of the interactions that a neutron of a particular energy can undergo in a particular material:

\[
\sigma_{\text{TOT}} = \sigma_{\text{ELAS}} + \sigma_{\text{INELAS}} + \sigma_{(n,\gamma)} + \text{etc.}
\]

The mean free path is the average total distance that a neutron travels before interacting:

\[
\lambda(m) = \frac{1}{N(m^{-3})\sigma_{\text{TOT}}(m^2)}
\]

The mean free paths of fission neutrons in light water or steel are typically a few centimeters. Thus, on the average, neutrons undergo several scatters in travelling from the reactor core to the pressure vessel. The cross sections for materials of interest in LWRs have been measured, evaluated, and tabulated in the nuclear data files available from the four nuclear data centers.

There are three main components in the neutron spectrum in the core of a LWR: the prompt fission neutrons (first-flight or unscattered), which have energies in the range of about 0.1 to about 15 MeV, the intermediate-energy or "slowing-down" neutrons, with energies between about 0.5 eV and about 0.1 MeV, and the thermal neutrons, having energies below about 0.5 eV. The fission neutrons can be described by the Watt or Cranberg curves as noted above. The intermediate-energy neutrons have an energy spectrum that is approximately proportional to 1/E, because the scattering cross sections are nearly constant with energy in the intermediate-energy range, absorption cross sections are small, the core is large compared to the mean
free path, and in each collision, a neutron loses nearly a constant fraction of the energy it has before the collision. The thermal neutron component is nearly in thermal equilibrium with the LWR core, and its energy spectrum is roughly approximated by the Maxwell-Boltzmann distribution. Deviations are caused by neutrons being added on the higher energy side by scattering down, and being removed preferentially on the lower energy side because absorption cross sections are proportional to $1/\sqrt{E}$.

As the neutrons diffuse from the core to the pressure vessel, their flux drops by some three or four orders of magnitude and their energy spectrum changes as a result of interactions with reactor materials. Figure 1 compares normalized neutron spectra in the core and in the pressure vessel of a BWR, down to 0.1 eV. These spectra were calculated using an $S_n$ transport code with 100 energy groups and $P_3S_8$ cross section expansion and quadrature. The structure in the spectra correspond to resonances in the absorption cross sections of uranium and iron, respectively.

When neutrons interact with solid materials such as pressure vessel steels, they produce nuclear transmutations and atomic displacements. Most of the atomic displacements are produced by fast neutron collisions with nuclei, producing primary knock-on atoms (PKAs), which in turn produce cascades of atomic displacements. Thermal neutrons are able to produce atomic displacements because of the recoil of the nucleus that results from gamma ray emission in radiative capture of neutrons, and also because the resulting gamma rays (having several MeVs of energy) can produce displaced atoms via Compton recoil electrons [28]. If the cross sections are known in detail for all possible neutron interactions with a material, it is possible to calculate the energy spectrum of the primary knock-on atoms from the neutron energy spectrum. From this, the number of atomic displacements that result from neutron interactions can be calculated, using the modified Kinchin-Pease (or Norgett, Robinson, and Torrens) [29] equation. This equation requires a knowledge of the energy required to displace an atom, and uses the Lindhard theory [30] for evaluating the fraction of the PKA energy that goes into atomic displacements. This calculation has been performed for iron and is the basis of the standard practice for calculating the displacements-per-atom or dpa [31]. Dpa is defined as the number of times, on the average, that an atom has been displaced during an irradiation. Calculation of dpa takes account of the fact that the number of displaced atoms that result from a neutron interaction depends on the energy of the neutron.
When a surveillance program is performed on a power reactor, the surveillance specimens are located between the reactor core and the inside wall of the pressure vessel. The rate of neutron exposure is therefore greater for the surveillance specimens than for the inner wall of the pressure vessel. The lead (pronounced "LEED") factor is defined as the ratio of the neutron flux density at the location of the specimens in a surveillance capsule to the neutron flux density at the reactor pressure vessel inside surface at the peak flux location. The neutron flux density is generally defined in terms of neutrons having energies above a cut-off energy such as 1 MeV in specifying the lead factor.

1.3 Neutron Exposure Parameters for Correlation with Damage

The search for the most appropriate neutron exposure parameter has gone on for a long time and has been the subject of some controversy. Initially, the technology for evaluating neutron exposure was very limited, so that there was little choice of parameters available. At present our evaluation capabilities are much improved, but the lack of a detailed understanding of the embrittlement process makes it difficult to specify the most relevant parameter to use for evaluating neutron exposure. It is well known that neutron exposure produces atomic displacements and transmutations in pressure vessel steels. It is not understood in detail how these basic processes translate into embrittlement of the steel.

Initially, very crude measures of neutron exposure were used, such as "megawatt-days per adjacent ton" (of uranium fuel) or specification only of the thermal neutron fluence. As dosimetry techniques improved and a greater appreciation of the effects of fast neutrons developed, neutron exposure came to be expressed in terms of the fluence of neutrons having energies above some cut-off energy, such as 0.1, 0.5, or 1.0 MeV. So long as comparisons were made between reactor locations or between different reactors in which the neutron energy spectra were similar, such measures were found empirically to correlate embrittlement data fairly well. They thus found their way into regulatory practice, where the fluence of neutrons with energy greater than 1.0 MeV is usually the parameter that is correlated with radiation damage in pressure vessel steels for Western reactors, while 0.5 MeV is used for VVER-type reactors.

As radiation damage theory developed, and concern grew about the importance of the shape of the neutron spectrum in determining the number of atomic
displacements, the concept of dpa was developed and in turn found its way into regulatory practice. Dpa was found to correlate property degradation data for the same material in different neutron environments better than did fluence (E>1.0 MeV) [32]. A recent informal international workshop adopted (with three dissensions out of approximately 40 participants) the consensus statement that "dpa is a useful exposure parameter, and it is clearly superior to the neutron fluence above 1 MeV for correlating embrittlement data obtained from irradiation experiments with different neutron energy spectra" [33].

The use of dpa is particularly indicated when comparing results from different reactors and in the analysis of through-vessel-wall embrittlement [34]. The "softening" of the neutron energy spectrum with radial depth within the vessel wall causes the relative decrease of dpa with depth to be smaller than the decrease with depth of fluence (E > 1 MeV). Exposure can be approximated within the vessel by means of an exponential function:

\[ f(r) = f(r_i) \cdot e^{-k \cdot r}, \]

where \( f (r_i) \) is the exposure value at the inner surface (r in cm). The exponent factor is about 0.13 cm\(^{-1}\) for fluences (E > 1.0 MeV) and in the range of 0.095 to 0.10 cm\(^{-1}\) for dpa, depending on reactor geometry. This means that the ratio of dpa to fluence (E > 1.0 MeV) can change by a factor of two in traversing from the inner to the outer PV-surface of a typical 1200 MW(e) PWR. Therefore conservative lead factors between surveillance and vessel positions should be based on dpa exposure.

In spite of the advantages of the dpa parameter compared to simply fluence (E > 1.0 MeV), few people in the radiation damage field believe that dpa is the ultimate, optimum neutron exposure parameter. One reason is that experimental electrical resistivity measurements and molecular dynamics computer modelling have both shown that dpa gives an overestimate of the true number of displacements (by about a factor of two for reactor neutrons in body centred cubic metals), and does not correctly account for the variation of displacement efficiency with primary knock-on atom recoil energy [35,36].

A second problem is that the number of atoms originally displaced is not likely to be a parameter of direct importance to embrittlement. The great majority of displaced atoms recombine with vacancies, leaving no lasting effects. Others form
clusters or diffuse as interstitial atoms. The number of "freely-migrating defects" (that is, vacancies and interstitial atoms that do not recombine or form clusters or become trapped) may be of significance, and this parameter has been found to be very sensitive to the recoil energy, with lower energy recoils producing a much greater fraction of free defects. In this connection a suggestion has also been made recently [28] about the possible importance of displacements produced by high-energy gamma rays resulting from low-energy neutron capture. These displacements are produced by Compton recoil electrons that result from gamma ray interaction with electrons, and the resulting atomic recoil energies are low. Thus, displacements produced by low-energy neutron interactions may have even more significance because of the resulting capture gamma ray displacements.

Another important point is that dpa takes account only of displacements. If transmutations play a major role, for example helium production due to high boron content in a particular steel, this would not be accounted for by dpa.

Finally, there have been questions about the possibility of flux dependence. Specification only of an integral parameter such as fluence or dpa does not take account of possible flux-dependent effects.

Until a better understanding of the embrittlement process is in hand, it will probably not be possible to make much progress in optimizing the neutron exposure parameter (or parameters). In this situation, the most prudent course of action is to evaluate the neutron exposure in as complete and detailed a manner as is feasible, so that the data collected can be converted to whatever parameter is found to be most useful in the future. In this way the value of the collected data will be preserved for use in future data bases.

With these considerations in mind, we recommend that the following parameters be specified when providing evaluations of neutron exposure for pressure vessel steel embrittlement work:

1. Full power neutron fluxes for locations of interest, and full power local flux changes during the fuel cycle for surveillance programmes, including thermal (<0.5 eV), epicadmium (>0.5 eV), and fast (both >0.1 MeV and >1.0 MeV), with uncertainties.
For VVER-type reactors, >0.5 MeV should also be included, because of conventional usage.

2. Fluences for the locations of interest (same energy ranges as in 1.) with uncertainties.

3. Displacements per atom (dpa) for locations of interest, with uncertainties.

4. Full power dpa rates for locations of interest, with uncertainties.

5. Listing and plots of complete neutron spectra for locations of interest, preferably shown as flux per unit lethargy versus energy in log-log format.

6. Spatial gradients of neutron flux (E>1.0 MeV), neutron fluence (E>1.0 MeV), and dpa throughout the array of metallurgical specimens in surveillance capsules, and in the pressure vessel.

2. CALCULATION OF NEUTRON EXPOSURE PARAMETERS

In analyzing radiation damage to a reactor pressure vessel, it is necessary to determine the detailed energy-dependent neutron flux distribution in the vessel as well as within surveillance capsules. For dosimetry comparison, estimates of the neutron spectrum and spectrum-averaged cross sections for the activation monitors must be calculated at various detector locations.

As noted in Section 1.1, the behavior of large numbers of neutrons in materials is described theoretically by the energy-dependent Boltzmann transport equation. This equation is based on conservation of neutrons and is written in terms of angular flux. The equation includes terms for source, translation, scattering, and absorption of neutrons. When applied to a nuclear power reactor, the Boltzmann equation is too complex for analytical solution. Accordingly, numerical methods such as discrete ordinates (or "Sn-method") [37] and Monte Carlo [38] are used.

The first calculation that must be performed is the determination of the neutron source distribution in the reactor core. The results of this calculation then serve as
input to the neutron transport calculation. The validity of these calculations depends on the accuracy of the knowledge of the materials and geometry of the reactor, and the quality of the nuclear data used, especially the neutron cross sections. Neutron source distribution codes and neutron transport codes can be obtained either from the Radiation Shielding Information Center at Oak Ridge National Laboratory, Oak Ridge, TN, USA, or from the Nuclear Energy Agency-Data Bank in Gif sur Yvette, France.

2.1 Neutron Source

A representative fission neutron source density $S(\vec{r},E)$ must be calculated within the reactor core spatial mesh for the time period to be analyzed, which can be a reactor fueling cycle or a shorter period. $S(\vec{r},E)$ may be calculated from the equation

$$S(\vec{r},E) = \bar{v}(\vec{r}) \cdot X(\vec{r},E) \cdot P(\vec{r})$$

where $\bar{v}(\vec{r})$ is the average number of neutrons per fission

$X(\vec{r},E)$ is the fission neutron energy spectrum,

and $P(\vec{r})$ is the spatially dependent fission rate.

To define $\bar{v}(\vec{r})$ and $X(\vec{r},E)$ for a reactor core, one must account for the facts that these parameters differ for fission of $^{235}\text{U}$ and $^{239}\text{Pu}$, and that the relative contributions of these two nuclides to the overall reactor neutron source vary significantly with burnup of the fuel. To define proper time-averaged $\bar{v}(\vec{r})$ and $X(\vec{r},E)$ values for a reactor core, the fuel loading and burnup tables consequently have to be analyzed. The average energy per fission also depends on the $^{235}\text{U} / ^{239}\text{Pu}$ ratio and thus on the burnup, and has to be taken into account when relating neutron production rate to reactor thermal power.

Normally the fission rate $P(\vec{r})$ is determined by diffusion theory calculations together with reactor power distribution measurements. The power distribution within the outer fuel elements must be accurately known, since they dominate the contributions to neutron exposure of the pressure vessel. This is also important in the case of "low-leakage" cores, where highly depleted fuel assemblies are placed at the core periphery.

In the axial direction, use of measured power distributions or form factors calculated from measured axial burnup distributions is preferable to assuming a
simple cosine variation. For boiling water reactors as well as for VVER (PWR type) reactors the changing source distribution within a reactor cycle must be taken into account. The varying neutron spectrum and flux must be calculated in short enough time steps to model the real situation to the desired accuracy.

2.2 Neutron Transport Calculations

The transport of neutrons from the core region to and through the pressure vessel can be modeled either by the method of discrete ordinates (Sn) or by Monte Carlo calculations. However, in practice the Sn method generally has been preferred because of its lower cost. After a brief description of Monte Carlo, we will focus on the Sn method.

The Monte Carlo approach involves tallying the paths of a large number of individual neutrons, the scattering and absorption of which are determined stochastically according to probabilities governed by the geometry of the problem, the materials, and neutron cross sections. This collection of paths is then taken to be representative of the overall neutron transport, within statistical uncertainty. By means of improved calculational acceleration techniques, deep penetration Monte Carlo calculations such as LWR-PV calculations have become practicable. Neutron exposure values for pressure vessel positions can be calculated with sufficient accuracy in a reasonable time. Computer codes such as MCNP, TRIPOLI, and MCBEND have been employed successfully for the analysis of LWR-PV neutron transport problems. The Monte Carlo approach is particularly attractive for calculating three-dimensional effects [39,40] and in cases with unsymmetrical geometry.

The Sn method has been shown to be the most cost-effective for characterizing the radiation environment outside an LWR core. Continual improvements since the early sixties can be followed best by reviewing the series of DOT transport codes [41]. Although the DOT codes can handle problems only in two-dimensional geometries, they have been successfully applied worldwide in analyzing LWR-PV surveillance programs as well as irradiation experiments in materials test reactors. Real three-dimensional (3-D) codes, such as TORT, have also been applied in pilot studies in PV exposure calculation [42], but the requirements for computer time and memory space are extremely high. 3-D synthesis using 2-D results is the more efficient method, and it gives acceptable accuracy at the pressure vessel and surveillance capsule positions that are of interest in PV damage analysis.
The actual details involved in setting up the input files for an $S_n$ code as applied to a particular problem are strongly dependent on the reactor and cycle to be modeled. Therefore, only some advice about the most important features can be given, and we hope this will be helpful to the readers. We recommend that the code manuals be consulted for details. The relevant benchmarks should be carefully reexamined for validating computational methodology. Further details can be found in relevant Standard Guides which have been published by the American Society for Testing and Materials (ASTM) [21].

2.3 Geometric Modeling

As a first step a one-dimensional code such as ANISN should be used to calculate spatially dependent fine group spectra. It is further used to collapse the fine-group data to a broad-group, problem-specific set of about fifty neutron energy groups. At this point, multidimensional transport calculations are practicable.

A real 3-D treatment on the basis of finite difference numerical methods is still not practicable today for routine application. But symmetry conditions in a reactor and slow variation of neutron flux in the axial direction normally allow construction of the 3-D flux distribution by combining the results of lower-dimensional calculations. This procedure, known as "flux synthesis" has been used often and successfully in the past. Normally a LWR is modelled in an $R-\theta-Z$ system of coordinates, and the synthesized 3-D energy group fluxes are calculated from the equation

$$\phi_g(R,\theta,Z) = \phi_g(R,\theta) \cdot \frac{\phi_g(R,Z)}{\phi_g(R)}$$

where $\phi_g(R,\theta)$ are the results from a 2-D axially infinite calculation, $\phi_g(R,Z)$ are the group fluxes from a azimuthally symmetric, $(R,Z)$ calculation, and $\phi_g(R)$ are the group fluxes from a 1-D calculation in cylindrical geometry.

$\frac{\phi_g(R,Z)}{\phi_g(R)}$ can be regarded as the axial form factor at each $R$ point. The synthesis procedure has been shown to provide sufficiently accurate 3-D fluxes at surveillance positions and for the pressure vessel, even at axial locations far away from the active
core height. Symmetry and boundary conditions can help in reducing the space mesh. Normally the analysis of a 45-degree octant is sufficient. In the radial direction accurate modelling of the inner biological shield is essential to calculating the flux level in the reactor cavity and attenuation through the vessel wall. Figure 2 shows a typical \((R,\theta)\)-mesh, used to calculate end-of-life fluence for the German BWR Gundremmingen A (KRB-A). The positions of trepans (A,B,C, etc.) in the vessel are indicated. The trepans were removed after decommissioning to compare calculated and measured neutron exposure data, together with material property change measurements [34].

Special attention has to be paid to modelling of surveillance capsules, which may include a relatively large quantity of iron. Their perturbation effect normally is not negligible. Changes in exposure values or threshold detector reaction rates can exceed 20%, compared with the unperturbed neutron field. Figure 3 shows calculated perturbation effects of iron specimens within a 5-cm diameter tube outside the core barrel of a 1300 MW(e) PWR.

Transport calculations for BWRs are more difficult than similar calculations for PWRs. This is the case principally because of void fraction and control rod movements in the core. By means of adjoint calculations one can determine if changes in the core are influential in affecting exposure values in the vessel. The influence of simplifications of the real 3-D geometry on the target quantities investigated should be carefully checked by comparison with a real 3-D calculation on the same reactor or a similar one.

Convergence studies should be performed to guarantee a sufficiently fine spatial mesh and angular quadrature for \(S_n\)-methods. An \(S_8\) quadrature together with \(P_3\) cross section expansion is recommended for pressure vessel exposure calculations.

2.4 Nuclear Data Libraries for Transport Calculations

The validity of transport calculations depends greatly on the accuracy of the nuclear cross-section data used. There are several processed multigroup libraries available which can be recommended [43,44]. Their energy group structure, with about 170 neutron groups, emphasizes the high energy range. Cross-section minima of important materials like iron are taken into consideration.
3. REACTOR NEUTRON DOSIMETRY

Having discussed methods of calculating the detailed energy-dependent neutron flux distribution in the pressure vessel and surveillance capsules, we now turn to a discussion of neutron dosimetry, which is the measurement of neutron exposure at certain discrete locations.

The accurate measurement of neutron exposure in power reactors requires careful attention to detail. While the main points are covered in this chapter, it is recommended that the reader refer to the current edition of the Annual Book of ASTM Standards, Volume 12.02 [21], for detailed procedures, since space is not available here to discuss everything of importance.

3.1 Types of Dosimeters Used for RPV Dosimetry

The most commonly used dosimeters for measuring neutron exposure of LWR pressure vessels are activation (sometimes referred to as radiometric) monitors. Other dosimeters that have been developed and have been used to some extent are solid state track recorders (SSTRs), helium accumulation fluence monitors (HAFMs), and damage monitors. A relatively recent development for power reactors has been the "scraping sampling" or "scratch dosimetry" technique, a special application of activation dosimetry. We will briefly discuss these dosimeters, and then describe the use of activation dosimeters in more detail. It should be noted that dosimetry methods used in benchmark facilities or experimental reactors include other techniques in addition to those used in power reactors, made possible by the shorter duration of irradiations and/or more ready access. These methods have included fission ionization chambers, nuclear emulsions, and proton recoil spectrometers. They will not be discussed further here, but interested readers can find information about them in Ref. [45].

The method of performing dosimetry with activation monitors consists of subjecting foils or wires to neutron activation at the desired location for a known period of time. They are then removed, and their decay radiation is counted in the laboratory. Reaction rates are calculated, and these can then be used to evaluate flux and fluence.
Advantages of activation monitors are that they are small, passive, convenient, and tolerant of the radiation environment. They can provide spectral information, their accuracy is adequate, and some can be used for long periods of time. A disadvantage is that it is not possible to measure that part of the neutron spectrum between about 0.01 and 0.5 MeV using known activation monitors.

The operating principle of the SSTR is that a thin layer of fissile or fissionable material is irradiated while in contact with mica or quartz. The resulting fission fragments produce damage along their paths in the dielectric. Chemical etching, performed later, reveals damage tracks. These are counted under an optical microscope. Fluences are calculated from the number of tracks per unit area. The chief advantages of this method are that no correction is needed for radioactive decay, that the fissile or fissionable material can be re-used, and that the method works for very low fluxes (down to 20 n/m²-s). Disadvantages are that only fissionable or fissile materials can be used, limiting the number of possible energy thresholds, and that difficulty is experienced in uniformly preparing and determining the mass of the thin layer of fissionable material. Also, track counting is tedious unless performed by automated equipment, tracks may anneal at reactor operating temperatures, and the maximum measurable fluence is limited to about 5x10²² n/m² by track overlap. With the current state of technology, SSTRs are useful for dosimetry in the cavity outside the pressure vessel, but not within surveillance capsules inside the vessel. Additional information about SSTRs is available in ASTM E 854 [21].

The principle of operation of the HAFMs is that helium-4 is produced in monitor materials by (n,α) reactions, and is retained in the material or in a capsule surrounding it. After irradiation, the monitor is vaporized and the amount of helium is measured by gas mass spectrometry. The neutron fluence above the reaction threshold is then calculated using a spectrum averaged cross section. The chief advantage of HAFMs is that helium does not decay or burn out, so they can be used for long times without corrections. Disadvantages are that rather specialized apparatus is needed to perform the helium analysis, and most materials have high energy thresholds for their (n,α) reactions. Exceptions are boron and lithium, which unfortunately show rapid depletion because of their large cross sections for thermal and intermediate (or resonance) neutrons. Another disadvantage is that high purity materials must be used to avoid significant production of helium from impurities. HAFMs have not been widely used in power reactor dosimetry, but the technique appears to offer advantages in determining the fluence directly from samples of steel.
irradiated for long periods of time, provided the chemistry of the steel is well characterized and contributions from impurities such as boron and lithium can be accounted for. Additional information about HAFMs is available in ASTM E 910 [21].

In the case of damage monitors, well-characterized solid materials are irradiated. After irradiation a characteristic physical property is measured which is sensitive to radiation damage. The change in this property is related to damage in steel. The materials that have been used, along with their corresponding properties are as follows: sapphire -- optical absorption due to aluminum vacancy centers; graphite and tungsten -- electrical resistivity; silicon -- electrical properties of P.I.N. diodes; and quartz -- density and refractive index. The advantages of damage monitors are that they provide a more direct, integral measurement of radiation damage, that a single monitor is sensitive to the entire damaging neutron spectrum, and that they are sensitive to the portion of the spectrum between about 0.01 and 0.5 MeV, where other monitors have little or no response. Issues that must be addressed are the usable fluence range, the degree of annealing of damage at reactor temperatures, and the development of an accurate damage correlation to steel. AEA Reactor Services in the UK are pursuing the application of the sapphire damage monitor to cavity dosimetry. Irradiation of reference samples of steel as correlation monitors can also be viewed as a type of damage monitor approach. These are helpful in comparing embrittlement results from one location to another.

The "scraping sampling" or "scratch dosimetry" method [46] involves removing small samples from the surfaces of structural materials during reactor shutdown, analyzing their radioactivity, and calculating either relative or absolute neutron exposures from the results. This method has been used on the inner stainless steel cladding of pressure vessels. The most useful product radionuclides have been $^{93m}$Nb and $^{54}$Mn. One advantage of this method is that the pressure vessel material serves as its own dosimeter, obviating the need to extrapolate or interpolate from other locations. Another is that dosimetry is available at essentially any location on the surface. This provides an excellent way to determine neutron exposure distributions within the reactor. Disadvantages are that one is restricted to using reactions of chemical elements that are already present in the structural material, that one cannot introduce fresh, non-irradiated material at will, and that care must be taken that the sample truly represents the material at the sampling location, and does not contain significant amounts of corrosion products that may have been transported by reactor coolant. It is preferable to perform dosimetry with monitors that are selected and well
characterized in advance. Nevertheless, this method is an important addition to reactor dosimetry, and is particularly useful in cases where neutron exposures must be quantified, but no dosimetry was planned in advance (see section 7).

3.2 Procedure for Dosimetry with Activation Monitors

In this section we will outline the main steps in performing activation monitor dosimetry. The reader should refer to the ASTM standards [21] for further details.

It must first be decided where the monitors will be placed. For test reactor irradiations, the monitors should be located so as to determine average exposure values and exposure gradients for the metallurgical samples to be irradiated. For power reactors, the locations where dosimetry is needed may be specified by national codes or regulations. Normally the locations of interest are (a) within surveillance capsules inside the pressure vessel ("in-vessel dosimetry"), particularly in the beltline region close to positions of maximum flux, and (b) in the reactor cavity outside the pressure vessel, but inside the concrete biological shield ("cavity dosimetry" or "ex-vessel dosimetry"). The former are necessary for evaluating the neutron exposure of the metallurgical samples in the surveillance capsules. The latter have come into wider use in recent years for several reasons: The cavity region is more accessible, and the environment there is less demanding. A larger azimuthal and axial range can be spanned with dosimetry. Dosimetry data can be obtained there to compare with the results of neutron transport calculations outside the thick steel pressure vessel, where the calculations are more sensitive to uncertainties in the scattering cross sections. Cavity dosimetry is also very helpful in monitoring long-term variations in the reactor core power distributions. This is particularly important in cases where fuel management strategies have been changed in order to minimize peak neutron exposure of the pressure vessel. Another application of cavity dosimetry is in evaluating exposure of pressure vessel support structures (ASTM E 1035) [21].

In selecting the monitor materials, a large number of factors should be considered:

- A product nucleus that emits gamma rays is preferred, for ease of analysis.
Monitor materials should be selected that are sensitive to different parts of the neutron energy spectrum, with as complete a coverage as possible.

The materials must be compatible with the environment, including temperature and radiation.

The product half-life should be at least comparable to the duration of irradiation, but not so long that the decay rate will be too slow to give significant counts in a reasonable counting time.

The excitation function (cross section as a function of energy) should be well-known, sufficiently large to produce a measurable product, but not so large that self-shielding and depletion (burn-up) are serious problems.

Product gamma rays preferably should have energies between about 0.06 and 2.0 MeV, with yields high and well-known.

The isotopic abundance of the nuclide to be activated should be high and well-known.

There should be no gamma ray interferences from products of competing reactions at energies too close to the energies of the desired gamma rays to be resolved.

For fission-type monitors, the fission yield of the product nuclide should be high and well-known.

Preferably there should be no highly-absorbing resonances, in order to minimize self-shielding and heating.

The materials should be available in sufficiently high purity and at reasonable cost.

Preferably the cross sections for burn-in and burn-out of the product should be small.
These selection factors describe the ideal activation monitor. In reality, however, our choices are limited to what exists in nature, and compromises must be made. Possible choices for the various energy ranges are given in ASTM E 844 [21]. Guidance on selection of monitors for an experiment to qualify as a calculational benchmark is given in ASTM E 482 [21], paragraph 3.2.1.7: "Reaction rates (preferably established relative to neutron fluence standards) must be reported for 237Np(n,f) or 238U(n,f) and 58Ni(n,p) or 54Fe(n,p); additional reactions that aid in spectral characterization such as provided by Cu, Ti, and Co-Al, should also be included in the benchmark measurements." With these monitors, the energies between which 90% of the activity is produced in a 235U fission spectrum are as follows (see also Fig. 4):

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Response (MeV)</th>
<th>Product Half-life</th>
</tr>
</thead>
<tbody>
<tr>
<td>237Np(n,f)137Cs</td>
<td>0.67 - 5.7</td>
<td>30.17a</td>
</tr>
<tr>
<td>238U(n,f)137Cs</td>
<td>1.51 - 6.7</td>
<td>30.17a</td>
</tr>
<tr>
<td>58Ni(n,p)58Co</td>
<td>2.09 - 7.6</td>
<td>70.82d</td>
</tr>
<tr>
<td>54Fe(n,p)54Mn</td>
<td>2.47 - 7.8</td>
<td>312.5d</td>
</tr>
<tr>
<td>46Ti(n,p)46Sc</td>
<td>3.86 - 9.4</td>
<td>83.81d</td>
</tr>
<tr>
<td>63Cu(n,α)60Co</td>
<td>6.13 - 11</td>
<td>5.271a</td>
</tr>
<tr>
<td>27Al(n,α)24Na</td>
<td>6.5 - 12</td>
<td>15.0h</td>
</tr>
<tr>
<td>59Co(n,γ)60Co</td>
<td>thermal and intermediate</td>
<td>5.271a</td>
</tr>
</tbody>
</table>

As can be seen, several of the product half-lives are short compared to the length of a power reactor fuel cycle, which may be 1, 1.5, or 2 years. If these monitors are used for such time periods, as in surveillance capsules, the measured activation will reflect only the last part of the irradiation, since earlier activation will have decayed. In this case, one must rely on an accurate knowledge of the neutron flux history in order to calculate the fluence for the entire period. Fortunately, there are some product nuclei with multiyear half-lives. For irradiations over shorter periods, other reactions having shorter-lived products can be chosen from the lists given in ASTM E 844 [21]. Detailed procedures for each of these monitors are given in the ASTM book of standards [21]. Special considerations for thermal neutron dosimetry are discussed in ASTM E 262 and E 481 [21]. The German standard DIN 25456 provides standard methods for Fe, Ni, Nb, and Cu monitors [22].
It should be noted that $^{24}\text{Na}$ has too short a half-life to be useful for long irradiations or long decay times, but aluminum, the target element from which it is produced, is useful as an alloying diluent for Co to reduce self-shielding.

Use of the Nb monitor has become more prevalent in recent years because of its response to the lower part of the fast neutron spectrum, down to about 0.5 MeV, and the long product half-life. The reaction is $^{93}\text{Nb}(n,n')^{93m}\text{Nb}$. The product half-life is 16.13a, and analysis involves counting X rays of energies 16.6 and 18.6 keV. High purity Nb is required, especially with respect to Ta. Otherwise, it is necessary to use a chemical extraction method. The Nb should be used in wire form, since very thin foils have been found to oxidize readily and to fall apart. However, for counting the x-rays, the Nb monitor must be converted to a thin source. Details are given in ASTM E 1297 [21].

Once the monitors are selected, their configuration, size, shielding, and encapsulation must be designed. Important factors are as follows:

- Monitors using nuclides with high cross sections should be thin or diluted in an alloy to minimize self-shielding. Thinness is also important to minimize self-absorption of emitted radiation during counting. For fissionable monitors, however, extreme thinness can permit excessive loss of fission fragments. The ASTM standards [21] should be consulted for guidance on thickness of particular monitors.

- Overall size and mass are determined by the space available, by the need to minimize perturbation of the neutron field, and by the need to minimize the corrections required to obtain reaction rates for several monitors at a common point in space. Also, the amount of radioactivity at the time of counting should be sufficient to provide good statistics in a reasonable time, but not so high that dead time is excessive. Monitors must be physically large enough for good precision in weighing and for convenience in labeling and handling. In order to insure that the activity of the monitors after irradiation will be within the dynamic range of the counting equipment, it is important to estimate it beforehand. The expected activity (D) of a monitor at
the time of counting, after the irradiation, can be estimated using the following equation:

\[ D(\text{bequerels}) = N_0 \bar{\sigma} \phi \alpha (1-\exp(-\lambda t_1)) (\exp(-\lambda t_2)) \]

where

- \( N_0 \) = the number of target element atoms
- \( \phi \) = the estimated flux density (m\(^{-2}\) s\(^{-1}\))
- \( \bar{\sigma} \) = the estimated spectral averaged cross section (m\(^2\))
- \( \alpha \) = the product of the nuclide fraction and (if applicable) the fission yield
- \( \lambda \) = the decay constant for the product nuclide (s\(^{-1}\))
- \( t_1 \) = the expected duration of the irradiation (s)
- \( t_2 \) = the expected decay time between the end of irradiation and the start of counting (s).

The spectral averaged cross section is defined by

\[ \bar{\sigma} = \frac{\int \phi(E)\sigma(E)dE}{\int \phi(E)dE} \]

where \( \phi(E) \) is the flux spectrum, and \( \sigma(E) \) is the excitation function for the activation reaction.

Note that the equation for expected activity is only approximate. \( \phi \) and \( \bar{\sigma} \) can only be estimated for an unknown spectrum. Various corrections will be applied to the actual observed activity to improve the accuracy, as shown in later steps below.

- In some cases monitors should be irradiated inside covers designed to absorb thermal neutrons. Thermal neutron shields are used for a variety of reasons: to eliminate extraneous activation in fast neutron monitors, to reduce burn-out of monitor or product nuclei, or to distinguish between thermal and intermediate energy neutrons in neutron capture type monitors. In the case of fissionable monitors such as \(^{238}\)U, shields are used to reduce thermal neutron fission of trace amounts of fissile materials, such as \(^{235}\)U, which produce the same fission products. Thermal neutron shields must have adequate thickness to shield thermal neutrons even after depletion, and
must have a sufficiently high melting point. These shields generally incorporate Cd, B, or Gd. (CdO is often used, rather than Cd metal, which melts at 321°C). Thermal neutron shields are discussed in ASTM E 262 [21]. The individual ASTM procedures for each monitor material should be consulted to determine the need for thermal neutron shielding [21].

- It is advisable to use more than one set of monitors in a surveillance capsule, placed on opposite sides or ends of the metallurgical samples to measure gradients in the neutron field. Wires called "gradient wires" can be arranged to span the entire capsule. After irradiation they are segmented, and the segments are counted individually to evaluate gradients. When thermal neutron shields are used, it is important not to place them too close to monitors intended to measure thermal neutrons.

- Capsules for activation monitors have several purposes:
  
  To position monitors at the correct, known location
  To prevent loss of monitors
  To prevent interference with reactor operation
  To prevent dispersion of monitors having low melting points, high vapor pressures, or vulnerability to radiation damage
  To prevent oxidation
  To control health hazards
  To conform to shipping regulations (In particular, fissionable material may be encapsulated in vanadium.)

- The requirements for capsules are as follows:
  
  They must tolerate the environment
  They must not shield neutrons or perturb the neutron field significantly.
  They must be easy to load and unload without damaging monitors.
  Minimum activation is desirable.
Materials used for capsules have included stainless steel, vanadium, copper, aluminum, and fused silica. Stainless steel is not desirable for encapsulation of thermal neutron monitors because of its high absorption. Fused silica is subject to breakage on impact during handling.

After the monitors, thermal shields, and capsules have been designed, materials must be obtained to fabricate them. In ordering material, it is important to specify sufficiently high purities. Certain impurities are particularly deleterious, such as cobalt in copper and tantalum in niobium. The ASTM standards should be consulted for details [21]. Supplies of activation monitors can be obtained from CEC-JRC-CBNM, Steenweg Naar Retie, B-2440 Geel, Belgium.

Before fabricating monitors, it should be verified by chemical analysis that the specified materials have actually been obtained. (This statement is not intended to disparage suppliers of activation monitors. It is simply good practice to check to be sure that material has not been inadvertently transposed.) The monitors, thermal shields, and capsules can then be fabricated. Monitors and capsules should be labeled by scribing numbers onto them, or permanently marking them in some other way that does not introduce chemical contamination. They should be weighed, and the numbers, materials, weights, and locations should be recorded in permanent records.

Plans should be made before the irradiation for handling and transportation of irradiated material. The radiation dose rates that will be present from the capsules after irradiation should be estimated. Appropriate shipping containers, hot cells, and counting facilities should be available. "Dry runs" (practices) of handling procedures are very helpful.

The capsules can then be assembled and installed. They must be attached securely. Attachment of capsules can be performed by welding, clamping, or pressing into holders built into reactor components. In any case, care must be taken not to cause adverse effects on reactor components or to allow loose parts in the reactor. Careful, permanent records of the installed locations should be kept.
The monitors are then exposed to neutron irradiation during reactor operation. (When possible, dummy dosimetry runs should be performed before irradiations that include steel surveillance specimens. This will enable the dosimetrist to obtain experience and to correct unforeseen problems that may arise before committing the steel specimens.) During the exposure, accurate records of the reactor power history, relative neutron flux in the fuel nearest the dosimeters, fuel loading, and control rod positions should be kept. The duration of irradiation should be sufficiently long to minimize contributions of start-up and shut-down and must be compatible with the reactor operating schedule or fueling cycle. Duration of surveillance capsule irradiation may be established by national codes or regulations.

After irradiation the capsules and monitors are removed from the reactor and transported to the analytical facility. Remote handling and transport casks will be required to limit radiation exposure of personnel to allowable amounts.

The capsules are then disassembled. Hot cell handling may be required to limit radiation exposure of personnel to allowable amounts. Care must be taken not to lose, damage, mix up, or cross-contaminate the monitors. They should be cleaned as necessary. They should be reweighed and the weights compared to the initial weights to verify identities and to correct for corrosion losses. The monitors are then mounted in standard holders or planchettes for counting. In the case of Nb, dissolution and further processing may be necessary (see ASTM E 1297) [21].

Radiation counting is then performed. In most cases this will be gamma ray counting, but beta counting (e.g. for sulphur monitors) and x-ray counting (e.g. for niobium monitors) are also used. For gamma ray counting, a high resolution spectrometry system is preferred. Such a system consists of a high purity germanium or lithium-drifted germanium diode, its liquid nitrogen cryostat, a preamplifier, a high-voltage power supply, a linear amplifier with a multichannel pulse-height analyzer and live time correction, and a data read-out device, such as a printer or teletype. A thallium-activated sodium iodide scintillator system can be used instead of the germanium diode for gamma ray peaks that are well separated. Beta counting can be performed with several types of instruments. X-ray counting is preferably done with a high-resolution lithium-drifted silicon or a germanium detector.

The detector should be well shielded and should be equipped with sample positioning hardware to insure reproducible sample locations. Careful background
measurements and checks for stability should be made. The system should be calibrated for energy and efficiency using national or certified radioactivity standard sources, as described in ASTM E 181 [21]. The counting position should be chosen to give the highest possible count rate without unacceptably high uncertainties due to dead time, summing, or geometry corrections. The duration of counting should be long enough to achieve a sufficiently low statistical uncertainty. As an approximate guide, 20,000 counts are usually enough. Careful records should be kept of the times when counting began and ended. Care should be taken to make sure that all times and dates are placed on the same basis, particularly for short-lived products (for example, be sure to account for daylight savings time if used, leap years, transport of monitors across time zones or the International Date Line, etc.).

The counting system should be calibrated against standard neutron fields. NIST in Gaithersburg, Maryland, USA, and PTB in Braunschweig, Germany, maintain fission spectrum standard neutron fields. Foils irradiated to specified fluences in such a field should be counted using the same procedures used for the activation monitors, and the results should be reported.

The value of the absolute activity (D) of each monitor product nuclide at the time when the irradiation ended is calculated. This is defined on the basis of the entire monitor.

\[ D \text{ (bequerels)} = AI S_R S_C E G P \exp(\lambda t_2) C \]

where
- \( A \) = average observed count rate (counts/s)
- \( I \) = correction for gamma self-absorption in monitor
- \( S_R \) = correction for random coincidence summing
- \( S_C \) = correction for true coincidence summing
- \( E \) = reciprocal of detector efficiency for photopeak and counting position used
- \( G \) = sample size and geometry correction
- \( P \) = reciprocal of gammas per disintegration
- \( \lambda \) = decay constant = \( \ln 2 / t_{1/2} \) (s\(^{-1}\))
- \( t_{1/2} \) = half-life (s)
- \( t_2 \) = time from end of irradiation to start of counting (s)
- \( C \) = correction for decay during counting = \( \lambda t_C / (1 - \exp(-\lambda t_C)) \)

and
- \( t_C \) = true elapsed counting time (s).
Detailed information on the various corrections in this equation as well as in the following two equations can be found in Ref. [21]. Space is not available to discuss them fully here.

The (theoretical) saturated specific activity is then calculated from the (actually observed) absolute activity. The saturated specific activity \( \lambda_s \) is defined as the activity (per unit mass of the monitor) which the monitor would have had if it had been left in the full power flux for a time very long compared to \( t_1/2 \) and if no depletion (burn-up) of the target nuclei or burn-out of the product nuclei had occurred.

If the flux was constant during the actual irradiation,

\[
\lambda_s = \frac{\text{bequerels}}{g} = \frac{D \, B_o \, C_D}{W(1 - \exp(-\lambda \, t_1))}
\]

where \( B_o \) = correction for actual burn-out of product nuclei
\( C_D \) = correction for actual depletion (burn-up) of target nuclei
\( W \) = mass of monitor (g).
and \( t_1 \) = duration of irradiation (s)

If the flux was not constant, the irradiation must be divided into \( N \) segments, in each of which the flux is approximated as constant:

\[
\lambda_s = \frac{\text{bequerels}}{g} = \frac{D \, B_o \, C_D}{W \sum_{i=1}^{N} R_i(1 - \exp(-\lambda \, \Delta t_i))(\exp(-\lambda (t_e - t_{oi}))}
\]

where \( R_i \) = the fraction of full power flux during time segment \( i \)
\( \Delta t_i \) = the length of time in segment \( i \)
\( t_e \) = the time at the end of the irradiation
and \( t_{oi} \) = the time at the end of segment \( i \).

\( R_i \) is determined from total reactor power for small test reactors or from self-powered (Hilborn-type) neutron flux monitors located in nearby fuel elements for power reactors. It should be noted that the relationship between \( R_i \) and reactor power changes with burnup because the relative contributions of \( ^{235}\text{U} \) and \( ^{239}\text{Pu} \) change.
The reaction rate for each monitor reaction is calculated from the saturated specific activity. The reaction rate \( (R_R) \) is defined as the rate at which individual, isolated nuclei of the target nuclide would undergo the reaction of interest if placed in the neutron field of interest:

\[
R_R \left( \frac{\text{reactions}}{\text{s. nucleus}} \right) = \frac{A_S M S_S B_I}{N f Y}
\]

where
- \( M \) = the gram-atomic mass of the target element
- \( S_S \) = the correction for neutron self-shielding
- \( B_I \) = the correction for burn-in of the product nuclide from competing reactions, if any.
- \( N \) = Avogadro's number
- \( f \) = the atom fraction of the target nuclide
- \( Y \) = the fission yield of the product nucleus if fission monitor is used \((Y = 1\) for non-fission monitors).

When fission foils are used, attention should be given to the possibility of significant contributions from gamma-ray induced fission (photofission). The gamma-ray fluxes should be estimated or calculated by a transport code to determine the contributions from photofission. If fission foils are used to high burn-up, contributions from fission of nuclides that have grown in must be considered (e.g. \(^{239}\text{Pu}\) in \(^{238}\text{U}\)).

4. DETERMINATION OF NEUTRON EXPOSURE BY COMBINING NEUTRON TRANSPORT CALCULATIONS AND DOSIMETRY

In order to compare the results of the transport calculations to those from dosimetry, it is necessary to calculate predicted dosimetry monitor responses using the neutron spectrum from the transport calculations. Before doing so, the perturbations due to the surveillance capsules should be modeled in the calculations.

\[
R_R(\text{predicted}) \left( \frac{\text{reactions}}{\text{s. nucleus}} \right) = \sum_{g=1}^{k} \sigma_g \phi_g
\]
where $\sigma_g$ are the group-averaged cross sections for the monitor reaction, obtained from an evaluated cross section file ($m^2$). These are available as the IRDF (International Reactor Dosimetry File) from the IAEA Data Section in Vienna. See also ASTM E 1018 [21].

and $\phi_g$ are the group fluxes from the calculated neutron spectrum ($n/m^2s$).

The comparison is usually expressed as a C/E ratio for each monitor reaction:

$$\frac{C}{E} = \frac{R_R(\text{predicted})}{R_R(\text{measured})}$$

If the calculated and measured dosimeter responses agree to within the required accuracy, the transport code is used to calculate the neutron field in the pressure vessel material. If not, the calculated spectrum must be adjusted. Note that the transport calculations normally are not carried down to thermal energies because of the long computer times required, so there will not be a predicted $R_R$ for the thermal neutron monitors. The thermal flux is normally calculated from the monitor results, and this is taken as the true value.

If the C/E ratios differ from 1.0 by too large a factor, it is necessary to adjust the calculated neutron spectrum. If the resources are available this is best accomplished using a computer program that will do a simultaneous best fit using all three sets of input: the evaluated neutron cross sections, the reaction rates based on the measured monitor results, and the neutron spectrum obtained from the neutron transport calculations. The program should take into account the uncertainties in each of these three inputs in terms of covariances and bias factors. The sources of uncertainty in the transport calculations include uncertainties in the inelastic scattering cross sections for steel, the neutron spectrum from thermal fission of $^{235}U$ and $^{239}Pu$, the dimensions and relative locations of the reactor components, the densities of materials, the three-dimensional flux synthesis, and streaming corrections. The dosimetry uncertainties include uncertainties due to random counting statistics, gamma-ray counter efficiency calibration, contributions from competing reactions, and normalization to reactor power.

The most advanced computation system that takes account of these uncertainties in a rigorous way is LEPRICON [22], available from the Radiation
Adjusted neutron spectra are computed with this system by a generalized linear least squares procedure that combines the nuclear data, bias factors, and calculated and measured dosimetry. Results for the neutron field in the pressure vessel material are then obtained from the transport code by making use of correlations that exist between the calculated spectra at the dosimetry and pressure vessel locations.

If the transport calculations are found to be incompatible with the dosimetry results, and errors cannot be found, then the one that predicts the higher fluxes should be used, and larger uncertainties should be assigned to the final results.

The full power thermal (<0.5 eV), epicadmium (>0.5 eV), and fast (both >0.1 MeV and >1.0 MeV) neutron fluxes should be calculated for the locations of interest in the pressure vessel material, using the neutron transport code, as adjusted, for the higher energies and the activation monitor results for the thermal energies. The fluences corresponding to these fluxes should be calculated per effective full power year. Estimates of fluences at future times are made by multiplying the fluence per effective full power year by the time in years and an estimated plant capacity factor. Caution should be used in projecting the fluence at long times from fluxes determined early in the reactor's life, because the core power distribution of new fuel changes as non-uniform burn-up occurs. Estimates of uncertainties should be made.

Displacements per atom (dpa) should also be calculated for the locations of interest. For details, see ASTM E 693 [21].

5. MOCK-UP EXPERIMENTS

As mentioned in section 1.1 above, benchmark facilities have been set up in which mock-up experiments have been run to simulate actual reactor geometries. Neutron exposure has been calculated and measured, and the comparisons have been very helpful in reducing uncertainties by identifying areas where improvements are needed. A discussion of mock-up work for VVER-type reactors is given in [27] and [33]. Figure 5 shows the geometry of the mock-up, and Figures 6 and 7 show relative
neutron energy spectra for the surveillance position (R2) and for the ex-vessel position (R6) respectively, as determined by proton recoil spectrometry. The relative spectra are expressed in terms of F(E)\*E, which is equal to flux per unit lethargy.

6. THE DOSIMETRY REPORT

It is very important to make the dosimetry report as complete as possible. It should provide the information that will be needed by all potential users. We recommend that the following be included:

1. Identification of monitors used
2. Source of monitor material and chemical purity
3. Thicknesses of monitors
4. Locations in reactor and relative arrangement
5. Thickness and material for thermal neutron covers
6. Thickness and material for dosimetry capsules
7. Power or relative flux history during irradiation
8. Dates and times of beginning and end of irradiation and counting
9. Standard sources used for energy and efficiency calibration of radiation counting system.
10. Calibration standards used for chemical balance
11. Description of calibration against standard neutron fields, if used.
12. Total count, duration of count, and distance from detector for each peak.
13. Background count and duration for each peak used.
14. Values of parameters used in equations to derive reaction rates from counting data
15. Gamma ray energy spectra and reaction cross sections used to make photofission corrections for fissionable monitors
16. Absolute activities, saturated specific activities, and reaction rates for all reactions used
17. Specification of activation cross section sets used in calculating predicted reaction rates
18. C/E ratios for reaction rates
19. Description of adjustment procedures for neutron spectra

20. Dosimetry results:

a. Full power neutron fluxes for locations of interest, including thermal (<0.5 eV), epicadmium (>0.5 eV), and fast (both >0.1 MeV and >1.0 MeV), with uncertainties. For VVER-type reactors, the flux for E>0.5 MeV should also be specified.

b. Fluences for the locations of interest (same energy ranges as in a.) with uncertainties

c. Displacements per atom (dpa) for locations of interest, with uncertainties

d. Full power dpa rates for locations of interest, with uncertainties.

e. Tabulation and plots of neutron spectra for locations of interest, preferably as flux per unit lethargy on a log-log plot.

f. Spatial gradients of neutron flux (E>1.0 MeV), neutron fluence (E>1.0 MeV), and dpa throughout the array of metallurgical specimens in surveillance capsules.

7. RETROSPECTIVE DOSIMETRY

Cases sometimes arise in which it is necessary to determine the past neutron exposure for a reactor that was not equipped with dosimetry monitors in advance. This presents a considerable challenge, since one must make use of whatever happens to be available.

If the reactor can still be operated, it may be possible to perform the type of planned dosimetry discussed in section 3.2 for a future period of operation, and then use the results to project back to the earlier operation that was not monitored, provided
there is sufficient knowledge of the previous history of fuel loading and power to perform reliable neutron transport calculations.

If the reactor is no longer in operation or if the neutron exposure results are needed to help in making a decision as to whether it is safe to resume operation, other means must be used.

An estimate may be obtained by comparison to other reactors of the same design, if any exist. This approach is of course subject to large uncertainties, particularly if the power histories and fuel loading histories are not well known for both reactors, or if the detailed geometries of the reactors are not the same.

A purely calculational approach can be attempted if there is enough knowledge of the geometry, materials, and history of fuel loading and power. Of course, there is then no independent, experimental check on the results.

Radiation spectrometry of irradiated structural parts can also be performed as in the "scratch dosimetry" approach discussed in section 3.1 above. Success with this approach depends on the presence of products of activation that have sufficiently long half-lives that they can give information about the irradiation that occurred throughout the period of interest. Unfortunately the product radionuclides with relatively long half-lives in pressure vessel steels tend to be those activated by thermal neutrons, such as $^{55}\text{Fe}$ (2.68a) and $^{60}\text{Co}$ (5.27a), whereas fast neutrons are often of more interest. $^{54}\text{Mn}$ (312d) is perhaps the best candidate. If $^{93m}\text{Nb}$ (16.13a) is present, it will be very helpful. Figure 8 shows a comparison of calculated and measured Mn-54 specific activities within the vessel wall of the decommissioned KRB-A reactor. The dosimetry data have been obtained from trepans taken out of the vessel at several locations [34].

Another requirement for successful scratch dosimetry is an accurate knowledge of the original composition of the material. This is particularly important if trace impurities are activated. Analysis of archive material will probably be adequate for major constituents, but if use is to be made of trace impurities, analysis will probably be needed on the actual irradiated sample, since the composition may not be uniform.
In using any of these retroactive approaches, one must make a careful estimate of uncertainties and keep them in mind in making decisions. Planned dosimetry is very much preferred when possible.

8. SUMMARY OF NEUTRON EXPOSURE DETERMINATION

The evaluation of neutron exposure is a necessary and vital part of efforts to monitor, understand, and predict pressure vessel embrittlement. It is performed by a combination of neutron transport calculations and dosimetry. Present methods have been developed over the past 35 years and have resulted from both individual and coordinated projects in many countries. It is now possible to evaluate integrated neutron exposure with uncertainties as low as 10% for power reactors.

The optimum neutron exposure parameter has still not been established. Until a better understanding of the embrittlement process is obtained, a complete evaluation and reporting of the energy-dependent neutron flux distribution in embrittlement studies is prudent. Dpa should also be calculated and reported.
Figure Captions

Figure 1. Normalized Calculated Neutron Spectra for the Core and Pressure Vessel of a Boiling Water Reactor (G. Prillinger).

Figure 2. Calculational Mesh Used for the German BWR Gundremmingen A (KRB-A) Trepans (G. Prillinger)

Figure 3. Calculated Perturbation of Iron Specimens on Flux (E>1MeV) and DPA Within a 5-cm Tube Outside the Core Barrel of a 1300 MW(e) PWR (G. Prillinger)

Figure 4. Energy Ranges of Response for Activation Monitor Reactions Having Products with Various Half-lives, Superimposed on a Representative Reactor Neutron Spectrum (Nuclear data supplied by ECN Petten, Netherlands).

Figure 5. Diagram Showing the Geometry of the VVER440 PWR Mock-up Experiment in the LR-O Experimental Reactor at the Nuclear Research Institute, Rez, Czech Republic (Ref. [47]).

Figure 6. Relative Neutron Spectrum for the Surveillance Position R2 in Fig. 5 as Measured by Proton Recoil Spectrometry (Refs. [47] and [48]).

Figure 7. Relative Neutron Spectrum for the Ex-Vessel (Cavity) Postion R6 in Fig. 5 as Measured by Proton Recoil Spectrometry Refs [47] and [48].

Figure 8. Comparison of Calculated and Measured Mn-54 Specific Activities Within the Vessel Wall of the Decommissioned KRB-A Reactor. Data Taken from Trepans (G. Prillinger).
Neutron spectrum per lethargy, normalized -BWR- (core-Y 1, pressure vessel-Y 2)  

FHTE-KL  
30.03.92  

FIGURE 1
FIGURE 2

1 reactor core
2 reflector
3 core barrel
4 downcomer
5 vessel cladding
6 vessel
7 reactor cavity
8 concrete
FIGURE 3
VVER 440 mock-up in LR-O experimental reactor

Water density reducing displacing tank

Barrel
Basket

LR-O tank

VVER 440 core simulator

Surveillance position R2 (1.625 m)
Pressure vessel (PV) outer surface R6 (1.92 m)

FIGURE 5
FIGURE 6

F(E)•E [relative]

energy [MeV]

90% response

F(E)•E
Figure 7

- 90% response

$F(E) \cdot E$ [relative]

Energy [MeV]

- Nb
- Fe
- Cu

$F(E) \cdot E$
FIGURE 8
REFERENCES


[15] Proceedings of the Fourth ASTM-Euratom Symposium on Reactor Dosimetry, Gaithersburg, Maryland, USA, Mar. 22-26, 1982, F. B. K. Kam, ed., Published as


[40] CARTER, M. D., CURL, I., MILLER, P., and PACKWOOD, A., "The LWR Radial Shield Benchmark Studies of the NESTOR Shielding and Dosimetry Improvement Program's NESDIP," see Ref. 39.


7.1 Introduction

It will be apparent from earlier chapters that characterisation of the neutron irradiation effects response of Reactor Pressure Vessel (RPV) steels is a key issue in the management of ageing in commercial nuclear plant. While this is a subject of intense scientific study, which demands well characterised closely controlled irradiation experiments, usually in a Materials Test Reactor (MTR), it is also one of direct regulatory concern in which nuclear plant operators are required to mount surveillance programmes to monitor the state of embrittlement in their plant. More specifically the objectives of these two types of irradiation experiment are as follows:

MTR -

- Provide sufficient control of environmental variables such as irradiation temperature, neutron flux and neutron energy spectrum for specific response functions to be defined;
- Permit accelerated irradiations to and beyond end-of-life doses for studying the effect of metallurgical variables such as composition, mechanical-thermal treatment and microstructure;
- Provide suitably well characterised databases for embrittlement model development and validation.
Surveillance - Monitor the state of embrittlement in 'typical' plant specific materials (usually plate, weld and HAZ) under conditions as close as possible to those in the critical component during operation;

Establish databases of plant specific or generic materials behaviour for the development of embrittlement trend curves.

Though the objectives of MTR and surveillance experiments may be different, many of the same metallurgical and environmental factors govern the materials response. Consequently it is appropriate to discuss in general terms the way in which the logistical and environmental constraints of each type of experiment conflict with the objectives and the degree to which they may be overcome.

In this chapter no attempt will be made to describe in great detail the numerous designs of irradiation capsule and MTR facilities employed during the last 30 years of irradiation experimentation. Rather it is intended to highlight in more general terms the most important experimental variables and design principles in order to provide guidance for optimisation of irradiation experiments.

7.2 General Aspects

As already indicated, the irradiation response of RPV steels is determined by a number of material and environmental factors which are summarised below:

Material Condition - Chemical composition, with regard to overall and impurity levels but particularly copper, nickel and phosphorus content, heat treatment, mechanical-thermal history and microstructure can influence both the rate of embrittlement and end-of-life embrittlement levels; because of the variation in properties and composition through the thickness of many RPV plates, the depth and location of all specimens should be carefully noted; wherever possible, well characterised, widely used standard reference materials (correlation monitors) with well defined response should be included in any irradiation:

Temperature - Irradiation temperature can strongly influence embrittlement response; typically Charpy transition temperature shift in
irradiation sensitive steels will decrease by about 1°C for every 1°C increase in irradiation temperature throughout the temperature range of interest for commercial power reactors:

**Neutron Fluence** -
Generally expressed as the time integrated flux of neutrons with energies greater than a threshold value usually 0.1, 0.5 or 1MeV; embrittlement increases with neutron fluence, generally at a reducing rate as fluence increases, in some cases appearing to saturate at high doses; the higher sensitivity at low doses necessitates high quality dosimetry methods to minimise scatter for trend curve development:

**Neutron Flux** -
Generally expressed as the flux of neutrons with energies greater than a threshold value usually 0.1, 0.5 or 1MeV; variations of embrittlement at a given fluence at different fluxes can be important, particularly at lower doses; the latter arise due to the complex interaction of time, temperature and dose with point defect diffusion and aggregation processes and irradiation induced or enhanced precipitation and segregation processes; it is probable that significant changes in flux, even within the same reactor, are likely to be associated with changes in energy spectrum:

**Neutron Spectrum** -
Neutron energy spectrum determines the relative proportion of low and high energy primary recoils and therefore the damaging power of a neutron environment; its accurate determination is important when comparing behaviour in different MTR's or different surveillance locations; in such cases calculation of the damage parameter, displacements per atom (dpa), generally provides a better correlation than fluence:

**Environment** -
The effect of chemical environment is possibly the least understood variable; close control of gaseous environment to avoid/minimise oxidation and carburisation/decarburisation are therefore advisable; additionally, care should be taken during specimen and capsule preparation to avoid contamination from potentially surface active reagents, such as chlorine based degreasing agents:
All specimens should be clearly marked, preferably by engraving, to ease later identification in hot-cells; the sampling location and orientation of all specimens in the irradiation should be accurately known, as should their precise location relative to flux and temperature monitors; sufficient archive material should be retained to permit additional materials characterisation or supplementary testing at a later date; if possible a simple and cost effective test such as pre- and post-irradiation hardness should be conducted on all specimens as an additional check against spurious results.

The methodology adopted for conducting experiments in MTR's and Power Reactor Surveillance will now be described. Particular reference will be made to the degree to which the various environmental and metallurgical factors enumerated above may be controlled and to the influence this has on the quality of embrittlement information.

7.3 MTR Irradiations

MTR irradiation experiments on pressure vessel steels can be categorised into three main types:

(a) General MTR irradiation of test pieces in "standard" irradiation rigs;

(b) Tailored MTR irradiations in which the reactor neutron spectrum is modified locally within the reactor to simulate that experienced at the pressure vessel wall of a LWR;

(c) Purpose built test facilities for simulating reactor surveillance and RPV environments and, in particular, assessing the effect of the attenuation of the neutron spectrum/energy and properties between surveillance positions and through the simulated vessel wall.

The important features of each of the above types of MTR facility will be discussed by reference to specific examples and an assessment will be made concerning future needs.
7.3.1 General Irradiation Rigs

Samples of RPV Steels have been irradiated in a wide range of MTR types including heavy water moderated and light water moderated systems. Operators of most MTR's have developed general irradiation rigs for particular types of irradiation of which RPV steels are one. The advantage of this type of rig is that it is usually relatively cheap to build, is to a standard design and it can be installed in virtually any empty location in the reactor given the constraints of flux indicated below. These rigs enable irradiation experiments to be mounted in rapid response to urgent operational or regulatory concerns or to perform scoping experiments on relatively large numbers of specimens to determine the relative effects of metallurgical variables.

Flux levels used for accelerated irradiations of pressure vessel steels are typically in the range $10^{15}$-$10^{17}$ n.m$^{-2}$ ($E>1$ MeV). Nuclear heating, sometimes referred to as $\gamma$-heating, becomes a limiting factor at fluxes $>10^{18}$ n.m$^{-2}$ when capsules containing sodium or similar heat transfer media may be needed to extract the heat. Consequently, such high flux levels are not generally used in the study of pressure vessel steels since target fluences are generally low, $10^{22}$-$5\times10^{23}$ n.m$^{-2}$ ($E>1$ MeV).

A typical simple type of irradiation rig used in the UK contribution to the IAEA Phase 2 Coordinated Research Programme[1] for the irradiation of pressure vessel steel Charpy impact samples in a light water moderated pool type reactor is shown in Figure 1. The samples are arranged in rows and are clamped in a copper carrier block thus ensuring good thermal contact and a predictable temperature distribution is obtained.

The sample assembly is suspended within a square aluminium irradiation tube with accurately defined gaps between the sample carrier and the tube. In the reactor such irradiation assemblies can be designed to be heated solely by nuclear heating of the steel samples and the copper block, sometimes assisted by a profiled central mass of stainless steel to even out differential heating effects due to the flux profile. The temperature of the samples is controlled by adjusting the thermal conductivity of the gas gap between the sample carrier block and the irradiation tube. This is achieved by adjusting the flow and/or composition of the gas (helium, helium/neon). This provides the simplest and most cost effective rig design but to obtain greater flexibility, particularly with respect to flux, electrical heaters are often incorporated into the design.
It is generally important to derive a good understanding of the thermal characteristics of such rigs by use of calibration rigs containing thermocoupled specimens and detailed heat transfer modelling, in order to interpret the temperature distribution derived from the thermocouples in the carrier assembly. In operational rigs, the temperature distribution within the specimen assembly is determined by an array of thermocouples which, because the rigs are usually required to be reloadable, are generally strategically distributed throughout the carrier assembly and not therefore directly in contact with the specimens. These thermocouples are continuously monitored during the irradiation. In reusable rigs, where a number of reirradiations are anticipated, some redundancy should be built into the number of thermocouples installed to allow for losses due to irradiation degradation. This factor is often the life limiting feature of these simple irradiation rigs.

The neutron dose received by the steel specimens is determined by flux monitors often in the form of wires and special dosimetry capsules containing foils of various materials placed at strategic positions within the rig. This allows the flux distribution and the fluence to be determined.

Typical monitor packs used comprise a series of Cu, Fe, Ti, Ni, Nb, Co foils and, in UK experiments, sapphire direct damage monitor. Fe, Ni or, more recently, Nb wire monitors are also sometimes placed along the notch of Charpy specimens in order to determine the variation of fluence along the length of the rig. The utility of the latter will be discussed in more detail below. The neutron environment experienced by the samples in a general MTR irradiation is often strongly dependent on the loading of other rigs in the MTR and operational factors such as fuel management and reactivity control, which may vary from cycle to cycle. Consequently, in this type of facility it is vital to derive as much dosimetry information as possible to be able to account for the fluctuations in both flux and flux spectrum that may occur.

Well characterised reference material may also be used to provide a correlation between different rigs, MTR's and power reactors. An example of such materials is the JRQ reference plate adopted as the standard reference material for the IAEA Phase 3 Coordinated Research Programme. Samples of this steel have been incorporated in all of the national irradiation programmes to enable such correlations to be made. Other common standard reference materials are A302B and HSST01/02 reference plates supplied by ASTM[2] and employed widely in US research studies and surveillance programmes since the early 1960's.
Experience from UK programmes suggests that Vickers hardness generally provides a sensitive and reliable indicator of irradiation hardening and therefore embrittlement. It has therefore become common practice to substitute small packs of 10mm square, 2mm thick slices of reference materials for one or two Charpy specimens in the rig loading, to allow such correlations to be made between different irradiations and provide a check in the event of suspected rig malfunction or spurious results.

Irradiation rigs of this generic type range in complexity and sophistication from simple reusable designs of the type illustrated above to those employed for example in the USNRC sponsored HSSI programme[3] (Figure 2) which can accommodate specimens of varying geometry from Charpy and 0.5T-CT specimens up to 4T-CT. In all cases the stringent requirements of temperature control and monitoring and need for comprehensive dosimetry apply. Even in the more sophisticated rigs, significant variations in flux distribution and spectrum can generally occur unless the reactor fuel and reactivity management is strictly controlled.

7.3.2 Tailored Irradiation Facilities

The neutron irradiation environment of an MTR can be tailored to simulate that of a power reactor, which is particularly important for heavy water moderated MTR's in which the thermal-fast ratio is generally very much greater. As part of the UK contribution to the IAEA Phase 3 programme, a flux converter facility (Figure 3) was constructed in the heavy water moderated PLUTO MTR at Harwell for the irradiation of pressure vessel steels in a simulated light water reactor neutron environment[4]. This facility was designed to provide:

(1) A neutron energy spectrum similar to that experienced by a light water moderated PWR pressure vessel, with a low ratio of thermal to fast neutron flux;

(2) A neutron fast flux similar to that obtained in light water moderated MTRs, typically $1-5 \times 10^{16} \text{n.m}^{-2}\text{s}^{-1}$;

(3) A low spatial variation in neutron flux, gamma flux and temperature across relatively large test pieces (60mm x 60mm compact specimens);

(4) A well characterised and controlled irradiation environment in terms of temperature and neutron spectra.
The fast flux in the Flux Converter was locally enhanced in a large peripheral hole in the reactor reflector by surrounding the specimen assembly with a volume of enriched fuel. This fuel was fissioned by the incident thermal flux from the core, thereby converting the predominantly thermal neutron flux to a fast flux within the rig volume. A series of reactor physics low power runs were made with multiple foil dosimetry packs to characterise the neutron spectra and tailor the local neutron environment to the required specification.

A series of calibration and reloadable operational rigs were designed for the converter, for different specimen geometries. The in-core part of the operational rigs (Figure 4) consisted of a split aluminium carrier with milled out recesses into which cassettes containing the samples could be loaded. Thermocouples were located at selected positions within the body of the aluminium carrier and also within specimens and cassettes in the early calibration rigs (Figure 5).

The required temperatures were achieved by a combination of gamma heating in the samples and specially shaped stainless steel nuclear heaters to compensate for axial flux gradients. Temperature control was maintained by controlling the thermal resistance of the gas gap between the rig and the converter coolant. This was achieved by changing the composition of the flowing He/Ne gas mixture within the gas gap.

The temperature and flux distribution within the converter were determined in fine detail from the calibration programme and from this the design and routine operation of the operational rigs was optimised in terms of temperature and flux profiles with respect to the sample positions. With such a facility this extensive early calibration pays dividends in that, once done, the task of routine operation becomes simplified by the well characterised response of the rig.

Experience gained with the Flux Converter facility is instructive of the type of problems encountered when mounting large irradiation programmes particularly with respect to the characterisation of the neutron environment. The details of the flux gradients in and around the specimen assemblies were found to be complex and only became understood because of the recognition early in the programme that iron or niobium wire monitors should be placed along the length of each specimen cassette to provide fine detail on the local flux gradients. This was accomplished by arranging the Charpy specimens so that their notches faced one another creating a hole along the
length of the cassette through which the monitor wire(s) could be threaded. Standard dosimetry capsules containing the range of monitors enumerated above were also placed in the end of each cassette. Typical iron monitor activities for the rig are illustrated in Figure 6, from which it is apparent that the capsule monitors, referred to by SDM (Sapphire Damage Monitor) reference numbers, usually indicated higher activities, by up to 20%, than the notch wire monitors, referred to by a code indicating cassette (top, middle or bottom) and position. From the rig design it was apparent that the capsule monitors, though mounted in the cassettes were at the end, within the aluminium body. The notch monitors, on the other hand, were surrounded by the more heavily absorbing steel of the packed specimens. The latter therefore more closely experienced the actual neutron environment of the specimens and in particular the root of the Charpy notch, for which reason they were originally included. There is clear evidence of neutron streaming between the masses of steel specimens, indicated by the high capsule activities and stronger absorption in the middle of each specimen pack.

The latter experience indicates a number of points regarding the conduct of irradiation experiments. Because of the need to exploit as much of the limited volume available in most MTR irradiation rigs, close attention should be given to obtaining the best estimate of flux gradients. Coupled with well documented rig loading information, the influence of such gradients on results can be investigated. Dosimetry, no matter how sophisticated (ie. specially designed capsules), is of little value if it does not reflect the actual environment of the specimens being irradiated. If such monitors are to be employed it would be prudent to sacrifice several specimens in favour of dummies consisting of capsules buried in steel of similar composition. A simple dosimetry technique, such as the use of long wire monitors along the notches of Charpy specimens, provides a more representative indication of specimen doses. Given the strong absorption characteristics of steel, it is wise to always orient specimens which have a uniquely defined testing direction in a self-consistent manner with respect to flux gradients and monitor positions.

The tailored facility has significant advantages over the simpler irradiation rigs, particularly in respect of neutron spectrum, since its spectrum is controlled by its own fuel and local light water environment. It is nevertheless still at the mercy of the reactor operation regarding flux gradients and temperature gradients through its reliance on passive nuclear heating. A further improvement in operation would therefore be gained by installing electrical heaters.
7.3.3 Purpose Built Irradiation Facilities

A good example of this type of facility is the Pool Side Facility (PSF) which was designed to simulate the surveillance-pressure vessel configuration in Light Water power reactors and test the validity of procedures to determine the radiation damage in the vessel from test results from surveillance capsules. This facility provides a useful blueprint for the type of facility for which there is increasing need as many ageing plant near the date for licence renewal in which the effect of attenuation of radiation damage through the RPV may be a critical issue.

This facility consisted of a Simulated Surveillance Capsule (SSC), Simulated Pressure Vessel Capsule (SPVC) and a Simulated Void Box Capsule (SVBC) which were positioned adjacent to the aluminium window of the Oak Ridge Research Reactor (ORR). The PSF supported the capsules and facilitated the movement of the capsules in and out of the neutron radiation field and relative to one another. A schematic view of the PSF with exploded views of the capsule assemblies is provided in Figure 7.

An exploded view of the specimen assemblies in the SSC and SPVC is provided in Figure 8 and 9. Each assembly, one in the SSC and three in the SPVC, consisted of a rigid outer frame into which the array of specimens was fitted. The three specimen frames of the SPVC, which correspond to the 0-T, \( \frac{1}{4}\)-T and \( \frac{1}{2}\)-T locations in a typical RPV, were separated by heater plate assemblies and cooling assemblies to provide thermal separation and minimise thermal gradients due to radiation energy deposition in the capsules. A range of different dosimeter capsules were distributed throughout the specimen assemblies, often utilising bolt holes or other spaces, as is apparent from Figure 10 for the surveillance and surface assemblies. Additionally, iron monitor gradient strips were fitted in the notches of Charpy specimens.

Temperatures of specimens were monitored and controlled to within ±7°C of the target temperature, 288°C, by 20 thermocouples attached strategically throughout each specimen assembly. This was accomplished by a combination of electrical heaters and internal heat generation. As with rigs described above, the thermal resistance within intercomponent gaps was varied by changing the composition of the mixture of helium and neon cover gas to bring control heater powers to within the optimum range.
Extensive dosimetry studies were carried out before the irradiation proper, in mock-up irradiations to characterise the flux in each specimen location. Consequently, considerable understanding of spectrum and flux distributions was available prior to the irradiation programme. Nevertheless, considerable differences, ~25%, were found between the final irradiation doses predicted on the basis of the mock-up experiments and the actual irradiation doses. These were traced to differences in core loadings and were readily accounted for by the comprehensive dosimetry conducted during the irradiation programme and supporting neutron transport calculations. Were this facility to have continued in operation this experience would have been fed back into the better definition of subsequent programmes.

Target fluences in the facility were 2 and 4 x 10^{23} n.m^{-2} for two irradiations in the SSC position of 46 and 92 days respectively. Comparable doses to the 1/4-T and 0-T positions respectively were accomplished by an irradiation of the SPVC of about 600 days. Though accelerated, these doses scale with surveillance and end of life doses to typical PWR's. In addition to providing embrittlement data for 5 different RPV steels, the irradiations yielded flux and damage dose attenuation information which is employed in the USNRC Reg. Guide 1.99 Rev2[6].

The reason that demand for a facility of this type is expected to increase in future is the fact that, while a consistent picture emerged for the attenuation of flux and damage dose through the vessel, no consistency was observed with embrittlement levels. Virtually all the materials studied appeared to exhibit a different sensitivity as discussed in detail by MCElroy et al[7]. The Reg. Guide understandably takes a conservative approach with respect to this data and potentially there is benefit to be derived from demonstrating greater attenuation of embrittlement on a plant or material specific basis.

A dedicated facility similar to the PSF would have significant advantages over the simpler forms of irradiation rigs in that, once fully characterised and operational, the irradiation conditions should be predictable and reproducible. The facility would also provide an suitably well characterised environment for benchmarking dosimetry and neutron transport methods.
7.4 Power Reactor Surveillance

National regulations of most countries[8-11] require that commercial nuclear reactor vessels have a surveillance programme to monitor the irradiation induced changes in mechanical properties of life limiting structural materials subjected to significant neutron fluence. The objective of the surveillance programme is to provide advance information concerning the state of degradation in mechanical properties of key structural components. For reactor pressure vessels this is generally achieved by determining the irradiation induced shift in Charpy V-notch transition temperature with reference to unirradiated archive material. The Charpy shift is compared with design predictions, regulatory guidelines and screening criteria to establish the margins and projected timeframe for safe operation of the reactor vessel. In the event that the data do not conservatively meet the design and regulatory criteria, the advance information provides the basis for planned remedial action in the form of changes to operating procedures or other corrective measures.

The remainder of this chapter describes the important features of current surveillance programme implemented in most Light Water Reactor (LWR) units and particularly in Pressurised Water Reactors (PWR's) constructed since about 1970 in the USA and countries of Western Europe, and, more recently in others throughout the world. Surveillance of the other main class of commercial reactors, gas cooled reactors mainly in the UK (Magnox), differs in certain respects and will not be dealt with in detail. The principal differences arise in the criteria for material selection, which is dictated by the very much larger number of welds employed in construction, the location of surveillance capsules and the wider operational temperature range (150-360°C) of the reactor vessel. These factors have necessitated a different approach to the interpretation of surveillance information and a less prescriptive, though no less rigorous, safety assessment philosophy.

The description of surveillance programmes is given below by considering the following topics successively:

- Principles of Surveillance Programmes;

- Initial materials characterisation;

- Materials selection;
7.4.1 Principles of Surveillance Programmes

Capsules containing specimens of specific reactor materials are placed inside the vessel, near the inner wall, before the plant starts to operate. The location of the capsules is closer to the core than the vessel wall so the neutron fluence received by the specimens is higher than that received by the vessel. Capsules are removed periodically throughout the life of the reactor such that the results of mechanical tests on irradiated specimens are in advance of (lead) the irradiation induced change in the behaviour of the vessel materials. The data from the surveillance programme are compared with the levels of embrittlement of the vessel predicted at the design stage and used to adjust the fracture toughness properties that are employed to assess the structural integrity of the reactor vessel. If the measured embrittlement exceeds the original design predictions or previous surveillance evaluations, a reevaluation of the vessel structural integrity must be performed taking into account the new irradiation embrittlement data or trend curves derived from the surveillance programme.

Following such a reevaluation, it may be necessary to modify the operation of the plant to reduce the severity of operational transients, such as thermal shocks, and additional measures may need to be taken to mitigate the irradiation damage to the reactor vessel (fluence reduction).

7.4.2 Initial Materials Characterisation

The surveillance programme includes a detailed description of the fabrication history of all beltline materials including austenitising, quench and tempering, welding parameters, welding qualification, acceptance tests of filler metals and post-weld heat treatment. In addition, it is also required that the reports contain the chemical analyses, Charpy data, tensile data, drop-weight data and initial RT_{NDT}. 
The chemical compositions of candidate materials for surveillance programmes are determined in the scope of acceptance tests on samples of actual materials of the RPV. It is required that those element, such as copper, nickel and phosphorus, which are known to increase the susceptibility of RPV steels to neutron embrittlement, be analysed for all beltline materials. The requirements on chemical analysis are generally extended, not only to other alloying elements, but also include a broad range of residual elements as for example sulphur, vanadium and gaseous elements (oxygen, nitrogen).

All material employed for the determination of pre-irradiation mechanical properties or machining specimens for surveillance capsules, as well as that kept for archive purposes, should be submitted to a heat treatment which is fully representative of the post-weld heat-treatment received by the reactor vessel.

The initial mechanical properties determined are the tensile characteristics, a full Charpy transition curve and the $RT_{NDT}$ reference temperature determined from drop weight tests.

### 7.4.3 Material Selection

Candidate materials for a reactor vessel surveillance programme are those located in the beltline of the reactor. The beltline is defined as that region of the reactor vessel which is adjacent to the reactor core for which one of the following criteria are met:

- the level of neutron fluence received at the vessel location is higher than a threshold value; this threshold of neutron fluence is $10^{22}\text{n.m}^{-2}(E>1\text{MeV})$ for [12] and as low as $10^{21}\text{n.m}^{-2}(E>1\text{MeV})$ for the other standard[13];

- the conservative prediction of the shift in $RT_{NDT}$ is higher than a prescribed value of $28^\circ\text{C}$[14].

From the materials located in the beltline region, those selected for inclusion in the surveillance programme are generally determined according to the following criteria:

- the adjusted $RT_{NDT}$ at end of life is determined by adding the predicted shift to the initial $RT_{NDT}$. 
- the selected base metal (plate or forging) is the one having the highest projected end-of-life $RT_{NDT}$;

- the heat affected zone (HAZ) of the latter base metal is also selected;

- the same criterion holds for welds, but, due to the large difference in neutron fluence between weld locations for current PWR designs, the weld joining the two core shells is quasi-systematically selected.

Deviations from this practice can be pointed out:

- the German standard[10] requires that material from both forged core shells be included in the surveillance programme, but such a practice could not be easily implemented for those reactor vessels that have core shells constructed of several different plates;

- a recent revision of the ASTM E185 standard[13] does not require HAZ material to be included in the surveillance programme.

7.4.4 Type and Location of Specimens

For each reactor unit, the surveillance programme monitors the irradiation behaviour of the selected base metal, HAZ and weld. A standard correlation monitor material is generally included in capsule loadings to aid interpretation in the event of apparently spurious results.

Three types of specimens are generally loaded into each surveillance capsule:

- tension specimens;

- standard Charpy-V notch specimens;

- small fracture toughness test specimens of compact tension type.

Figure 11 shows the different types of specimens and their orientations. The base metal specimens are removed at a quarter-thickness depth from the inside surface and weld specimens not less than 12.7 mm from the root pass. The major axis of the different specimens from base metal is oriented in a direction parallel to the surface
and normal to the principal rolling direction for plates or to the major working
direction for forgings (TL orientation).

The axis of the notch of the Charpy specimens or the fatigue pre-crack of compact
tension specimens is oriented perpendicular to the surface of the material both for
base and weld metals. For HAZ, only Charpy specimens are extracted with the notch
located at 0.9mm from the weld fusion line.

Most regulatory standards[12,13] require a minimum number of test specimens for
each irradiation exposure set (capsule). The minimum is generally 12 Charpy and 3
tension test specimens. In practice, not less than 15 Charpy specimens are employed
in order to reduce the uncertainty on the transition temperature determination.

7.4.5 Capsule Design, Location and Withdrawal Schedule

A typical standard capsule dimension is 25mm square, and approximately 1.5m long.
The capsule is generally manufactured from thin (0.9mm) austenitic stainless steel
sheet to protect specimens from corrosion at the operating temperature and to ensure
that the temperature of the specimens is close to the temperature of the coolant at the
inner wall of the reactor vessel. Specimens, dosimetry and temperature monitors are
inserted and the half capsules welded together. Finally, end plugs are welded on to
the capsule to provide a completely sealed volume.

Hydrostatic pressurisation is generally performed to collapse the capsule stainless
steel sheet onto the specimens in order to ensure optimum thermal conductivity
between the PWR environment and the specimens. Final inspection consists of a
helium leak test to verify capsule integrity. A typical design of capsule is illustrated
in Figure 12.

The capsules are often inserted in baskets which are attached to the core barrel,
thermal shield or attached by brackets to the inner wall of the reactor vessel. The
location is at mid-plane of the core and at angular positions selected with respected to
the peak neutron flux.

The "lead factor" is defined as the ratio of the neutron flux at each capsule location to
the maximum calculated neutron flux at the inner surface of the reactor vessel wall.
As the capsules are located nearer the core than the vessel wall, they receive a neutron
damage dose that is representative of the vessel at a later time of life. The lead factor
is usually within the range 1 to 3. A recent ASTM Standard[13] recommends that the lead factor should not be higher than 5, but other practices such as that in Germany[10] allow lead factors as high as 10 to 40.

Choice of lead factor is a source of considerable discussion concerning how representative the mechanical property changes are for specimens receiving highly accelerated dose rates. Additionally, there are concerns that highly accelerated surveillance may not reflect the changes in neutron energy spectrum and flux experienced by the vessel during subsequent changes in enrichment or management of fuel elements later in life.

The minimum number of capsules is determined as a function of the predicted maximum embrittlement of the selected material. For most current standards[ ] the number is never less than three and increases to four when the predicted end-of-life embrittlement is higher than 56°C. Four capsules is the current practice for PWR's of the 1990 generation. Earlier plants often have up to six capsules[15].

The withdrawal schedule generally conforms to the following pattern for monitoring neutron embrittlement:

- the first capsule is scheduled for withdrawal early in the vessel life to obtain a comparison of the surveillance material response to the actual radiation environment with design predictions. Practically, the results from the first capsule are available before the first hydrostatic pressure retest performed generally after 10 years of operation;

- the withdrawal schedule of later capsules is adjusted to obtain the expected end-of-life fluence in such a way that the results from the (n+1)th capsule will be available before the fluence at the vessel wall reaches that received by the nth capsule.

A typical schedule of capsule withdrawals is presented in Figure 13. It is apparent from the schedule that a comfortable time is available to carry out remedial actions in the event of unexpected embrittlement behaviour or, in favourable cases, to cater for the possibility of extensions to the reactor vessel life by the introduction of additional capsules.
7.4.6 Monitoring the Irradiation Environment

The irradiation damage dose (expressed as neutron fluence E>1MeV, E>0.5MeV or E>0.1MeV and dpa) experienced by the surveillance specimens is determined from measurements of the activity of dosimeters placed at reference points within the capsule. From these measurements the axial and radial neutron flux gradients within a capsule can be evaluated. This information is also important since it provides the principal source of data upon which updated vessel fluence estimates are made. Recently, ex-vessel or cavity dosimetry has been routinely performed to provide additional information in this regard, especially for those plant in which significant changes in fuel management or other fluence reduction methods have been adopted.

Two types of surveillance dosimeters, activation and fission monitors, are employed and these are inserted into holes in dummy specimens contained in the capsules. A list of recommended dosimeters and their characteristics are given in Table 1.

To provide a satisfactory exploitation of the measurements, it is recommended that:

- high purity materials should be used for dosimeters to avoid interference from activation of impurities;
- boron nitride should be used to improve the neutron filtering for the fission dosimeters;
- pure titanium should be used for the shell box of fission dosimeters instead of nickel.

An analytical neutron energy spectrum, sometimes modified to account for the dosimeter activities using an adjustment code, is employed to convert the measured response of the detectors to appropriate radiation damage units.

Temperature monitors are also located within the capsule to obtain estimates of the maximum temperature reached at each location during irradiation exposure. The monitors comprise low melting point eutectic alloys sealed in pyrex tubes. A list of possible detectors is given in Table 2. A set of three detectors having different melting points is recommended. The selection of the lowest melting point detector is made by choosing the alloy with a melting point just above 5°C of the highest calculated temperature reached along the capsule length. The location of the
temperature monitors is defined in a way to cover the entire length of the specimen assembly.

7.4.7 Evaluation of the Data

The irradiation effect is first qualitatively evaluated by determining the amount of irradiation strengthening by comparing unirradiated and irradiated tension test results. This provides an indication of the anticipated Charpy transition temperature enabling testing temperatures to be specified to provide the most efficient use of the limited number of available Charpy specimens.

The main embrittlement evaluation comprises a detailed analysis of the Charpy transition behaviour. This is achieved by comparing the unirradiated and irradiated transition temperatures at a specified fracture energy level (41J, 56J or 68J) or for a specified lateral expansion level (0.9mm) and deriving transition temperature shifts. These are compared with the Charpy shift predicted at the design stage or regulatory screening criteria in order to assess the relative embrittlement of the reactor vessel.

Some regulatory bodies, notably the USNRC[14], further require an assessment of the absolute level of Charpy upper shelf energy and the irradiation induced change in this parameter. Failure to meet the appropriate upper shelf energy criterion, 50ftlb (68J) in the case of USNRC, requires the plant operator to demonstrate equivalent margins of safe operation using conservative elastic-plastic fracture mechanics methods being developed for this purpose.

If the Charpy shift is less than predicted, the plant can continue to be operated without change to existing procedures. If not, a number of different actions may need to be carried out, depending on the magnitude of the deviation from the prediction and the withdrawal number under consideration.

In all cases, the scatter of the data has to be considered and, if necessary, additional examinations of the fractured surfaces of the Charpy specimens may need to be conducted in order to provide information on the origin of any special behaviour.

If the deviation happens early in the surveillance program and the safety analyses, taking into account the increased irradiation sensitivity, demonstrate that the end of life can be reached with reduced but sufficient margin, it would be prudent to await
the results of the next withdrawal and scrutinise them for confirmation of the unexpected behaviour.

When deviations occur in the later stages of the surveillance programme, other more directly relevant toughness evaluation methods may need to be considered such as additional results from supplementary fracture toughness tests of small irradiated specimens.

In cases where higher than predicted sensitivity to irradiation embrittlement is confirmed, it is necessary to consider modified operating conditions or the feasibility of present or future reduction in vessel embrittlement. When, on the other hand, the materials appear significantly less susceptible to irradiation embrittlement than expected, the possible benefits of reactor vessel life extension can be considered.

7.4.8 Guidelines for Remediation and Perspective for Plant Life Extension.

When evidence is provided by the surveillance programme that the actual radiation embrittlement exceeds earlier predictions and will necessitate a reduction of vessel life or significantly affect the possibility for plant life extension, several options are available and need to be assessed.

If the vessel is very much more strongly affected by the radiation embrittlement than expected, an annealing heat treatment of the vessel (around 450°C depending on the steel) is able to recover most of the initial fracture properties of the steel. A good example of such remedial action is provided by the recovery annealing of the Soviet VVER 440/230 reactor vessels, which has been successfully conducted on more than ten vessels prior to 1992[16].

When the need to improve the fracture toughness is not so stringent, it is possible to reduce the severity of some operational transients. For example, heating the emergency cooling water, used during loss of coolant transients, has been performed in some cases[17] and demonstrated to be an efficient action to provide the required safety margin in the event of such transients occurring.

A reduction in neutron flux received by the inner wall of the reactor vessel can also be employed in those cases where the current state of embrittlement is acceptable but the expected end-of-life embrittlement will exceed acceptable margins. Such a reduction could lower the future rate of embrittlement to levels which would permit
full life attainment and even life extension of the vessel. The flux reduction methods employed range from well controlled fuel management, in which almost fully burnt-up fuel is shuffled to the outside of the core, to the insertion of flux absorbers in critical locations adjacent to limiting welds. The latter can be dummy fuel assemblies, which brings a possible economic penalty by reducing reactor output, or absorbers located at the reactor vessel inner wall or thermal shield.

An optimisation of these different remedial actions is applied on a plant by plant basis. The surveillance programme may need to be modified as a result of these actions and the reassessed state of embrittlement of the vessel based on the findings from earlier withdrawals. This may necessitate changes in withdrawal scheme in respect of frequency or, in other cases, the insertion of new capsules using qualified archive materials or irradiated specimens reconstituted from earlier surveillance capsules.
References


Table 1
Characteristics of Recommended Dosimeters

<table>
<thead>
<tr>
<th>TYPE</th>
<th>REACTION</th>
<th>ENERGY RANGE (MeV)</th>
<th>PERIOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Activation</td>
<td>$^{63}$Cu(n,α)$^{60}$Co</td>
<td>6.1-11.3</td>
<td>5.27y</td>
</tr>
<tr>
<td></td>
<td>$^{54}$Fe(n,p)$^{54}$Mn</td>
<td>2.3-7.8</td>
<td>313d</td>
</tr>
<tr>
<td></td>
<td>$^{58}$Ni(n,p)$^{58}$Co</td>
<td>2.7-7.5</td>
<td>70.8d</td>
</tr>
<tr>
<td></td>
<td>$^{93}$Nb(n,n')$^{93m}$Nb</td>
<td>&gt;1</td>
<td>16.0y</td>
</tr>
<tr>
<td></td>
<td>$^{59}$Co(n,γ)$^{60}$Co</td>
<td>0.02-2.4</td>
<td>5.3y</td>
</tr>
<tr>
<td>Fission</td>
<td>$^{237}$Np(n,f)$^{137}$Cs</td>
<td>0.6-5.6</td>
<td>30y</td>
</tr>
<tr>
<td></td>
<td>$^{235}$U(n,f)$^{137}$Cs</td>
<td>1.5-6.7</td>
<td>30y</td>
</tr>
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</table>

Table 2
Melting Points of Various Low Melting Point Eutectic Alloys Used as Monitors of Maximum Surveillance Capsule Temperature

<table>
<thead>
<tr>
<th>EUTECTIC ALLOY</th>
<th>MELTING POINT (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PbAg1.7Sb6</td>
<td>263</td>
</tr>
<tr>
<td>Bi</td>
<td>271</td>
</tr>
<tr>
<td>PbAg1.9Sb5</td>
<td>272</td>
</tr>
<tr>
<td>PbAg1.9Sb4.5</td>
<td>273</td>
</tr>
<tr>
<td>PbAg1.9Sb4.3</td>
<td>278</td>
</tr>
<tr>
<td>PbAg2Sb4</td>
<td>280</td>
</tr>
<tr>
<td>PbAg2Sb3.5</td>
<td>284</td>
</tr>
<tr>
<td>PbAg2Sb3</td>
<td>288</td>
</tr>
<tr>
<td>PbPt5</td>
<td>290</td>
</tr>
<tr>
<td>PbAg2Sb2</td>
<td>293</td>
</tr>
<tr>
<td>PbAg2.5</td>
<td>304</td>
</tr>
<tr>
<td>PbAg1.75Sn0.75</td>
<td>308</td>
</tr>
<tr>
<td>PbIn5</td>
<td>314</td>
</tr>
<tr>
<td>PbZn0.5</td>
<td>318</td>
</tr>
<tr>
<td>Pb</td>
<td>327</td>
</tr>
</tbody>
</table>
Figure Captions

Figure 1 Typical simple rig for irradiating Charpy specimens in a pool type light water MTR.

Figure 2 Example of a more complex irradiation assembly designed to accommodate large CT specimens in the USNRC sponsored HSSI programme.

Figure 3 Schematic representation of the PLUTO Flux Converter irradiation facility.

Figure 4 Schematic of Reloadable Charpy specimen rig.

Figure 5 Detail of the specimen section of the Flux Converter Charpy Calibration Rig showing thermocouples leading to positions in and around the specimens.

Figure 6 Variation in activation monitor activation throughout a Flux Converter Charpy rig. Points designated SDM refer to standard capsules positioned at the end of assemblies of Charpy specimens. Points designated T1, T2 etc., M1, M2 etc. and B1, B2 etc. refer measurements from gradient wires inserted in the notches of Charpy specimens.

Figure 7 Schematic view of the ORR Pool Side Facility with exploded view of the capsule assemblies.

Figure 8 Exploded view of the Simulated Surveillance Capsule (SSC) in the PSF.

Figure 9 Exploded view of the Simulated Pressure Vessel Capsule (SPVC) in the PSF.

Figure 10 Schematic view of specimen and dosimetry loadings of the Surveillance and Surface capsules in the PSF. Note the extensive use of Charpy notch gradient strips and the utilisation of bolt holes for dosimetry capsules.

Figure 11 Location and orientation of surveillance programme specimens.

Figure 12 Surveillance capsule design and encapsulation of specimens.

Figure 13 Typical surveillance capsule withdrawal scheme.
FIGURE 1

CHARPY RIG.

- Titanium locating spring
- 2 Gas pipes
- Thermocouple T₁T₂
- Face adjusting screws
- Monitor hole
- Side adjusting screw
- Bottom gas pipe
- Reactor Lattice Plate Locator
- Reactor water
- Copper rig block
- Aluminium Tube (square) containing rig
- Corrugated Shim
- Charpy specimen 10mm x 10mm
- Centre block
- Monitor holes
- Suspension
- CHARPY RIG. block length 260 mm
- Gas pipes
Photograph of heater plate showing swaged, stainless-steel-clad, nichrome elements, prior to installation in the irradiation capsule.

Photograph of specimens after installation in the irradiation capsule.
Parameters of Flux Converter facility

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Irradiation volume</td>
<td>8.5 cm $\phi$ x 40 cm</td>
</tr>
<tr>
<td>Fast flux (&gt; 1 MeV) on sample</td>
<td>$5.7 \times 10^{16}$ cm$^{-2}$s$^{-1}$</td>
</tr>
<tr>
<td>Thermal flux on sample</td>
<td>$2.7 \times 10^{15}$ cm$^{-2}$s$^{-1}$</td>
</tr>
<tr>
<td>Nuclear heating in sample</td>
<td>0.5 W/cm$^2$</td>
</tr>
<tr>
<td>Converter power</td>
<td>212 kW</td>
</tr>
<tr>
<td>Useful fuel load</td>
<td>160 g</td>
</tr>
<tr>
<td>Usefu heat source</td>
<td>6-9 months</td>
</tr>
<tr>
<td>Coolant flow</td>
<td>3 l s$^{-1}$</td>
</tr>
<tr>
<td>Charpy size</td>
<td>10 x 10 x 55 mm</td>
</tr>
<tr>
<td>Charpy loading</td>
<td>32 samples</td>
</tr>
<tr>
<td>Operational temperature range</td>
<td>220-360°C</td>
</tr>
<tr>
<td>Specimen temperature variation within sample</td>
<td>5°C</td>
</tr>
<tr>
<td>Variation of mean specimen temperature within rig</td>
<td>9°C</td>
</tr>
<tr>
<td>Temperature control during reactor cycle</td>
<td>±5°C</td>
</tr>
</tbody>
</table>

FIGURE 3
To Rig Head (All Drawings)

Main body - specimen carrier assembly

Thermocouple retaining plates

Clamp plates

Section X - X

Charpy specimens

Thermocouples

Typical thermocouple position

Thermocouple groove

Flux monitor wire full length of notch

Thermocouple

Standard dosimeter capsule

Charpy specimens

Charpy specimens

Charpy specimens

Charpy Specimen Pack

Charpy cassettes (3 off)

"Nuclear heaters" (2 off)

Charpy "Cassette"

Charpy "Cassette"

Clamp faces

Rear location face

Bolted frame

Dosimeter capsule location

Gas gap

FIGURE 4
UK1 Mn 54 specific activity

Dosimeter identity and relative position in rig

FIGURE 6
FIGURE 7

Exploded View of the Simulated Surveillance Capsule, Simulated Pressure Vessel Capsule, and Void Box Relative to the ORR Poolside Facility.
Exploded View of the Simulated Surveillance Capsule (SSC).
FIGURE 9

Exploded View of Simulated Pressure Vessel Capsule (SPVC).
FIGURE 10

SURVEILLANCE (4x10^19 AND 2x10^19) AND SURFACE CAPSULES

SURVEILLANCE CAPSULE FISSION

IRON FILLER PLUG

LEGEND
BB - BACK BONE DOSIMETRY SET
DM - DAMAGE MONITOR
GS - GRADIENT SET
CC - COMPRESSION CYLINDERS
H - HARDNESS DISKS
TEM - TRANSMISSION ELECTRON MICROSCOPY DISKS
### Actual Years of Reactor Vessel Operation

<table>
<thead>
<tr>
<th>Capsules</th>
<th>ACTUAL YEARS OF REACTOR VESSEL OPERATION</th>
<th>LEAD FACTOR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Y</td>
<td>4</td>
<td>2.25</td>
</tr>
<tr>
<td>Z</td>
<td>9</td>
<td>1.96</td>
</tr>
<tr>
<td>V</td>
<td>10</td>
<td>1.79</td>
</tr>
<tr>
<td>U</td>
<td>17</td>
<td>1.58</td>
</tr>
<tr>
<td>W</td>
<td>Corresponding period of reactor vessel radiation exposure</td>
<td></td>
</tr>
<tr>
<td>X</td>
<td>Archive Material (End of Life Monitoring or Life Extension)</td>
<td></td>
</tr>
</tbody>
</table>

- Actual period of radiation exposure of the capsule
- Corresponding period of reactor vessel radiation exposure
- Insertion
- Withdrawal

**Figure 13**
8. EFFECTS OF IRRADIATION ON MECHANICAL PROPERTIES

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1 INTRODUCTION

This chapter deals with the effects of neutron irradiation on the mechanical properties of reactor pressure vessel steels. Neutron exposure is a growing concern for operating reactors because of the change in the strength and fracture toughness properties of the vessel wall materials. This change in properties is known as radiation embrittlement, and the long-term effects of neutron exposure could challenge the structural integrity of reactor pressure vessels. Thus, it is important to monitor and control the amount of radiation damage since the success of current industry efforts to assure full life time operation of nuclear plants depends importantly upon the long-term integrity of reactor pressure vessels.

In order to control the effects of irradiation, it is essential to have a good understanding of the mechanisms causing embrittlement in the plates, welds, and forgings that are used in fabricating reactor pressure vessels. Early vessels were constructed from moderate strength carbon steels. Newer vessels were made from manganese-molybdenum steels with the addition of nickel to increase strength. Unfortunately, all steels have the same basic response to neutron irradiation, that is, radiation hardens the steel causing an increase in strength which, in turn,
reduces the fracture toughness. This trend is common for ferritic steels that tend to undergo a classic ductile-to-brittle transition over a range of temperatures. Thus, it is appropriate to establish general trends in mechanical properties in describing the radiation embrittlement effects of vessel steels. Once the basic features of these effects are understood, differences in radiation sensitivity as the result of variations in chemistry and microstructure, flux and fluence, and irradiation temperature are examined.

Our knowledge of the effects of neutron irradiation damage comes from studies of unirradiated and irradiated materials in power and test reactors. Mechanical test specimens in these reactors have provided a significant amount of data for determining the relevant damage parameters as well as mechanical properties trends. Most of the data currently available are from Charpy V-notch and tension test specimens which are now used to measure the change in properties due to irradiation exposure. These data have been utilized to develop trend curve prediction methods for the embrittlement process which include the effects of variables such as copper, nickel, phosphorus, and neutron fluence in a given temperature range.

While not all reactor vessel steel types are covered by this chapter the majority of those in light water reactors are covered by emphasis on the types of steels and vessels used in the USA and the former USSR. Variations of these steels are used in Europe and Japan, however the extent of these variations are such that the conclusions of this chapter are still valid since the nature of the
steels used throughout the world for reactor vessels are similar to those discussed here.

2: NEUTRON IRRADIATION DAMAGE

2.1 How does irradiation exposure cause damage?

Research to understand the mechanisms of neutron irradiation embrittlement in vessels fabricated in the United States has resulted in improved radiation damage trend curves as given in USNRC Regulatory Guide 1.99, Revision 2 [1]. Similarly, trend curves have been developed in the former USSR [2,3] for assessing the radiation damage in nuclear vessels. The basic mechanisms of embrittlement can be described as follows:

1. Embrittlement is primarily due to microstructural changes which result in irradiation-induced increases in yield strength which can be quantitatively related to changes in the Charpy V-notch curve, as well as hardness.

2. Yield strength increase-related changes in fracture properties are due to irradiation-induced fine scale microstructures (about 1 nm) which act as obstacles to dislocation motion.

3. The most likely resultant microstructures are precipitates, vacancy clusters (micro-voids), and interstitial clusters (dislocation loops). Copper-rich precipitates, possibly alloyed with elements such as nickel (or combined with vacancies), are believed to be a dominant hardening (strengthening) feature in sensitive steels containing significant concentrations of these impurity elements. Other possible types of irradiation-induced or
enhanced precipitates are phosphides and small carbides.

4. Other important effects of irradiation are enhanced diffusion rates and defect clustering.

5. These microstructural changes are kinetic phenomena and are functions of neutron exposure and temperature as well as composition and initial steel microstructure. In general, the changes in the microstructure can be understood from basic principles of alloy thermodynamics and precipitation kinetics, coupled with rate theories of radiation damage.

A noticeably greater difference in radiation sensitivity has been observed in weld materials as compared to plates and forgings for approximately the same chemistry and neutron exposure. While trace amounts of copper are the primary contributor to irradiation embrittlement in USA steels, it has also been discovered that nickel in the presence of copper tends to increase irradiation sensitivity, particularly in the case of welds. This observation has led to physically-based models of the embrittlement process which are slightly different for welds and base materials. Test data from the former USSR and European plants exhibit other effects due to the presence of higher concentrations of elements such as phosphorous and vanadium. These effects are discussed separately.

2.2 What properties are affected?

Embrittlement due to radiation hardening can be quantitatively related to changes in the yield strength following neutron exposure.
This hardening effect can be measured in tension tests and can also be inferred (but not directly quantified) using hardness (or micro-hardness) test techniques. The embrittling effect of irradiation on reactor vessel steels is a complex process, and numerous theoretical studies, testing programs, and microstructural evaluations have been performed to better understand the structure-property correlation. The changes in microstructure described above have been confirmed using atom probe/field ion microscopy (APFIM), small angle neutron scattering (SANS), and high resolution transmission electron microscopy (TEM). These studies have revealed radiation-induced precipitation or clustering of copper, nickel, and phosphorous on a very fine scale. As a result, the more recent models of radiation embrittlement include a dispersion strengthening component based on the age-hardening model developed by Russell and Brown [4] which has been modified to account for the increased diffusion rates of vacancies due to neutron bombardment. Some researchers now believe that age-hardening is the dominant process of radiation embrittlement in vessel steels [5, 6].

The age-hardening effect can be measured by changes in the mechanical properties. In fact, good correlations have been observed between tension tests and Vickers microhardness test data on reactor vessel welds. The changes in ductility and fracture toughness are important measures of the material's ability to resist fast fracture, but these properties
are difficult to measure directly. While hardness is not the critical material property, it is a convenient means of monitoring embrittlement, and a method to correlate with other key changes in mechanical properties. Our ability to model the embrittlement process has led to an improved understanding of how steels are damaged by neutron irradiation, but as yet we cannot predict changes in fracture toughness solely from a mechanistic standpoint. These property changes must be measured using more expensive larger size specimens such as used in Charpy V-notch tests or even larger ones such as those used in compact fracture toughness tests.

2.3 How is radiation damage measured?

The fracture toughness of carbon steels is very temperature-dependent as exhibited by a marked change in fracture behavior from brittle (cleavage) at low temperature to ductile (tearing) at higher temperatures. Characteristic changes in fracture toughness from low to high values with increasing temperature are exhibited. This transition behavior is most apparent with a test specimen containing a sharp notch or crack. The most common specimens used to monitor the effects of radiation embrittlement are the Charpy V-notch (ASTM E370-88a) and the drop weight tests (ASTM E208-87a). The Charpy test measures the energy required to produce fracture of a notched three-point bend specimen. Charpy data as a function of temperature can be obtained for unirradiated and irradiated pressure vessel steel materials.

The
embrittlement effects are noted as an upward shift of the Charpy curve in temperature and a reduction of the maximum upper shelf energy at the higher temperatures. In the U.S., the extent of radiation damage is determined by the shift in the 30 ft-lb (41 Joule) Charpy energy level frequently referred to as the transition temperature shift. This shift is important as it relates to the change in fracture toughness ($K_{IC}$) of the steel in the irradiated condition which exhibits the same kind of transition temperature shift. The change in the toughness curve due to irradiation must be taken into account when establishing safe operating limits to prevent brittle fracture of the vessel. For obvious reasons of reactor vessel integrity, the fracture toughness of the material is the most important mechanical property. Unfortunately, it is difficult to determine vessel toughness directly since "valid" fracture toughness measurements require large specimens at the temperatures of vessel operation.

It is valuable, even essential, to correlate changes in fracture toughness with other, more easily measured, mechanical properties. For example, Charpy specimens are routinely used in surveillance programs to monitor the effects of embrittlement on mechanical properties with the assumption that the change in fracture toughness transition temperature is equivalent to the change in Charpy temperature shift. Based on the work of Cottrell, it was recognized that the transition temperature shift as the result of radiation damage should be directly proportional to the change in yield strength. In fact, a robust correlation between
yield strength increase ($\Delta \sigma_y$) and transition temperature shift ($\Delta T$) has been observed [9] which follows the simple form:

$$\Delta T = C \sigma_y$$

For U.S. reactor vessel welds, the correlation factor $C$ was determined to be 0.65°C/MPa based upon the existing U.S. surveillance database. A similar analysis for plate materials yielded $C = 0.5$°C/MPa. This correlation provides evidence that changes in hardness can also be related to transition temperature shift. Caution must be exercised whenever using such correlations since they may be only applicable to specific environmental parameters (e.g., temperature and fluence rate) and specific material types (e.g., welds versus base metal or even specific material types within welds or base metals). This correlation process has been utilized in the former USSR to measure recovery in toughness properties following reactor vessel thermal annealing by performing in situ hardness measurements before and after the anneal.

Thus, it is apparent that small-scale testing techniques can be used in conjunction with larger specimens to monitor the effects of embrittlement on reactor vessel steels so that the contributing factors to irradiation damage can be accurately assessed.
2.4 What are the key factors affecting reactor vessel embrittlement?

The results from the many test programs and mechanistic modelling studies identify three key factors which affect the radiation damage of reactor pressure vessel steels. These factors include the neutron environment (fluence, fluence rate, fluence energy spectrum) and the irradiation temperature. The other key factor is the material itself which can be different in chemical elements, microstructure, and processing history. The time integrated neutron flux exposure results in continuing loss in toughness and other changes in mechanical properties for most pressure vessel steels until, in some cases, a saturation level is reached. The degree of radiation damage depends largely on the particular materials involved, especially their chemistry and microstructure, plus the specific neutron environment. The effects of these factors will be described in the next part of this chapter with emphasis on how mechanical properties change based upon the neutron environment and materials. These differences will be highlighted based upon results obtained primarily in the USA and the USSR. The design of reactor vessels along with the size and placement of the core are crucial to radiation effects.

3 USA VESSEL DESIGN AND FABRICATION

Essential to the understanding of the background within which vessel steel properties are changed by neutron exposure is a description of the design and fabrication of the vessel.
Consequently a summary of these features for both USA and former USSR reactors are provided.

3.1. USA Vessel Design

Most reactor vessels now in service in the USA were fabricated in the 1960s and 1970s. A general description of these vessels is outlined in Reference [10].

Table 1 presents typical sizes and weights of LWR pressure vessels for plants in excess of 1000 MW of electric power capacity. The boiling water reactor (BWR) vessel is much larger, and somewhat heavier, than the pressurized water reactor (PWR) vessel. The direct-cycle BWR vessel contains not only the reactor core, but also the steam separators and dryers, and the feedwater spargers. In addition, most domestic (USA) BWR vessels accommodate internal jet pumps, which are part of the coolant recirculation system. The BWR typically operates at 1000 psi (6.9 MPa) and <550°F (<290°C).

The PWR system uses external steam generation and separation, so the PWR vessel is much shorter than the BWR vessel. The two types of vessels are shown schematically in Figure 6 [11]. The weld locations in a typical PWR [10]. The PWR operates at a higher pressure, typically 2250 psi (15.2 MPa) and in a temperature range of 520-600°F (270-315°C). The PWR wall thickness is typically about 10 in. (250mm), while the BWR wall thickness is about 6 in. (150mm).
3.2 Reactor Vessel Fabrication in the USA

Similar techniques are used to fabricate BWR and PWR pressure vessels, and also PWR steam generators and pressurizers. The PWR vessel can be constructed entirely from forged rings, flanges, and nozzles. Alternatively, rolled and welded plate can be used for the shell courses, and forgings for flanges and nozzles. All BWR vessels have used the latter construction method, with shell courses of rolled and welded plate.

The reactor vessel manufacturer typically receives steel plate in the as-rolled and stress-relieved condition. The steel making and hot rolling of the plate is not discussed here. The first step in the fabrication sequence is to hot-form the plate into 120° steel segments. Hot forming is performed at a temperature of approximately 1650°F (900°C). The formed segment is austenitized at 1600°F (870°C) followed by a water quench. The quenching treatment is followed by a tempering treatment of 1250°F (675°C) for two hours. Three 120° segments are then welded together onto a shell course using a welding procedure to be described later. The shell courses are clad with an austenitic stainless steel weldment using either the multiple wire or strip cladding submerged arc process. Three shell courses make up the cylindrical portion of a typical PWR vessel.

The upper (or nozzle) shell course is generally 20% to 50% thicker than the intermediate and lower shell courses. The additional thickness is required to reduce the stresses associated with nozzle penetrations. Six to eight nozzle forgings are welded
into large diameter holes (60 in., or approximately 1.5 m) in the nozzle shell course. The vessel flange is welded to the top of the nozzle shell course, and the intermediate shell course is welded to the bottom of the nozzle shell course, forming one of two vessel assemblies.

The second vessel sub-assembly consists of the lower shell course and the bottom head. The final steps are to weld the upper and lower vessel subassemblies together and to stress-relieve the assembly.

A forged vessel is constructed in a similar fashion, except that the shell courses are one-piece ring forgings. This construction avoids the longitudinal beltline region welds.

It generally requires between three and five forged rings to construct a reactor vessel. The forging procedure for such a ring is described next. The ingot is cast and the upper and lower ends are discarded. The remainder of the ingot is repeatedly upset in a large hydraulic press until a disk approximately four to five feet thick is prepared. This disk is pierced, and the pierced disk is placed on a mandrel and continually worked in a large hydraulic press until the shell forging is formed. The finished forging is austenitized at 1650°F (900°C) for two hours. The ring is then quenched and tempered at 1275°F (690°C). As with the plate-formed vessel courses, the forged ring must be clad with austenitic stainless steel to prevent general corrosion.
3.3 Material Specification

Reactor vessel shell plate specifications have evolved since the beginning of the commercial nuclear power industry in the mid-1950s. Table 2 [10] lists the principal steel plates used in construction of nuclear pressure vessel components. The original steels, the A212 as well as the A302B, were in widespread use in the construction of fossil power plant components at that time. All of the steel plates are of the low-alloy ferritic variety. The "A" designation indicates the ASTM material specification. The "S" prefix indicates material acceptable by the ASME Boiler and Pressure Vessel Code for construction of power plant components. The A212 steel was used only in very early plants which are now decommissioned. The current equivalent of this carbon-manganese-silicon steel is SA 515 Grade 60. A much more widely used material, A302B, is a carbon-manganese-molybdenum steel that was used in the quenched and tempered condition. In the mid-1960s, with the size of the nuclear components increasing, greater hardenability was required. The addition of nickel in quantities between 0.4 and 0.7 wt% provided the necessary increased hardenability to achieve the desired mechanical properties. This steel was known initially as SA 302B Modified. Later, it became the present grade SA 533 Grade B Class 1, which is the most widely used material for construction of reactor pressure vessels and pressurizers in the USA.

In 1973, limits were imposed in the percentage of copper and phosphorus permissible for use in the so-called beltline region of
reactor pressure vessels where the neutron flux is high. The reduction of copper and phosphorus minimized the embrittlement sensitivity of the steel.

The last entry in Table 2 is SA 533 Grade A which is used in the construction of the steam generator shell. This material is also used in the quenched and tempered condition, and is the current equivalent of the original A302 Grade B. Typical heat treatments of these materials are as follows: The material is austenitized at a nominal 1600°F - 1650°F (870°C - 900°C) followed by quenching, generally in an agitated water bath. The material is then tempered at 1220°F - 1250°F (660°C - 675°C) for two hours. After welding, the entire vessel is stress-relieved at 1150°F (620°C). Typical ranges on these heat treatment temperatures are ±25°F (±14°C).

The pressure vessel forging materials (Table 3) have also evolved since the mid-1950s. The earliest grade, A105 Class 2, was used in the normalized and tempered condition for flanges and nozzles. This simple carbon-manganese steel has been used to a very limited extent in vessels. A forging material of greater usage in the 1950s and 1960s was the SA 182 F1 Modified, used in the quenched and tempered condition. This manganese-molybdenum-nickel steel was used principally for flanges and nozzles. Another forging material in use at this time is a carbon-manganese-molybdenum steel, SA 336 F1, used in both the normalized and tempered, and the quenched and tempered conditions.

One of the production problems encountered with the SA508-2
forging material is the occurrence of small underclad cracks with certain cladding procedures. In the early 1970s, typical European practice was to apply cladding in a strip fashion using the submerged arc process. Small cracks on the order of several millimeters in depth occurred with great frequency in some of the heat-affected zones of strip-clad welds. It was eventually discovered that the presence of chromium in this forged material was the root cause of the cracking. Such underclad cracking has never been observed in the SA 533 Grade B Class 1 plate material or the submerged arc weld metal.

To eliminate underclad cracking, SA 508 Class 3 material is used in place of SA 508 Class 2. The Class 3 material is essentially the same as the earlier SA 182 F1 (Modified) specification. The industry appears to have come full circle with regard to forging materials. Hydrogen blistering is no longer a problem in SA 508 Class 3 material.

3.4 Welding Procedures in USA Reactors

Full thickness welding is required to assemble the shell courses, the nozzle forgings, the flange forgings, the top and the bottom heads, and any internal or external support pads. Table 4 lists typical welding techniques used in the construction. By far, the most frequently used technique is the automatic submerged arc welding procedure. The protective environment is provided by a granular flux, part of which is vaporized during welding. The weld wire has the required alloy content. The only heat treatment
required is the normal stress relief, which reduces residual stresses and tempers the martensite found in the heat-affected zone. Automatic submerged arc welding is used wherever possible because it provides excellent mechanical properties and has a very high deposition rate relative to other welding techniques. A tandem wire technique may be used to increase the deposition rate.

Narrow gap submerged arc welding is a variant of the technique used primarily in the fabrication of circumferential or girth seam welds. The benefit of the narrow gap technique is reduced weld volume and fabrication time.

A frequently used manual welding technique is the shielded metal arc procedure, which is used for complex configurations, for repairs of base material, or possibly for areas of weld buildup. Although the deposition rate is low, the technique is extremely flexible and the weld metal has excellent mechanical properties.

The electroslag technique was used for full thickness welds in some of the earlier BWR pressure vessel and PWR steam generator shells. This automatic procedure provides extremely high deposition rates. Due to its large coarse cast microstructure, electroslag welds must be austenitized, quenched, and tempered similarly to the treatment for base metal. Unfortunately, electroslag welding requires such close dimensional tolerances on weld preparations that the technique is often not cost-effective.

All interior surfaces of the PWR vessel are clad with austenitic stainless steel to inhibit general corrosion and the buildup of radioactive crud. BWR vessels are clad below the steam-
water interface. Three principal types of cladding processes are used. The shielded metal arc process is used wherever possible due to its high deposition rate. The process uses either multiple wires of strip electrodes of Type 308 or 309 stainless steel. In areas where an automatic process is not possible, shielded metal arc or gas tungsten arc welding is utilized.

3.5 Heat Treatment in USA Reactors

The heat treatment of the base material has been previously discussed. The postweld treatment is described here. Following any welding procedure, such as the through-thickness weldment and cladding procedure, or even a small repair, an interstage stress relief is required before dropping preheat. Generally, this requires exposing the welded component to 1130°F (610°C) for approximately one hour. When the vessel is completely assembled, the entire unit is postweld stress-relieved. Typically, the reactor vessel would be heated to 1130 ± 25°F (610 ± 14°C) for approximately one hour per inch of thickness. The total stress relief time of a typical reactor vessel, including the final as well as the intermediate stage stress relief, is typically 20 to 25 hours. Due to the uncertainty of this time, the qualification test for the reactor vessel materials may require that all materials receive a simulated postweld heat treatment of 40 to 50 hours. This stress relief time for reactor vessels can increase if the unit is subjected to a number of repair cycles.
4 GENERAL INFLUENCE OF NEUTRON EXPOSURE ON USA VESSEL STEELS

4.1 USA Background on Radiation Damage Assessment

Ferritic materials are susceptible to radiation-induced embrittlement. The beltline region of a PWR receives fluence ranging from $9 \times 10^{18}$ to $4 \times 10^{19}$ n/cm$^2$ (E $>$ 1MeV) during its design life. This level of exposure to fast neutrons can cause significant reduction of fracture toughness. The embrittlement sensitivity of a particular heat or weldment depends on the presence of trace elements, particularly copper, in the alloy. The safe operating life of some reactor vessels, and the justification for full life attainment or extension, depends on a prediction of the adequacy of fracture toughness for the specific heats of material in the beltline region of a reactor vessel at a specified future date.

A Reactor Vessel Surveillance Program (RVSP) is conducted for each vessel to monitor radiation damage using Charpy V-notch, tensile, and in some cases, fracture toughness specimens. Statistical models have been developed to relate such data, and these statistical models are thus related to static, dynamic, and crack arrest fracture toughness. Interpolation schemes are used to predict radiation damage at locations where flux, temperature and even microstructure differ from the RVSP test specimens.

In the period from the mid-1950s to 1972, there were no limits imposed on the presence of copper. Although copper is not an alloying element, small amounts of copper (up to 0.4 wt%) were present as a "tramp" element. In the plate and forging materials,
the origin of the copper is traceable to the automotive scrap used in the production of ingots. It is impossible to remove all electrical, hence copper, wiring from the automotive scrap. The base material for the submerged weld wire has always been very low in copper content; however, copper was intentionally plated on the wire to minimize oxidation of the wire during storage. Also, copper plating provided the additional benefit of improved electrical conductivity in the welding head. This copper plating was standard practice for ferritic weld wire. In the period of 1950 to the mid-1960s, large research programs were introduced to evaluate the impact of fast neutrons on mechanical properties of pressure vessel material. During this period of time, it was observed that there was a large variability of radiation embrittlement sensitivity. In the late 1960s, the element copper was determined to be the prime contributor to this variability.

Experiments designed and conducted in the late 1960s and early 1970s confirmed the fact that copper as a trace element was a primary contributor to radiation embrittlement sensitivity. It was also believed that phosphorus, an element well-known for its deleterious effects in other embrittlement mechanisms, was a contributor to radiation embrittlement, and this was also confirmed. Responding quickly to these observations, limits were imposed by the ASME Boiler and Pressure Vessel Code on the level of acceptable copper and phosphorus in those materials exposed to significant fast neutron bombardment. Consequently, all reactor vessels made since 1972 have significantly reduced embrittlement
sensitivity.

In the mid- to late 1970s, a greater radiation embrittlement database was developed. Careful scrutiny of data indicated that accounting for copper did not completely account for the variability of observed sensitivity. In the late 1970s, it was suspected that nickel, a close relative to copper on the periodic table, was a secondary contributor to radiation embrittlement. It should be remembered that nickel was intentionally added as an alloy element in the mid-1960s to improve the hardenability of the base materials and also to improve the initial mechanical properties of the weldments. In the 1980s, nickel was, in fact, confirmed as a secondary contributor to the embrittlement of steels, particularly in combination with copper. The effects of both copper and nickel are recognized in terms of producing small microvoids or copper-nickel precipitates in the presence of neutron flux. These act as the primary source of embrittlement. Improved radiation damage trend curves have been developed using these physically based models and data from reactor surveillance programs. These improved radiation damage trend curves provided the basis for Regulatory Guide 1.99, Rev. 2 for calculating vessel \( RT_{\text{NDR}} \) (nil-ductility transition temperature), and the implementation of the Regulatory Guide has had an impact on the plant operating pressure and temperature limits which vary from plant to plant.

4.2 Neutron Effects in Light Water Reactors

The nature of neutron irradiation effects for the temperature
range of operation in light water (LWR) reactors and for the types of ferritic steels used in "western" Boiling and Pressurized Water Reactors (BWR and PWRs) is essentially the same. (It should be noted that the PWR is the most widely used system of all LWRs.) However, the significant factor of change (usually defined as embrittlement) varies between BWR and PWR types with the PWR vessel showing more embrittlement. Unfortunately, many early commercial reactors in the "west" must be treated as "individuals". Nevertheless the design of the two types of reactors (e.g., the water gap) explains the greater fluence of energetic (>1 MeV) neutrons impinging in the PWR vessel wall. This generally higher level of neutron exposure dictates that the level of embrittlement is greater in the PWR. Further, the nature of the composition and microstructure leads to greater sensitivity to damage. (Composition differences in WWER reactor vessels are shown in Tables 6 and 7.) Consequently, following the noted development of a statistical body of data from surveillance, measured changes in fracture toughness provide the bases for the widely used U.S. Nuclear Regulatory Guide 1.99, Rev. 2 trends which often are used to project both the vessel embrittlement condition and the response to a pressurized thermal shock (PTS) event.

The most significant effects of neutron fluence on mechanical properties of reactor vessel steels have led to a general view of these effects which are affected by the level and energy of neutron fluence accumulation over time, the specific composition and microstructure of the steel exposed and the temperature during
exposure. Some data indicate, as well, that the rate of exposure to the number of energetic neutrons bombarding the steel each second may affect the ultimate changes in critical mechanical properties for some materials in certain ranges. The most significant of these properties include: strength (yield strength increases along with ultimate tensile strength), notch toughness (the crucial measurement) decreasing in terms of Charpy V-notch (Cv) curve position and shape (with a shift to higher temperatures and a lower ductile shelf level), and static (Kic) and dynamic (KId) fracture toughness levels which unfortunately must be deduced from Cv data since valid Kic and KId numbers currently can only be determined using specimens too large for the surveillance programs. Accordingly, the strength and toughness measures which are applied to assess fracture potential or to judge structural integrity must be deduced from small surveillance specimens. Such knowledge combined with that of steel composition, exposure temperature, flaws present (and stress levels at these flaws) are essential to assessment of vessel integrity under normal and accident conditions.

4.3 Tensile Properties of Irradiated Steels

Neutron radiation effects were first identified as radiation hardening and strengthening. Casual interpretation of this change would suggest a good (or positive) effect on the key properties of pressure vessels steels. However, both engineering and fundamental analysis of these increases in tensile properties carried to the
conditions of pressure vessel service can be interpreted as potentially detrimental.

First, while both yield and ultimate strength are raised by neutron exposure, yield strength is increased more rapidly to the point that it may in some cases approach the ultimate strength. This increase then limits the degree of uniform elongation possible thereby creating a "less forgiving" medium under conditions which could be serious to the integrity of a potentially flawed vessel since elastic deformation is reduced. Thus, there is less tolerance for correction in a deformed and flawed vessel if an "overpressure" transient should occur. This projection may be extended to the potential ductile rupture or brittle fracture of a residual (unruptured) section coupled with a rapid separation in the vessel. This scenario must be avoided at all costs in the primary system nuclear pressure vessel.

Fundamentally linkage of the radiation strengthening and potential fracture can be described as follows: microscopically it is possible to describe the role of radiation produced defects as barriers to dislocation motion which results in hardening and strengthening of the irradiated material. The increase in the yield strength resulting from the presence of defect aggregates has been shown to be temperature independent in irradiated iron. This temperature independent increase in yield stress can in turn be used to explain the shift in transition temperature as shown schematically in Figure [11]. If the fracture stress is assumed to be roughly temperature independent and the intersection of the
fracture stress curve and the flow stress curve is taken as the brittle-to-ductile transition temperature for the unflawed condition, an increase in flow stress produced by irradiation can be shown to shift the point at which the fracture stress and flow stress curves intersect and increase the brittle-to-ductile transition temperature.

Tension tests provide measurements of yield stress, ultimate tensile strength, percent elongation, and percent reduction in area. The change in yield strength is usually considered to be the most sensitive property to neutron irradiation. Trends for these properties have been observed from various national and international programs [11].

Yield strength increases after neutron irradiation of advanced pressure vessel materials are shown to provide results which follow a pattern that is most similar to the shift in Charpy transition temperature when plotted against neutron fluence. Such curves are usually described by a relationship

\[ \Delta \sigma_{0.2} = A (\phi t)^n \]

where \( A \) and \( n \) are constants and \( \phi t \) is the neutron fluence in terms of \( E > 10^{19} \) n/cm². The increase in yield strength for a number of advanced steels is given by

\[ (\Delta \sigma_{0.2})_{20^\circ C} = 44.24 \ (\phi t)^{0.356} \]

As will be seen later, the value for the exponent, \( n \), for the Charpy transition temperature shift may be as high as 0.5; there is, however, a larger scatter in the results which may arise from tension test variables as well as from scatter from heat-to-heat
variations in the steel. A comparison of the change in yield strength with irradiation between a reference steel (HSST-03) and several advanced steels shows that irradiation of the older HSST-03 steel induces a larger increase in yield strength at lower neutron fluences, and an increased sensitivity with increasing neutron fluence.

4.4 Changes in Transition Temperature (Fracture Potential)

The ferritic low alloy steels most widely used for reactor pressure vessels commonly exhibit the phenomenon of ductile-to-brittle transition over a relatively narrow temperature range (about 100°C or less). This range involves a transition from ductile (fracture only under conditions of plastic overloading or tearing) to brittle (fast fracture under elastic loading conditions) as the temperature is reduced. Over this range the micro- and macroscopic nature of the fracture gradually changes from ductile dimpled rupture, with attendant plastic deformation of the adjacent matrix, to cleavage along crystallographic boundaries. There is a broad range of mixed fracture modes between these points. Two distinct interrelated patterns of embrittlement are definable: a) the very distinct increase in the ductile-to-brittle transition temperature (ΔT) and b) the less predictable but important reduction in fracture resistance known as the ΔUSE (reduced upper shelf energy). A crucial development of the former is the relative equivalence in the measured NDT and the Charpy V-notch transition temperature after irradiation. This fact made
possible the $RT_{NDT}$ (reference transition - nil ductility temperature) which is initially defined at the 50 ft-lb (68J) level in combination with the measured NDT and adjusted by the 30 ft-lb (41J) shift after irradiation and forms the principal criterion for defining the vessel's embrittlement condition.

The design and operation of nuclear reactor pressure vessels includes the consideration of radiation-induced embrittlement of ferritic, low alloy steels in the fast neutron fluence range of $10^{18}$ to $10^{20}$ n/cm$^2$ for energies (E) greater than 1 MeV. The effects of neutron irradiation are manifested principally by an increase in yield and ultimate tensile strengths and decrease in fracture toughness. The change in toughness generally is monitored by using Charpy V-notch ($C_{v}$ or CVN) test results as a function of temperature for specimens that have been irradiated in surveillance capsules. The results from irradiated CVN testing show both an increase in the ductile-to-brittle transition temperature measured at the 30 ft-lb (41J) energy level and a drop in the upper shelf energy (USE).

Both the shift in $T_{30}$ (41J) and the drop in USE are key regulatory issues for reactor pressure vessels operation. Ideally, the shift in measured linear elastic fracture toughness ($K_{IC}$) as well as the drop in measured upper shelf $J$-resistance fracture toughness should be used, but surveillance programs are generally limited to only a few small fracture toughness test specimens usually compact fracture specimens with thicknesses less than 1-in. (25 mm). Recent developments have enhanced the potential for use of such "quantitative" specimens in
irradiation studies.

4.5 Upper Shelf Toughness

Since the ideal J-R tests are not acceptable for direct use in vessel surveillance, USE values or USE limits are imposed to avoid conditions for ductile tearing of a vessel ligament. Under USA Federal Code 10 CFR 50, Appendix G, a minimum upper shelf Charpy impact energy requirement of 68 J (50 ft-lb) (for specimens with transverse TL orientation) is specified. Further, this code requires notification to the US NRC three years in advance of the date when it is estimated that the 50-ft-lb criterion will be transgressed. If data fall below 50ft-lb level, then the completion of the following three steps is required:

1. A 100% volumetric inspection of the reactor beltline region materials whose USE is less than 50 ft-lb following the requirements of American Society of Mechanical Engineers Section XI [12]

2. Supplemental tests to provide additional evidence of fracture toughness changes in the beltline region materials (i.e. test reactor irradiations of archive material tested to validate the adjusted K_{IR} curve).

3. A fracture mechanics analysis that conservatively demonstrates that adequate margin still exists for continued safe operation; this requirement sometimes can be used without the inspection and toughness results (1 and 2 above) if
conservative estimates of both are used and an adequate margin is easily justified.

The first step may require a revision of the inspection schedule. The second step is difficult because most surveillance capsules do not include fracture mechanics specimens; however, correlations between USE and fracture toughness have been developed. Some surveillance capsules do include small fracture mechanics specimens, but adequate testing techniques for standardization of these specimens are still under development. Therefore, characterization of the upper shelf toughness is a major problem. Guidelines and requirements for a plant-specific analysis to assure safe continued operation when a plant has violated the 50 ft-lb level have only recently been developed through ASME Code Section XI activities. US Federal Code 10CFR50 indicates that thermal annealing of affected beltline material is an acceptable approach for restoring toughness properties. In the USA the number of older plants facing USE limits is significant so that a quantification of these effects or plant operational implications of these effects becomes urgent for a number of older plants.

4.6 Other Factors Affecting Mechanical Properties of USA Vessel Steels

The microstructure of reactor vessel alloy steels generally determines its basic material properties. Several types of microstructural imperfections can be created by irradiation,
resulting in changes in tensile and fracture properties. Examples of these microstructural changes are: clustering of vacancies (empty lattice sites) clustering of interstitials (extra atoms forced between lattice sites), forming microvoids, and forming dislocation loops. These imperfections can be created when high energy neutrons collide with individual atoms, knocking some of them out of their lattice positions. Because of the relatively slow speed of the displaced atoms and their uniform size, the displaced atoms interact with other lattice atoms in a billiard ball manner, creating what is called a cascade phenomenon. Once the cascade has dissipated, local zones of depleted atoms draw interstitial atoms by diffusion through the metal. The interstitials and microvoids act as barriers to dislocation movements.

Another example of microstructural changes that can occur during radiation exposure is irradiation-enhanced precipitate formation and alteration. Detailed characterization of these actions is difficult because of the small precipitate sizes (1 to 2 nm). However, research has indicated that precipitation of small, complex copper-enriched clusters and their interactions with dislocations may be a dominant embrittling mechanism in most radiation-sensitive vessel steels. Increased vacancy population created by neutron irradiation can enhance the copper precipitation process at PWR operating temperatures. The thermodynamic situation created by irradiation drives the system away from normal equilibrium and as such is strongly dependent upon the displacement
damage rate and the irradiation temperature. Precipitation influences with other elements such as nickel and phosphorus are also important.

Other factors than microstructure and composition affect the nature and extent of properties changes in a nuclear environment. Key among these factors are exposure temperature, radiation fluence, and possibly flux. A general pattern of the temperature effect is illustrated in Figure 1 [11]. Of course, effects resulting from copper content may dominate the physical environment.

While the large reductions of $\Delta T$ suggest thermal annealing as a solution, the implications in terms of the general vessel's fracture toughness, both $\Delta T$ and $\Delta USE$ requires a more quantitative general approach using fracture mechanics parameters ($K_{IC}, J_{IC}$) and $T$). However, serious limitations exist at this time since, as noted, earlier; larger specimens than can be accommodated in vessel surveillance programs are required. The solution then has been to test large $K$ and $J$ specimens in non-power reactors and thereby establish, by correlation, limit curves which can be applied using smaller surveillance specimens. The combination of a carefully established $K_{IC} - K_{IK}$ curve (collectively called the ASME Code $K_{IR}$ limit curve) coupled with a projected $RT_{NDT}$ (index to reference temperature, NDT) can accommodate $\Delta T$ or $\Delta RT_{NDT}$ values based on CVN specimens from surveillance programs. A similar initial action $K_{IC}$ curve has been established in the ASME Code, Section XI, Appendix A, for assessing indications discovered during
inservice inspection. This approach has been accommodated in regulatory limit curves. In the USA limits from adjusted surveillance data curves derived for US Regulatory Guides for irradiated $\text{RT}_{\text{NRT}}$ and for PTS (Pressurized Thermal Shock) limit curves. The weakest link in this approach is the underlying basic dependence on values derived from bounding curves from dynamic fracture toughness, $K_{\text{Ia}}$, and crack arrest toughness, $K_{\text{Ir}}$, for unirradiated steels or from large specimens irradiated in non-power reactors (i.e., at accelerated exposure rates). The types of curves for USA regulatory limits thus derived are shown in Figure 10. An elaborate correlation for chemical content, copper and nickel, is an underlying guide to these (and other) limit curves. Also shown are earlier correlations for comparison. Note that the current correlation predicts more damage at lower fluences and less at higher fluences.

It should be noted that after projecting the $\text{RT}_{\text{NRT}}$ for irradiation, a radiation-adjusted value from the $K_{\text{Ir}}$ curve is used $t_c$ for an identified or postulated defect to evaluate potential failure during emergency and faulted conditions. The $K_{\text{Ir}}$ and $K_{\text{Ic}}$ curves are used in lieu of measured fracture toughness data for a plant specific material.

Neutron energy spectrum differences clearly should and do have an effect on measured properties changes of a given steel at a given temperature. This effect is because the usual technique of measuring dosage involves a neutron exposure in terms of the number of neutrons having energies above a single level, usually 1 MeV.
This technique has worked well because of the similar environments of light water reactors in which the most damaging neutrons are of similar magnitudes and the energy spectrum shape is quite similar. However, it is important to note that the 1MeV neutrons are probably more damaging because of their numbers and because their collision cross-sections (probability for displacing iron atoms, and thereby the cause for damaging secondary collisions) are dominant in the usual LWR neutron spectrum. Thus, an explanation exists for the reproducibility of data >1MeV from reactor-to-reactor. Of course, neutron spectra do vary and lower energy neutrons do cause damage. Hence, there is a recent move to use displacements per atom (dpa) to measure damage. However, the difficulty in retroactive dpa analysis, or even in spectrum analysis, degrades the chances for gaining any significant benefit from "backfitting" using dpa techniques.

Studies over many years have sought possible flux (fluence rate) effects. Many results between test and power reactor exposures of the same (reference) steel suggest that variations in flux between one and one thousand (3 orders of magnitude) caused no significant effect. However, caution and logic have led to the use of only surveillance, near one-to-one exposure comparisons, for projecting trends. This approach eliminates the rate variable though it has not been demonstrated conclusively to be a significant variable for all steels in radiation-induced changes to key mechanical properties.

In order to minimize effects of neutron radiation to
mechanical properties of a given reactor steel, it is essential to isolate key factors; these factors are: the steel's composition (especially content of copper and to a lesser extent nickel and phosphorus), the neutron fluence (E >1MeV) or dpa, the dominant temperature of exposure and the index for these factors for commonly used classes of steel establishing mechanical performance curves. These factors affecting fracture potential when applied to the key operating factors (such as applied stress on known flaws and operating temperature gradients) establish probabilities for serious failures under abnormal operating conditions and the bases for critical conditions which must be controlled.

In conclusion, a number of factors impinge on vessel life assessment which revolve around irradiated mechanical properties of the vessel steel and its condition during operation. These factors together permit establishment of options for estimating and extending vessel life considering neutron radiation effects. Some of the technical issues essential to quantification of vessel degradation by neutron damage and emerging options for life assessment and possible life extension in irradiated vessels are outlined in Figure 4.

5 GENERAL INFLUENCE OF NEUTRON EXPOSURE

IN THE FORMER USSR VESSEL STEELS

5.1 Former USSR Vessel Design Considerations

(Design considerations for the WWER pressure vessels are presented in Chapter 4 of this book.)
5.2 Effect of Irradiation Temperature

Temperature has a marked influence upon radiation embrittlement, the level of embrittlement being reduced progressively with higher temperatures. The reason for this phenomenon is that with higher temperatures, the ability of displaced atoms to return to a vacancy site or other relatively innocuous location is enhanced, thereby relieving part of the damage.

It is reasonable to suppose that the process of self-annealing takes place at elevated temperatures resulting in a decrease in the shift of the brittle-to-ductile transition temperature.

Figures 5 and 6 [13] give the data on the effect of irradiation temperature on radiation embrittlement factor of steels, used for fabrication of reactor pressure vessels and weld metals in the former USSR.

The data for former USSR steels are given as dependence of radiation embrittlement factors against temperature. $A_r$ is the factor which determines the rate of shift of brittle-to-ductile transition temperature due to radiation according to the following formula:

$$T = A_r F_0^{1/3} ; F_0 = 10^{18} \text{n/cm}^2$$

(Note that $F$ is fluence for $E > 0.5 \text{ MeV.}$)
5.3 Fluence Rate (Flux) and Spectrum Effects in Former USSR

The scheduled time of power reactor operation exceeds, as a rule, the time of specimen irradiation in a research reactor with the fast neutron fluence corresponding to the end-of-life on the inner wall of the reactor vessel by up to three orders of magnitude. Thus, the issue of radiation stability of reactor vessel materials also involves the allowance for a possible effect of the neutron flux (fluence rate) on irradiation embrittlement.

In all countries where vessel-type LWRs are operated, the monitoring of the reactor vessel materials condition is currently accomplished by a surveillance program. However, as is typical in most types of reactor vessels, the flux in the surveillance capsules exceeds by several times the flux in the vessel wall. The question of a flux effect on the mechanical properties of a steel, such as variations in the transition temperature shift, is also possible when the surveillance capsule program utilizes high lead factors (e.g., greater than a factor of 10 times). Most U.S. surveillance programs do not exceed a lead factor of about 3 to 5, and therefore little flux effect is expected in USA LWRs.

It must be pointed out that the experiments on detection of a flux effect are very complicated and time consuming. For a correct establishment of a flux effect, constant specimen temperature should be maintained in both flux conditions during the whole irradiation period and the neutron energy spectrum must be nearly equal.

Though efforts in this direction have been made both in the
former USSR and in other countries, nobody has succeeded in
detecting a consistent flux effect. The results depend upon the
chemical composition of the steel, ranges of flux, and integrated
fluences within which the experiment is carried out. Information
in the literature does not provide data on the accuracy of
maintaining capsule temperature during irradiation. A small change
in the irradiation temperature within the range from 200°C to 300°C
has a very strong effect on steel as was noted earlier.

Figure 7 shows the calculational integral power spectra of
the fast neutron fluxes for the channels with surveillance capsules
in the WWER-440 reactors fully loaded (curve 1) and loaded with 36
fuel shielding assemblies (curve 2). It is clear that the shapes
of the neutron flux spectra in Figure 5 are similar, thus
indicating that the use of the shielding fuel assemblies does not
adversely affect the spectrum shape of neutrons hitting the
surveillance specimens.

The maximum values of the neutron flux were $4.2 \times 10^{12}$ and $4.75 \times 10^{11}$ n/cm$^2$/s, respectively. It follows from this that
installation of the shielding fuel assemblies results in reduction
of the neutron flux by about nine times for the surveillance
irradiation channels. Using the calculated shape of the spectrum
shown in Figure 15 and the cross section displacements for iron,
the numerical values for the rates of formation of displacements
per atom (dpa) have been calculated [14].

The maximum rates of formation of dpa exist in WWER reactor
surveillance channels in which those of a normal core are
3.7 x 10^{-9} and 4.1 x 10^{-10} for the shielded fuel elements. The
difference in fluence rate by using dpa is also approximately nine
times.

The values of $\Delta T$ depended importantly on the total content
of phosphorus and copper in the steel for the specimens irradiated
in the WWER fully charged (i.e., flux = 4 x 10^{12} n/cm^2/s). The
analysis of the data shows that the experimental points obtained
from the specimens irradiated in a WWER with a load of 313 fuel
assemblies (neutron flux equal to 4 x 10^{11} n/cm^2/s), in most cases,
lies above the upper envelope of weld data. This discrepancy is
particularly significant for the steels with elevated contents of
phosphorus and copper.

It should be pointed out that the values of $A_F (\Delta T)$ for a
low flux were obtained at neutron fluences between 1 x 10^{19} n/cm^2
and 5 x 10^{19} n/cm^2, while for high flux, the data were obtained at
fluences greater than 5 x 10^{19} n/cm^2. Therefore, a more correct
assessment of the flux effect can be achieved by the actual changes
in measured shift.

The program of surveillance investigations of the reactor
vessel materials at the Armenian NPP-2 (ANPP-2) prescribed use of
weld metal surveillance specimens cut from the same weld as in the
vessel. Thus specimens of the identical weld were irradiated under
identical conditions at ANPP-2 with a flux = 4 x 10^{12} n/cm^2/s and at
ROVNO (RNPP-1) with a flux = 4 x 10^{11} n/cm^2/s. The spectral
characteristics of neutrons affecting the specimens in both
reactors are essentially identical. The only difference was in the
irradiation periods and the neutron fluxes.

Figure 9 presents the experimental values of transition temperature shift ($\Delta T$) as a function of the fast neutron fluence for surveillance specimens at RNPP-1 (□) and ANPP-2 (O) and in the channel of the research reactor (MR) at the I.V. Kurchatov Institute of Atomic Energy. In the latter experiment the tests were from control specimens of the weld metal from the RNPP-1 irradiations. The irradiation of the specimens was carried out in contact with the coolant at a temperature of 270°C - 276°C. The neutron flux in the specimen locations was $7 \times 10^{12}$ n/cm$^2$/s. It follows from the analysis of Figure 9 that at a flux of $4 \times 10^{12}$ n/cm$^2$/s a saturation effect is not observed up to a fluence of $4.9 \times 10^{20}$ n/cm$^2$ ($>0.5$ MeV). This level is nearly twice as high as the lifetime fluence for the base metal of the WWER-440 reactor. At a flux of about $4 \times 10^{11}$ n/cm$^2$/s the embrittlement process occurs more intensely than at a flux of $4 \times 10^{12}$ n/cm$^2$/s within the fluence range of 1 to $5 \times 10^{15}$ n/cm$^2$.

The experimental values of $\Delta T$ are identical for fluences of $3.4 \times 10^{15}$ and $5 \times 10^{15}$ n/cm$^2$. But whether it is a tendency toward saturation of irradiation effects at a flux of about $4 \times 10^{11}$ n/cm$^2$/s will be shown by testing the available assortment of specimens from RNPP-1 with a higher fluence level. This question is especially important in view of the fact that on the inner surface of the WWER-440 pressure vessel, the flux is equal to $3 \times 10^{11}$ n/cm$^2$/s, that is, it is very close to the flux values obtained with the same types of specimens in RNPP-1.
6 CONCLUSIONS

This chapter covers the effects of neutron irradiation on mechanical properties of reactor pressure vessel steels as viewed from the USA and former USSR perspectives for specific primary systems used in these countries. It is intended that the summary provided serves as a proxy for all Light Water Reactor Systems (LWRs) and provide background on the critical elements controlling such neutron damage effects.

The major properties affected by high energy neutron exposure are hardness (a physical property) and the related mechanical properties strength and toughness. These and variants caused by the environmental and materials factors can have serious effects upon vessel integrity and hence the life of a LWR plant. The factors of neutron environment (flux, fluence, and energy spectrum) and temperature also have major effects. However both types of nuclear systems (USA and the former USSR) operate in similar ranges of neutron and temperature conditions so that no major discrepancies in conclusions are reached between the two. Further the evolution of vessel steels, construction methods in both countries, and aspects related to metallurgy and microstructure for plate, weld, and forging materials are crucial to the level of embrittlement attained.

A most critical factor in the quantitative assessment of embrittlement is steel composition, especially the levels of copper, nickel, and phosphorus. The former USSR investigators have considered other elements as well: vanadium, silicon, aluminum,
manganese, nitrogen, boron, and oxygen. Copper, nickel, phosphorus plus vanadium are agreed to be most prominent in causing embrittlement, though the most detrimental threshold levels of content are yet to be defined. Further, the role of microstructure is a qualitative factor in the change of mechanical properties with neutron exposure, but more research is required to provide specific and quantitative guidance to these effects.

The mechanisms of radiation hardening and embrittlement are defined physically as submicrostructural features: precipitates, vacancy clusters, and interstitial clusters. The shape, size, and distribution of these features are sensitive to thermal and nuclear dynamics of service environments. Suggestions of fluence rate effects have been made, but surveillance conditions in the USA show limited rate effects. In the former USSR, while the rate differences may reach an order of magnitude, investigators believe temperature variations can be so overwhelming as to cover any flux effect. More research on this subject is required before quantitative assessment of a rate effect can be assigned.

The extent of our knowledge of the most critical factors to steel embrittlement response, however, is such that damage or embrittlement can be assessed and assigned in most vessels based upon knowledge of the steel (composition and metallurgical microstructure), neutron environment (primarily fluence) and temperature. From this knowledge, trend curves relating these factors to fracture toughness provides a limiting condition from which operators may judge integrity provided flaw sizes and
stresses at critical vessel locations can be assigned with confidence.

Acknowledgments

The authors wish to thank Mr. Lendell Steele for his valuable and critical review and editing of this manuscript.
Chapter 8 References


3. CMEA Standards of Nuclear Power Plants; Vol. 4, Components and Piping for Nuclear Power Plants; Part 2, Standards for Strength Calculations; Checking Calculations; Calculation of Resistance against Brittle Failure, Interatomenergo, Moscow, 1984.


Chapter 8 Tables

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Table 2 Pressure Vessel Plate Materials
Table 3 Pressure Vessel Forging Materials
Table 4 Welding Techniques for Nuclear Components
Table 5 Technical Characteristics of Reactor Vessels
Table 6 Specifications for heat composition of base metals used in the beltline materials of former USSR reactor vessels
Table 7 Specifications for composition of submerged arc weld metal in the beltline for former USSR reactor vessels
## Table 1
Sizes and Weights of Typical Reactor Vessels

<table>
<thead>
<tr>
<th>Type (Vendor)</th>
<th>Size MW(e)</th>
<th>Overall Height (m)</th>
<th>Inside Diameter (m)</th>
<th>Wall Thickness (mm)</th>
<th>Dry Weight (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BWR (G.E.)</td>
<td>1065</td>
<td>22.2</td>
<td>6.4</td>
<td>150</td>
<td>680,000</td>
</tr>
<tr>
<td>PWR (B&amp;W)</td>
<td>1213</td>
<td>13.8</td>
<td>4.9</td>
<td>248</td>
<td>540,000</td>
</tr>
<tr>
<td>PWR (W)</td>
<td>1106</td>
<td>13.3</td>
<td>4.4</td>
<td>222</td>
<td>390,000</td>
</tr>
<tr>
<td>PWR (C.E.)</td>
<td>1270</td>
<td>15.3</td>
<td>4.6</td>
<td>254</td>
<td>510,000</td>
</tr>
</tbody>
</table>

G.E. - General Electric  
B&W - Babcock and Wilcox  
W - Westinghouse  
C.E. - Combustion Engineering

## Table 2
Pressure Vessel Plate Materials

<table>
<thead>
<tr>
<th>Grade</th>
<th>Heat Treatment</th>
<th>Vessels</th>
<th>Usage</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>A212B</td>
<td>N&amp;T, Q&amp;T</td>
<td>All</td>
<td>Mid 50s-60s</td>
<td>C-Si</td>
</tr>
<tr>
<td>SA302B</td>
<td>Q&amp;T</td>
<td>All</td>
<td>Mid 50s-60s</td>
<td>Mn-Mo</td>
</tr>
<tr>
<td>SA302B (modified)</td>
<td>Q&amp;T</td>
<td>All</td>
<td>Mid-Late 60s</td>
<td>Mn-Mo-Ni</td>
</tr>
<tr>
<td>SA533B-1</td>
<td>Q&amp;T</td>
<td>RPV, pressurizer</td>
<td>70s to present</td>
<td>Mn-Mo-Ni</td>
</tr>
<tr>
<td>SA533B-1 (low Cu, P)</td>
<td>Q&amp;T</td>
<td>RPV beltline</td>
<td>1973 to present</td>
<td>Mn-Mo-Ni</td>
</tr>
<tr>
<td>SA533A</td>
<td>Q&amp;T</td>
<td>Steam Generator</td>
<td>70s to present</td>
<td>Mn-Mo</td>
</tr>
</tbody>
</table>

N - Normalized  
Q - Quenched  
T - Tempered
### Table 3
Pressure Vessel Forging Materials

<table>
<thead>
<tr>
<th>Grade</th>
<th>Heat Treatment</th>
<th>Applications</th>
<th>Usage</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>A105 II</td>
<td>N&amp;T</td>
<td>Flanges, nozzles</td>
<td>50s (limited)</td>
<td>C-Mn</td>
</tr>
<tr>
<td>SA182F1 (modified)</td>
<td>Q&amp;T</td>
<td>Flanges, nozzles</td>
<td>50s-60s</td>
<td>Mn-Mo-Ni</td>
</tr>
<tr>
<td>SA336</td>
<td>N&amp;T, Q&amp;T</td>
<td>Flanges, nozzles</td>
<td>50s-60s</td>
<td>C-Mn-Ni</td>
</tr>
<tr>
<td>Code Case 1236</td>
<td>Q&amp;T</td>
<td>Flanges, nozzles</td>
<td>1957-60s</td>
<td>Low Ni-Cr-Mo</td>
</tr>
<tr>
<td>SA508-2</td>
<td>Q&amp;T</td>
<td>Flanges, nozzles, rings</td>
<td>Current</td>
<td>Same as code case 1236</td>
</tr>
<tr>
<td>SA508-2a</td>
<td>Q&amp;T</td>
<td>Tube Sheets, flanges</td>
<td>Current</td>
<td></td>
</tr>
<tr>
<td>SA508-3</td>
<td>Q&amp;T</td>
<td>Flanges, nozzles, rings</td>
<td>Current</td>
<td>Same as SA182F1 modified</td>
</tr>
</tbody>
</table>

### Table 4
Welding Techniques for Nuclear Components

<table>
<thead>
<tr>
<th>Technique</th>
<th>Type</th>
<th>Heat Treatment</th>
<th>Application</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sub Arc</td>
<td>Auto</td>
<td>Stress relief</td>
<td>Wherever possible</td>
<td>Good properties high deposition</td>
</tr>
<tr>
<td>Sub Arc - narrow gap</td>
<td>Auto</td>
<td>Stress relief</td>
<td>Girth seams</td>
<td>Reduced weld volume</td>
</tr>
<tr>
<td>Shielded metal arc</td>
<td>Manual</td>
<td>Stress relief</td>
<td>Complex or irregular</td>
<td>Very flexible</td>
</tr>
<tr>
<td>Electroslag</td>
<td>Auto</td>
<td>Q&amp;T</td>
<td>Longitudinal seams in some BWRs</td>
<td>Very high deposition rate</td>
</tr>
<tr>
<td>Parameter</td>
<td>WWER-440</td>
<td>WWER-1000</td>
<td></td>
<td></td>
</tr>
<tr>
<td>--------------------------------------------------------------------------</td>
<td>---------------</td>
<td>---------------</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1. Coolant temperature at the reactor inlet under nominal conditions (°C)</td>
<td>267 ± 2</td>
<td>289 ± 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2. Coolant pressure in the reactor vessel under nominal conditions (MPa)</td>
<td>12.3</td>
<td>15.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3. Fast neutron fluence for the reactor vessel base metal during design service life of 40 years, n/cm² (E ≥ 0.5 Mev)</td>
<td>$2.6 \times 10^{20}$</td>
<td>$5.7 \times 10^{19}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4. Fast neutron fluence for the weld metal during a service life of 30 years</td>
<td>$1.86 \times 10^{20}$</td>
<td>$5.7 \times 10^{19}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$2.48 \times 10^{20}$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Steel</td>
<td>C</td>
<td>Si</td>
<td>Mn</td>
<td>Cr</td>
</tr>
<tr>
<td>-----------------------</td>
<td>------</td>
<td>------</td>
<td>------</td>
<td>------</td>
</tr>
<tr>
<td>15X2M0A-A</td>
<td>0.13</td>
<td>0.17</td>
<td>0.30</td>
<td>2.5</td>
</tr>
<tr>
<td>(WWER-440) (old design)</td>
<td>0.18</td>
<td>0.37</td>
<td>0.60</td>
<td>0.8</td>
</tr>
<tr>
<td>15X2M0A-A</td>
<td>0.13</td>
<td>0.17</td>
<td>0.30</td>
<td>2.5</td>
</tr>
<tr>
<td>(WWER-440)</td>
<td>0.18</td>
<td>0.37</td>
<td>0.60</td>
<td>0.3</td>
</tr>
<tr>
<td>15X2HM0A-A</td>
<td>0.13</td>
<td>0.17</td>
<td>0.30</td>
<td>1.8</td>
</tr>
<tr>
<td>(WWER-1000)</td>
<td>0.18</td>
<td>0.37</td>
<td>0.60</td>
<td>2.3</td>
</tr>
</tbody>
</table>

1 Steel 15X2M0A-A is currently being used (from about 1980) for the manufacture of reactor shells positioned opposite the core. 15X2M0A-A and 15X2HM0A-A steel are smelted using pure original iron ore mixture (i.e., not scrap metal).
Specifications for composition of submerged arc weld metal in the beltline for former USSR reactor vessels

<table>
<thead>
<tr>
<th>Base metal joining to base metal</th>
<th>Content of elements, %</th>
<th>Cu</th>
<th>S</th>
<th>P</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>C</td>
<td>Si</td>
<td>Mn</td>
<td>Cr</td>
</tr>
<tr>
<td>15X2M0A with 15X2M0A (WWER-440 design)</td>
<td>0.04-0.12</td>
<td>0.20-0.60</td>
<td>0.0-1.3</td>
<td>1.2-1.8</td>
</tr>
<tr>
<td>Any joint with 15X2HM0A-A (WWER-440)</td>
<td>0.04-0.12</td>
<td>0.20-0.60</td>
<td>0.0-1.3</td>
<td>1.2-1.8</td>
</tr>
<tr>
<td>15X2HM0A-A with 15X2HM0A-A (WWER-1000)</td>
<td>0.05-0.12</td>
<td>0.15-0.45</td>
<td>0.50-1.00</td>
<td>1.40-2.10</td>
</tr>
<tr>
<td>15X2HM0A-A with 15X2HM0A-A (WWER-1000)</td>
<td>0.04</td>
<td>0.15</td>
<td>0.45</td>
<td>1.20</td>
</tr>
</tbody>
</table>

<sup>x</sup>By new requirements upper content of nickel is not more than 1.5%.
Fig. 1 Schematic diagram illustrating how irradiation strengthening produces a shift in transition temperature [11].
Fig. 2 Effect of irradiation temperature on transition temperature increase for an A302-B reference steel [14].
Fig. 3 A comparison of shift predictions for welds and base metal (0.35 wt% Cu and 0.60 wt% Ni) calculated using Revision 1 and 2 of Regulatory Guide 1.99 and other PTS correlations.
Fig. 4 Parameters and options available for estimating and extending licensed life when considering radiation embrittlement.
Fig. 5. Effect of irradiation temperature on radiation embrittlement factor of steels 15X2MOA-A (a) and 15X2HMOA-A (b).
Fig. 6 Effect of irradiation temperature on radiation embrittlement factor of weld metal of steels 15X2MOA (a), and 15X2HMOAA (Cu ≤ 0.08%; P ≤ 0.012%) (b)
Fig. 7 - Integrated energy spectrum of fast neutrons $E > 0.5$ MeV at irradiation locations of WWER-440 surveillance specimens.

1. Normal loading of core
2. Loading of core with shielding assemblies
Fig. 8 - Integrated energy spectrum of fast neutrons at irradiation locations of VVER-440 surveillance specimens.
$P = 0.028\%$
$Cu = 0.18\%$

- $\varphi = 4.10^{11}$ cm$^{-2}$s$^{-1}$, RNPP-1
- $\varphi = 4.10^{12}$ cm$^{-2}$s$^{-1}$, ANPP-2
- $\varphi = 7.10^{12}$cm$^{-2}$s$^{-1}$, MTR

- calculated curve in accordance with norm

$$\Delta T = 800 \ (\%P + 0.07\%Cu) \ (F/F_0)^\mu$$
$$= 800 \times 0.04 \ (F/F_0)^\mu$$
$$= 32 \ (F/F_0)^\mu$$
$E > 0.5$ MeV

Fig. 9 - Radiation embrittlement of weld metal at irradiation temperature of 270$^\circ$ C showing fluence rate effects
CHAPTER 9
Microstructural Evolution in Neutron Irradiated Reactor Pressure Vessel Steels
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1. Introduction

The neutron irradiation experienced by the reactor pressure vessel (RPV) of a thermal reactor can produce significant changes in the mechanical properties of ferritic steels used in the construction of such vessels [1,2]. Typically, an increase in the tensile yield strength is observed, accompanied by an increase in hardness and a decrease in work hardening rate and ductility. The ferritic steels used in the RPV exhibit a ductile to brittle transition temperature (DBTT) in which the energy of fracture increases with increasing temperature on passing through the transition, followed by a region of relatively constant high fracture energy (upper shelf). The effect of irradiation manifests itself in an increase in the DBTT with a decrease in the upper shelf energy. Mechanical property changes of this nature are clearly of considerable importance in the safe operation of reactor systems and play a major role in plant life extension studies. It is the purpose of this chapter to consider the microstructural changes that give rise to these changes in macroscopic properties.

It has been recognised for many years that the dominant mechanism is the formation of small clusters during neutron irradiation which impede dislocation motion and give rise to an increase in tensile strength. These clusters are formed by the action of the mobile point defects created under irradiation, either through irradiation-induced diffusion and subsequent precipitation of solute atoms or through the clustering of the irradiation produced point defects. In this chapter we discuss the fundamental radiation damage mechanisms underlying the formation of these small clusters, the techniques available to study them, and illustrate the present status of such studies.
Within this context it is important to note the attention that this phenomena is achieving world-wide. On the micro-mechanisms the International Group on Radiation Damage Mechanisms in Reactor Pressure Vessel Steels (IGRDM) was founded in 1987 with USNRC sponsorship to bring together in a workshop environment scientists and engineers involved directly with RPV embrittlement issues [3]. Involvement within the group has resulted in many world-wide inter-laboratory collaborations. These have made an impressive addition to our understanding of the embrittlement phenomena; many of the references cited reflect such collaboration and the advantages of applying various techniques to the same material.

2. Damage Mechanisms

An operating RPV is exposed to a flux of neutrons throughout its life. The exact neutron energy spectrum and flux depends on the location in the vessel and the precise reactor design. Examples of a typical spectra are presented in Figure 1 for the belt-line region. The energy spectrum ranges from thermal neutrons to neutrons with energies above 1MeV extending up to 10MeV. As the neutrons penetrate the vessel wall the low energy neutrons are absorbed and the shape of the spectra change. This is illustrated in Figure 2 by comparing the ratio of neutrons > 0.1MeV and 1MeV, where the decreasing importance of neutrons above 1MeV at 1/4T and 3/4T can be seen. In the belt-line region the vessel is exposed to typically \(6 \times 10^{19} \text{n/cm}^2 > 1\text{MeV}\) during its lifetime.

In a typical PWR spectrum the great majority of the damage is created by neutrons above 0.1MeV, but thermal neutrons will also damage the RPV. Several of the processes occurring during the initial damage event can strongly influence microstructural evolution and these are considered in section 2.1 below. The subsequent evolution and an overview of current understanding of the mechanisms of embrittlement is presented in section 2.2.
2.1 Importance of initial damage event.

Collisions between the fast neutron and the lattice atom can transfer energies ranging from a few eV to tens of keV. Indeed, a high fraction of the primary knock-on atoms (PKA) have energies above a few tens of eV. These PKA lose energy through interacting with both the electrons and atoms of the solid. If the energy transferred to a lattice atom is greater than some threshold value, \( E_d \) (typically >40eV) then the atom will be displaced from its lattice site, creating a Frenkel defect, i.e. a vacancy (empty lattice site) and an interstitial (an atom occupying a non-lattice site). Typical recoil spectra are shown in Figure 3 for a PWR surface and 1/4T positions for energies above this threshold value; it can be seen that although the majority of recoils are below 5keV, a significant fraction have energies above 30keV.

If the PKA energy is much greater than a few keV the PKA is able to displace many atoms, rapidly entering a regime where the collisions occur every lattice spacing. This sequence of events is called a collision or displacement cascade. It is important to realise that in the evolution of a cascade not only is a heavily damaged region containing a large number of displaced atoms established, but that during the subsequent evolution considerable point defect motion, recombination and clustering may occur. Indeed, there is general agreement that the centre of the cascade is very hot, with "temperatures" typical of the liquid state, commonly termed a thermal spike (see for example [4]). The final configuration is generally a vacancy rich region surrounded by interstitial atoms (which may be clustered), as envisaged in the classical pictures of cascades, [5,6]. The main stages in cascade evolution are detailed in Table I and the current status of cascade studies can be seen from papers such as Diaz de la Rubia and Guinan [7], English et al [8].

Certain cascade processes, particularly during the thermal spike phase can have a profound impact on subsequent microstructural development and it is important to consider these in
detail and to comment on the specific case of an iron matrix. Most important to the development of the microstructure in RPV steels are the number of point defects produced by each cascade, this is the basis of one of the foremost exposure parameters, dpa, and the clustering of point defects within the cascade.

The accepted method of evaluating the amount of damage is given by the formalism developed by Norgett, Torrens and Robinson [9]. It is also incorporated into an ASTM Standard E693 [10]. The critical aspects are that it describes the number of displaced atoms, \( N_d \), by evaluating the partitioning of energy between nuclear (displacement producing) and electronic losses and by a statistical process evaluates the number of displacements resulting from the energy transferred in collisions. In Norgett, Torrens and Robertson (NRT) \( N_d \) is given by:

\[
N_d = k \frac{E_{\text{dam}}}{2E_d}
\]  

where \( k \) is the damage efficiency factor and is energy independent with a value of 0.8, \( E_{\text{dam}} \) is the energy available for damage after energy loss to the electrons is taken into account, and \( E_d \) is the mean displacement energy.

This is the basis for the Internationally agreed exposure parameter displacements per atom, dpa [9]. Evidence from varied experimental studies of RPV embrittlement suggests that the dpa parameter provides an adequate exposure parameter. In spite of some limitations the physical basis behind it is superior to other damage functions. However, some debate still continues stimulated by the recommendation in the USNRC Regulatory Guide 1.99 rev 2 that dpa should be used for describing the attenuation of damage through the thickness of an RPV.

Recently, evidence from fundamental experimental studies and from computer simulations of cascade evolution point to a limitation in the formalism for dpa; more specifically, that
the damage efficiency $k$ is in fact energy dependent \([11, 12]\). Here, we illustrate this from molecular dynamics simulations. Figure 4 shows the NRT efficiency factor $k$, versus the recoil energy $E$ for copper \([13]\). These results are based on over a hundred cascade simulations at 100 K in copper with random knock-on directions and energies ranging from 60 eV (2 $E_d$) to 10 keV. It is clear that $k$ decreases from a value of 0.8 to 0.37 between 60 eV and 250 eV, from 250 eV to 2 keV there is a steady linear decrease in total efficiency to 0.28, and $k$ continues to decrease more slowly on going from 2 keV to 10 keV. We also include on figure 4 values of $k$ obtained from the alpha iron simulations of Calder and Bacon \([14]\) with $E_d=30$ eV. Although based on a more limited data set (of 9 cascades), the efficiency factors are similar to those of copper.

This energy dependence of $k$ has achieved international prominence in the RPV embrittlement area with the HFIR embrittlement problem. Here, very much higher than anticipated Charpy shifts in the HFIR Pressure Vessel surveillance programme when compared with similar material irradiated in Material Test Reactors \([15]\). This higher embrittlement effect was variously postulated to be a fluence effect or a neutron spectrum effect. Early assessments of the HFIR surveillance locations suggested a very high thermal to fast ratio of 100:1, and enhanced damage was suggested because the low energy (n,\gamma) recoils are significantly more damaging, relative to high energy recoils, than the present NRT code allows. It was the potentially large magnitude of this effect which provided a driving force for a re-evaluation of the specification of radiation exposure parameters. However, more recent evaluations of the HFIR neutron spectrum suggest more modest thermal to fast rates which places greater emphasis on a fluence effect.

The second fundamental process affecting microstructural evolution is the clustering of both interstitial and vacancy point defects within individual cascades. The majority of evidence is for relatively simple metal systems, either through experimental studies or through computer modelling of cascades (see for example \([8]\)). Evidence of interstitial clustering in isolated cascades comes primarily from molecular dynamics simulations of
cascades in copper and other pure metals. Here, it is observed that the self-interstitial atoms in the periphery of the disordered central damage region frequently coalesce to form clusters. (SIA) As the knock-on energy is increased the number of SIA produced in this peripheral region rises. The strong elastic interaction between the stress fields of the SIA enhances the probability that an individual SIA will combine with another of its kind rather than recombine with a vacancy. This clearly demonstrates the steady increase in the production of SIA clusters as the energy is increased, so that at 2 keV and above an appreciable fraction of the SIA are being produced in cluster form. As yet there is no direct evidence that these will be formed in iron or RPV steels at the temperatures of interest, or of the role of these in subsequent microstructural evolution. However, they cannot be ruled out.

In many pure fcc and bcc metals there is extensive evidence for vacancy loop formation within individual cascades [16]. This can occur with a quite high efficiency, particularly in fcc metals such as copper. The very high generation rate of cascades and thus vacancy loops in neutron irradiation means that such loops can at low temperatures have a dominant effect on the microstructure. However, in contrast in alpha-iron there is very little evidence for vacancy cluster formation within individual cascades at room temperature and above. Indeed, neutron irradiation of high purity iron to doses where considerable cascade overlap occurred produced no discernible microstructure within the grains, suggesting that under fission neutron irradiation no small clusters are formed (either interstitial or vacancy. This strongly suggests that cluster formation under irradiation is dominated by diffusion and interaction of the point defects outside the cascade, particularly with impurity or solute atoms. This is the subject of the next section.

2.2 Microstructural Evolution in RPV steels.
As described above microstructural evolution in RPV steels is dominated by the interaction of the radiation induced point defects after they have left the cascade region. Also of significance is their interaction with the impurities in the steel. Both vacancy and interstitial point defects are expected to be mobile in the temperature range of most operating pressure vessels. However, they are also expected to interact with solute atoms. The key interstitial impurities in iron and steel are C and N and there is a wide range of experimental evidence confirming the strong interaction between C and N solutes and irradiation induced defects [1]. The partial or complete trapping of self-interstitials by C and/or N solutes will cause heterogeneous cluster nucleation and a fine cluster distribution. Further, in steels containing residual levels of elements such as copper, which are in super-saturated solution, radiation-enhanced diffusion will occur at these temperatures, which leads to the formation of small clusters which can again harden the matrix. It is now generally accepted that the mechanical property changes produced during neutron irradiation can be rationalised in terms of this underlying microstructure. Generally, after neutron irradiation the processes described above lead to a microstructure consisting of small clusters (<5nm in diameter) which create obstacles to the free movement of dislocations thereby producing an increase in the yield stress, hardness and the ductile-brittle transition temperature of the material.

Physically based models for the prediction of changes in yield stress and ductile-brittle transition temperature [17-20] have been developed and they all have the common feature of considering at least two irradiation-induced microstructural components. The first component is a matrix radiation damage contribution produced by the coalescence of point defects created by neutron - lattice atom interactions, possibly interacting with C and N as described above. The resulting obstacles may comprise dislocation loops or microvoids. The second component is a copper-related contribution produced by the radiation-enhanced diffusion and precipitation of impurity copper from solid solution in the steel. The enhancement arises because the steady state vacancy concentration present during irradiation is significantly higher than that arising from thermal exposure alone. It is not
the purpose of this chapter to consider these mechanism and models in any great detail as this is covered elsewhere in this volume. However, in order to place the need for microstructural characterisation in context we give below a summary of the essential features of each component of the damage, and highlight a selection of critical issues and how they can be addressed in Table II (see also [21]).

The greatest insight has been obtained into the copper component of the microstructure, as under both thermal ageing and irradiation it has been possible to examine its precipitation from solid solution. Initially, this occurs as coherent bcc precipitates producing efficient pinning sites inhibiting dislocation motion. Under irradiation the precipitates have not been observed to over-age; instead they remain at about 2 nm diameter under all conditions reported to date. Evidence from various studies strongly suggests that the hardening from the copper-rich clusters reaches a plateau once the copper has precipitated from solution. The rate at which the plateau value is reached is controlled by factors such as dose rate, and the level of the plateau is determined by factors such as composition and pre-irradiation heat treatment, which can influence the initial distribution of the copper is clearly of importance to any subsequent precipitation. Indeed, there is a continuing need to understand and document the effect of environmental and metallurgical variables, such as dose rate, heat treatment and composition, on the precipitation.

There is as yet no direct evidence for the nature of the matrix defects, they may be "sponges" (vacancy rich regions), microvoids, dislocation loops (vacancy or interstitial), or solute-point defect clusters. The nature, size and number density of such defects remains largely unknown. As in the case of precipitation there is a need to understand the factors controlling the production and stability of these defects. The irradiation environment and materials variables (both microstructure and impurity levels e.g. C, N, & O) are known to be important but the details of how these factors influence the processes are not fully understood. All the data suggests that for the dose range experienced by RPV's this
component is continuously formed under irradiation. The different dose dependencies of the two components is illustrated in Figure 5 [22].

There might also be potential for non hardening embrittlement in some RPV materials. Non hardening embrittlement is generally associated with intergranular failure resulting from grain boundary segregation. Elements such as P, S, As, Sn and Sb are known to segregate to the grain boundaries under thermal conditions. There is a clear need for the identification of the mechanism(s) and factors influencing the segregation of these elements during irradiation.

A complete characterisation of the microstructure requires a knowledge of the number density, composition, structure, and size distribution of the irradiation-produced clusters and precipitates, an estimate of the grain size and dislocation density, and the spatial distribution of known embrittling elements such as copper, phosphorus and nickel. In practice this can only be achieved by focusing a range of microstructural techniques on model alloys and commercial steels under irradiation. It is important to emphasise the practical benefits of such characterisation. It is clear from the above that the detailed microstructure and thus mechanical property change is sensitive to a wide range of material and irradiation variables. Frequently, in developing trend curves for operating RPV's, it is necessary to extrapolate outside the database and to do this with confidence requires underpinning by a knowledge of the mechanisms controlling the microstructural evolution. Such an approach can help reduce the conservatism by providing a firmer basis for choosing the form of the trend curve and correlating data more effectively by taking into account variations in environmental and metallurgical variables. In particular, the usefulness of the modelling approach in establishing the dose dependence of hardening and embrittlement in copper-bearing steels irradiated in different fluxes has increased the demand for microstructural characterisation. This serves to underpin model development by confirming the basic mechanisms and critical assumptions, as well as providing data to remove uncertainties in the model parameters.
3. TECHNIQUES FOR EXAMINING THE MICROSTRUCTURE

It is only in recent years that microstructural examination has started to play a major role in understanding the degradation of RPV steels. This is contrast to phenomena such as void swelling in fast reactor cladding where electron microscopy made a major contribution in discovering and documenting the phenomena. This is primarily, because in commercial RPV steels many of the techniques required considerable development before they could successfully be applied to this problem. However, there have been rapid advances made in the last five to ten years. It has been found that no one technique can fully characterise samples, and frequently a combination of techniques have been employed, successful inter-correlation of data from different techniques is a feature of work in this area. In this section we focus on the techniques available and type of analysis they can be used for (section 3.1), and illustrate the success of such studies by drawing on recent examples from the UK. (section 3.2).

3.1 Techniques Available

Table III has been compiled to show the techniques available to characterise the microstructure/chemistry of the material. The table indicates the strengths and limitations of the techniques together with suitable references where available. The techniques can be broadly split into two groups, namely the 'mature' techniques that have been used extensively in the RPV area, and new, in many cases novel, techniques that tend to be used to answer specific questions when applied to model materials. Many of the techniques can be used to provide information on all three of the components of embrittlement as mentioned above, however some are specific as indicated by the following letters in the table, i.e. precipitates (P), matrix (M) and grain boundaries (B). Techniques such as SANS and Positron Annihilation sample relatively large volumes of material and yield
information on bulk averaged properties of the cluster distributions. In contrast in techniques such as electron and field ion microscopy individual features of the microstructure are analysed, often in sufficient numbers to give statistically meaningful data. Frequently, it is by combining such contrasting techniques that the greatest insight can be obtained. An example of this is that microscopy observations of the composition of precipitates can be used to refine the interpretation of SANS data, as in SANS it is impossible to unambiguously fix the composition of the small clusters.

Many of these methods have been validated by studying the distributions of copper precipitates in irradiated or thermally-aged simple model alloy systems. A comparison between techniques is illustrated in Figure 6 where the size distributions of small copper precipitates formed in thermally aged Fe-Cu-Ni, measured by SANS and TEM, are compared [23]. The agreement is excellent, both techniques showing a bi-modal distribution after 10hrs ageing. Another important microstructural variable is the amount of copper in the matrix. Table IV shows the Cu levels measured in-between second phase particles in aged Fe-Cu alloys; again there is good agreement between the techniques used [24].

### 3.2 Examples of recent applications.

It is beyond the scope of this chapter to critically review the progress made in the numerous studies cited in Table III. The purpose here is to illustrate the mechanistic insight possible from studies of model materials and the progress made in the study of commercial materials.

#### 3.2.1 Micro-mechanisms of particle hardening.

A critical aspect of developing micro-mechanistic models of the embrittlement by copper precipitates is to verify the currently accepted Russell and Brown modulus hardening
theory [25] for the particle hardening. Recent work in the UK has focused on a multi-
technique study of the observed hardening on ageing or irradiation with the hardening predicted from the measured populations of copper precipitates. A problem in making such a comparison is to separate the hardening due to other components such as non-
copper precipitates or dislocations, grain boundaries etc. This is complicated by the small decrease in the dislocation density component and the more significant effect due to the reduction in matrix copper content caused by ageing, which have to be determined from experimental measurements. Figure 7 shows the variation in hardness of the Fe-Cu alloy after ageing at 550°C [24]. The hardening contributions from the measured grain structure and dislocation density are shown. To these are added the contributions from copper in solid solution, based on the composition predictions of SANS and measured values obtained by STEM and AP/FIM given in [23] combined with the hardening effectiveness of copper [26]. To this an allowance is made for a third of the vacancies present during the solution heat treatment to be retained after quenching in the form of microvoids. The microvoids are assumed to evaporate during the first few minutes of ageing, in keeping with an interpretation of the SANS data.

A comparison is made in Table V between the copper precipitation-hardening component, deduced from figure 7, and that calculated using the Russell and Brown [9] modulus hardening theory. The calculated values were based on the precipitation size and number densities measured by SANS [23]. These data were fixed by STEM and FIM analysis of the matrix and precipitate composition [23, 24]. The appropriate form of the Russell-
Brown model depends on the angle made by the line dislocation as it leaves the precipitate. For the Fe-Cu system this was obtained by in-situ TEM straining experiments on a range of model alloys. The precipitate hardening is dependent on the difference in shear moduli between the precipitate and matrix. The appropriate Russell-Brown equations were solved using the above information for both fcc and bcc shear moduli for copper. The bcc modulus was obtained from the MD study [24] using a modified embedded atom interatomic potential of Ackland et al. [27] MD simulations have in turn
been cross checked by comparing the bcc lattice parameters and the transformation structure with experimental TEM, EXAFS [24] and HREM [28] data. An encouraging agreement is obtained using this approach, with the calculations using the bcc shear modulus appearing the more appropriate.

3.2.2 Application to commercial materials

Many of the techniques listed in Table III have been applied successfully to commercial materials. An initial significant observation by Fisher and co-workers [29,30] was the presence of copper sulphide, Cu$_{1.8}$S, in submerged arc and manual welds used in Magnox pressure vessels; Buswell [31] subsequently found this phase in the UK submerged arc weld in Phase II of the IAEA programme. These results show that not all the copper is initially in solid solution and available to embrittle the vessel, a proportion may be removed by sulphide formation.

English [32] studied the micro-distribution of copper and nickel in unirradiated and irradiated weldments using TEM techniques on both the ferrite and second phase particles. In the ferrite copper levels exhibited considerable spatial variation and for high copper welds the matrix copper content was significantly lower than the bulk chemical analysis. This was due to copper precipitating on dislocations during the post-weld heat treatment. Copper levels in the matrix recovered when the material was given a heat treatment of 5h at 750°C (Table VI). This result shows that in high copper material it is clearly necessary to take account of the post-weld heat treatment, as well as the effects of the potential for copper sulphide formation, in assessing embrittlement.

In a joint USA/FRG/UK study, archive samples and material trepanned from the vessel wall of the decommissioned 250 MW BWR Gundremmingen Reactor (KRB-A) have been examined by a combination of SANS and TEM/STEM [33]. Small precipitates (2 to 3 nm diameter) were found in both the service irradiated trepan sections and in archive material after accelerated irradiation. The SANS data is consistent with the anticipated effects of dose rate variation [34] in producing larger precipitates in lower dose rate exposure and
apparently enhancing the development of copper precipitation at a given dose. The presence of copper-rich precipitates alloyed with both manganese and nickel has been confirmed in these irradiated steels by TEM/STEM techniques. The matrix copper contents in the irradiated trepan were determined by STEM to be $0.14 \pm 0.04$ wt% (cf. nominal bulk value 0.16 %) in good agreement with SANS data, indicating that only a small fraction of the bulk copper has precipitated during irradiation. Unexpectedly, in this class of material, a number of small vanadium-rich precipitates were detected. These were enriched with copper, but using windowless EDX, the levels of C and N were qualitatively assessed and appeared to be lower than expected from a VC or VN precipitate. In an FIM investigation of service and accelerated irradiation material, diffuse regions rich in copper, phosphorous and nickel were observed [35]. Molybdenum rich carbo-nitride platelets and precipitates rich in vanadium were also observed. No copper could be detected with these precipitates due to the problems of overlap between copper and vanadium nitride ion masses. The deviation from ideal stoichiometry observed in the STEM is in agreement with FIM results [35]. It remains to be established whether these precipitates were irradiation-induced.

4. Discussion and Conclusions

It is now well-established that the dominant mechanisms of RPV embrittlement are the formation of clusters by point defects escaping from collision cascades. The subsequent microstructural development is largely determined by two processes, copper precipitation and matrix damage. The resultant microstructure is dominated by a high dispersion of small clusters, frequently < 2nm in diameter. It is only in the last decade that techniques have been sufficiently refined to successfully analyse these features. The majority of recent data has focused on the role of copper in these materials and has clearly been successful in the identification of the factors affecting the precipitation of the copper to produce effective dislocation pinning sites with resultant increase in hardness and shift in DBTT. The critical issues are centred around the factors controlling the nucleation and growth, and precise
hardening mechanism of these copper rich precipitates. Here, dose, dose rate, temperature and steel composition, product form (weld vs plate) and thermal/mechanical history have also been identified as important factors. The second component of matrix damage has been inferred from numerous experiments and semi-empirical models developed to describe the hardness contribution. However, as yet, there has been no unambiguous determination of the nature of these clusters. Phosphorous has been observed to form phosphides which contribute to hardening and embrittlement, but is also subject to irradiation-induced segregation to the boundary causing non hardening embrittlement.

Finally, it is important to stress the increasing importance of microstructural techniques to problems in operating reactors. For example, there is an increasing trend, which is most advanced in the UK, for dose-damage relationships for operating RPV's to be underpinned by mechanistic models []. The advantages of the models are that they can remove unnecessary conservatism in the development of the trend line or in the estimation of the scatter band. Microstructural techniques are then essential to validate the fundamental assumptions of the models and help parametise them. The activities range from verifying major assumptions such as demonstrating that copper precipitation reaches a plateau, or that over-ageing does not occur under irradiation, to investigating the finer mechanisms such as the rate of copper precipitation, and the composition of the precipitates. Individual issues relevant to older plant can be addressed. Here there are frequently problems over archive material; either in saying that a given material is a genuine archive or in gathering evidence that there have not been differences in heat treatment: etc. The former can be achieved by fairly routine microscopy etc. The latter, in high copper welds, is more sophisticated, as the STEM techniques can be used to measure the amount of copper available for irradiation embrittlement. Some may have precipitated out during the post-weld heat treatment, and differences between different samples can be investigated. Specimens may be in the form of scrapings or small volumes of material cut from the RPV: techniques have been developed to interrogate these.
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Table I. Stages in cascade development.

<table>
<thead>
<tr>
<th>Stage</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt; 0.2 psec (Collisional phase)</td>
<td>Heavily damaged region expanding rapidly, large number of displaced atoms.</td>
</tr>
<tr>
<td>0.2 - 0.3 psec</td>
<td>Still exploding but interstitials escape along replacement collision sequences (r.c.s). Considerable motion in the cascade centre, region becomes very hot. Increased density ring around heavily damaged region.</td>
</tr>
<tr>
<td>0.3 - 0.6 psec (Thermal Spike)</td>
<td>Thermal spike, molten region in the cascade centre which cools slowly. Energy released by recombination of interstitials and vacancies.</td>
</tr>
<tr>
<td>0.6 - 5 psec (Cooling Phase)</td>
<td>Cooling phase with appreciable defect motion and recombination in hot region. Clustering of point defects may occur.</td>
</tr>
<tr>
<td>4 - 10 psec</td>
<td>Final cooling down, only interstitials in clusters or those ejected along r.c.s survive.</td>
</tr>
</tbody>
</table>
Table II  Critical issues and outstanding questions.

<table>
<thead>
<tr>
<th>Critical Issues/Conclusions</th>
<th>Results to date</th>
<th>Outstanding work required/underway</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Outstanding Questions.</strong></td>
<td>Technique(s) used to address them</td>
<td></td>
</tr>
<tr>
<td>Precipitates and matrix composition. Most attention to Cu ppts. Precipitates alloyed with Mn and Ni. No evidence to support overaging in the irradiated material</td>
<td>Good agreement between techniques SANS, TEM/STEM and AP/FIM capable of providing information on Size, Nc and composition. Transformation of ppts followed in thermally aged alloys by TEM, HREM, EXAFS and MD. Structure of irradiation induced ppts confirmed by EXAFS to be bcc as in 'peak hardness' thermally aged condition.</td>
<td>Matrix composition can be assessed experimentally by STEM and AP/FIM Role of annealing on ppt stability requires post irradiation annealing and re irradiation experiments Controlled accelerator based irradiation may be beneficial in understanding the role of cascades in the stability of precipitates under irradiation.</td>
</tr>
<tr>
<td>Factors controlling nucleation and growth. Dose, Dose rate, Temperature and steel composition. Factors controlling the initial distribution of embrittling species such as copper. Other types of precipitates (eg Phosphides, Mo,V Carbides/Nitrides etc.). Why no overaging of copper ppts in irradiated material.</td>
<td>Limited results by TEM,HREM, FIM and SANS Growing PA data. Interpretation currently very difficult due to other contributions to the signal eg precipitates. A combined PA,IF and e Res on a series of well characterised model alloys, linked into a well controlled annealing kinetics experiment. Experimental work using accelerator based irradiations combined with modelling may provide useful insight.</td>
<td></td>
</tr>
<tr>
<td>Matrix defects, they exist but not understood in what form. What are the matrix defects? (Sponges, microvoids, loops (vacancy or interstitial)) Size and number density. Factors controlling production, irradiation and materials variables (both microstructure and impurity levels eg CNO)</td>
<td>Techniques exist to analyse grain boundaries (AP/FIM, STEM, Auger and SIMS) Computer modelling successful in other systems (eg IASCC in Austenitics)</td>
<td></td>
</tr>
<tr>
<td>Non hardening embrittlement, generally associated with grain boundary segregation of elements such as P,S,As Sn etc. Problem confined to specific materials Identification of the mechanism(s) and factors controlling segregation. what are the important irradiation and materials variables. Understanding of the balance between thermal and irradiation induced segregation processes.</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>


### Table III Techniques Available for the Characterisation of RPV Steels

<table>
<thead>
<tr>
<th>Technique Application (Example References)</th>
<th>Information Available Limitations/comments.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transmission Electron Microscopy (TEM)</td>
<td>Strain field of small precipitates and defect clusters. Number density, size and structure of features with diameter above the visibility limit of ~2 nm. In-situ experiments on dislocation pinning and precipitation kinetics. Standard sample geometry 3mm diameter 0.05mm thick discs. Analysis limited to relatively small areas (~mm²) less than 0.1-0.5µm thick. Microchemical information by electron energy dispersive X-ray (EDX) and electron energy loss spectroscopy (EELS). Resolution may be adversely affected by magnetic nature of material or oxide formation on the surface.</td>
</tr>
<tr>
<td>P (M) B Example Refs 35-49</td>
<td></td>
</tr>
<tr>
<td>Field Emission Gun Scanning Transmission Electron Microscopy (FEGSTEM)</td>
<td>Microchemical information on the spatial distribution of (embrittling) elements i.e. composition of small precipitates or precipitate free regions of the matrix, and grain boundaries. Beam size ~1.5nm, positioned with nm resolution to give elemental composition by (EDX) or (EELS), typically can give concentrations (Cu) above ~0.05 wt%</td>
</tr>
<tr>
<td>P B Example Refs 35,36,40,48,50</td>
<td></td>
</tr>
<tr>
<td>Electron Probe Micro Analysis (EPMA)</td>
<td>Chemical analysis of the bulk composition by EDX or X-ray wavelength spectroscopy. Sample volume analysed - µm³. Elemental mapping of bulk composition changes across weld runs etc. Analysis of inclusions and second phase particles.</td>
</tr>
<tr>
<td>P Example Refs 51</td>
<td></td>
</tr>
<tr>
<td>High Resolution Electron Microscopy (HREM)</td>
<td>Lattice imaging thin regions containing precipitates and/or matrix defects such as dislocation loops. Size and structure of the precipitates/defects. Limited to extremely thin samples &lt;20nm thick, requires very clean samples, gun damage can be a problem in the identification of lattice defects.</td>
</tr>
<tr>
<td>P M B Example Refs 36,52</td>
<td></td>
</tr>
<tr>
<td>Field Ion Microscopy and 3D Atom Probe (FIM) (POSAP or OAP)</td>
<td>Atomic resolution and atomic composition of small regions of the matrix, precipitates and grain boundaries. Very high spacial resolution, chemical composition isotope sensitive, can be both an advantage and a disadvantage. Relatively small volume analysed, typically only ~5000 nm³.</td>
</tr>
<tr>
<td>P M B Example Refs 35,36,42,53-56</td>
<td></td>
</tr>
<tr>
<td>Small Angle Neutron Scattering (SANS)</td>
<td>Size and number density of precipitates in bulk specimens. Typically 10 x 10 x 2 mm. Magnetic A ratio measurements also permit some insight into the chemical composition of the scattering centres. Resolution ~1 nm.</td>
</tr>
<tr>
<td>Diffuse Elastic Neutron Scattering (DENS)</td>
<td></td>
</tr>
<tr>
<td>P (M) Example Refs 35,36,41,55,59,60</td>
<td></td>
</tr>
<tr>
<td>Technique</td>
<td>Description</td>
</tr>
<tr>
<td>-----------------------------------------------</td>
<td>-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>Anomalous Small Angle X-Ray Scattering (ASAXS)</td>
<td>Size and number density of precipitates, elemental specific. Technique similar to SANS but with limited depth penetration.</td>
</tr>
<tr>
<td>Example Refs 66,69</td>
<td></td>
</tr>
<tr>
<td>Extended X-Ray Absorption Fine Structure (EXAFS)</td>
<td>Provides data on the average atomic environment of embrittling species such as copper and nickel. Currently limited to simple model alloys as data interpretation difficult in complex steels. Bulk sampling technique.</td>
</tr>
<tr>
<td>Example Refs 36,70,71,72</td>
<td></td>
</tr>
<tr>
<td>Positron Annihilation (PA)</td>
<td>Lifetime, Doppler broadening and angular correlation techniques. Information on both matrix defects and precipitates. Association of vacancy clusters and contribution of vacancies to the precipitate kinetics. Non destructive bulk sampling technique, samples typically 10 x 10 x 2 mm. Full mechanistic understanding currently limited to model alloys, as data interpretation difficult in complex steels. Increasingly being used in an empirical way to correlate with mechanical properties data. Can also be employed in ND testing of plant components.</td>
</tr>
<tr>
<td>Example Refs 65,66,73-78,79-81</td>
<td></td>
</tr>
<tr>
<td>Internal Friction (IF)</td>
<td>Low frequency: Gaseous impurities C,N and O in or out of solution (Snoek effect). Orientation changes in point defect complexes related to elastic anisotropy. High frequency: Dislocation interactions (Granato-Lucke effect)</td>
</tr>
<tr>
<td>Example Refs 82-87</td>
<td></td>
</tr>
<tr>
<td>Mossbauer Spectroscopy (MS)</td>
<td>Recoilless nuclear resonance absorption of gamma radiation. Bulk analysis technique, average environment of the mossbauer nucleus. Precipitation of copper, carbon in solution or in carbides. Interpretation difficult in complex steels.</td>
</tr>
<tr>
<td>Example Refs 88</td>
<td></td>
</tr>
<tr>
<td>Muon Spin Rotation (μSR)</td>
<td>Bulk technique requiring high energy particle accelerator. Point defects, defect clusters and precipitation kinetics followed by changes in depolarisation, commencement of ppt ripening detected by changes in precession frequency. Application to complex steels not fully understood.</td>
</tr>
<tr>
<td>Example Refs 89</td>
<td></td>
</tr>
<tr>
<td>Barkhausen Emission and Magneto Acoustic Emission (BE,MAE)</td>
<td>Related non destructive bulk analysis techniques. Detect interactions between moving magnetic domain walls and pinning points (copper precipitates?) Currently lacks sensitivity in application to RPV steels.</td>
</tr>
<tr>
<td>Example Refs 90-94</td>
<td></td>
</tr>
<tr>
<td>Molecular Dynamics (MD)</td>
<td>Computer simulation of the atomic interactions in materials. Understanding of the properties and transformation of bcc copper. Used in the study of irradiation induced defect complexes. Good elemental potentials but currently no realistic Fe alloy potentials as yet available. Limited to small volumes due to computer and memory intensive calculations.</td>
</tr>
<tr>
<td>Example Refs 36,95-97</td>
<td></td>
</tr>
<tr>
<td>Analysis</td>
<td>Description</td>
</tr>
<tr>
<td>----------</td>
<td>-------------</td>
</tr>
<tr>
<td>Scanning Auger Microscopy (SAM)</td>
<td>Analysis of the fracture surface providing surface specific microchemical information on embrittling species such as P. In situ fracture of specimen required to avoid surface contamination.</td>
</tr>
<tr>
<td>Static / Dynamic Secondary Ion Mass Spectroscopy (SSIMS / DSIMS)</td>
<td>Microchemical information on materials provided by spectral analysis of sputtered ions. In principle SSIMS can be used for assessment of segregation to grain boundaries (fracture surface), requires good time of flight detector. DSIMS good qualitative elemental mapping, grain boundary segregation on cross section samples. Through thickness composition of large inclusions and precipitates. Diffusion kinetics can be followed due to isotopic sensitivity.</td>
</tr>
<tr>
<td>Scanning Tunnelling Microscopy (STM)</td>
<td>Surface structural assessment with atomic resolution. May provide information on large overaged precipitates, offers possibility to identify subsurface features by magnetic anomaly when used in conjunction with a magnetic tip. Sensitive to any oxide layer and/or surface contamination giving rise to problems in data interpretation.</td>
</tr>
<tr>
<td>Electrical Resistivity</td>
<td>Bulk sampling technique used in the assessment of point defect and defect complexes. Interpretation more difficult in the complex steels.</td>
</tr>
<tr>
<td>Lattice Parameter Measurements</td>
<td>Changes in lattice parameter associated with solute additions, precipitates and matrix defects. Requires further work to validate the technique and its application to model alloys and RPV steels.</td>
</tr>
<tr>
<td>High Precision Dilatometry/Density</td>
<td>Accurate assessment of volume changes associated with the production of matrix defects. Currently unknown capability in the area of RPV steels. Considerable data and expertise exists in applying the technique to other systems ie. irradiation induced growth and void swelling.</td>
</tr>
<tr>
<td>Computer modelling</td>
<td>A range of modelling codes are available to assist in a wide range of RPV embrittlement issues, these range from codes designed to help interpret experimental data, predictions of irradiation induced phenomena, through to modelling of the embrittlement behavior of vessels. Examples include: dosimetry, recoil spectra, segregation, precipitation, TEM image simulation, positron lifetimes, SANS maximum entropy, etc... All models require a degree of experimental input for validation.</td>
</tr>
</tbody>
</table>
Table IV. Matrix Cu Levels Measured in Fe 1.1 at% Cu aged at 550C [8]

<table>
<thead>
<tr>
<th>Technique</th>
<th>SANS Cu at%</th>
<th>STEM Cu at%</th>
<th>FIM/AP Cu at%</th>
</tr>
</thead>
<tbody>
<tr>
<td>A/Q</td>
<td>1.1</td>
<td>1.1</td>
<td>1.0</td>
</tr>
<tr>
<td>10min</td>
<td>0.99</td>
<td>0.8</td>
<td>-</td>
</tr>
<tr>
<td>2 hr</td>
<td>0.56</td>
<td>0.4</td>
<td>0.4</td>
</tr>
<tr>
<td>10 hr</td>
<td>0.20</td>
<td>-</td>
<td>0.1</td>
</tr>
</tbody>
</table>
Table V.
FeCu After Thermal Ageing at 550°C,
Values in Vicker Hardness.[10]

<table>
<thead>
<tr>
<th>Ageing time at 550°C</th>
<th>Measured Hardness</th>
<th>Precipitate hardening predicted from Russell and Brown (1972) theory</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>ΔHAV(DT)</td>
<td>fcc Cu modulus (Θ&gt;50°)</td>
</tr>
<tr>
<td>5 min</td>
<td>37</td>
<td>23</td>
</tr>
<tr>
<td>10 min</td>
<td>55</td>
<td>34</td>
</tr>
<tr>
<td>2 h</td>
<td>100</td>
<td>72</td>
</tr>
<tr>
<td>10 h</td>
<td>76</td>
<td>58</td>
</tr>
</tbody>
</table>

Shear Modulii; fcc - 4.8x10^4 MPa; bcc - 3.0x10^4 MPa
Table VI.
Effect of heat treatment on high copper weld [17]

<table>
<thead>
<tr>
<th>Condition/Heat Treatment</th>
<th>Analysis method/ Cu level in the ferrite</th>
</tr>
</thead>
<tbody>
<tr>
<td>As received</td>
<td>Wet chemistry</td>
</tr>
<tr>
<td>Bulk Chemistry</td>
<td>0.56</td>
</tr>
<tr>
<td>As received</td>
<td>Calculated from bulk data</td>
</tr>
<tr>
<td>Copper effective</td>
<td>0.52</td>
</tr>
<tr>
<td>(assumes all S in the</td>
<td></td>
</tr>
<tr>
<td>form of Cu$_1$S)</td>
<td></td>
</tr>
<tr>
<td>Post-weld heat</td>
<td>Measured by STEM</td>
</tr>
<tr>
<td>treatment of 12h at</td>
<td>0.39 ± 0.04</td>
</tr>
<tr>
<td>600°C and 6h at 600°C,</td>
<td>min 0.36</td>
</tr>
<tr>
<td>Followed by a quench</td>
<td>max 0.45</td>
</tr>
<tr>
<td>from 950°C and</td>
<td></td>
</tr>
<tr>
<td>tempered for 42h at</td>
<td></td>
</tr>
<tr>
<td>600°C and then 6h at</td>
<td></td>
</tr>
<tr>
<td>650°C</td>
<td></td>
</tr>
<tr>
<td>As above plus</td>
<td>Measured by STEM</td>
</tr>
<tr>
<td>a further 5h at 750°C</td>
<td>0.49 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>min 0.42</td>
</tr>
<tr>
<td></td>
<td>max 0.53</td>
</tr>
</tbody>
</table>
Figure Captions.

Figure 1. Typical light water reactor spectrum at various depths in the vessel wall. (Distances in depth from inner surface).

Figure 2. Percentage of neutrons at the surface, 1/4T and 3/4T which are greater than 0.1MeV and 1MeV respectively.

Figure 3. Histograms of recoil spectra for the inner surface and 1/4T position of the vessel wall.

Figure 4. The damage efficiency factor, k, for defect production at 100 K in copper with recoil energies from 60 eV to 10 keV and random knock-on directions (solid line). The production of single and di-interstitials is also shown (dotted line).

Figure 5. Modelling interpretation of Magnox submerged-arc weld surveillance data [26].

Figure 6. Particle size distributions for FeCuNi thermally aged for 2 and 10 hrs. at 550°C (a) derived from maximum entropy analysis of SANS data (b) TEM derived size distributions. In both cases a bi-modal distribution can be seen.

Figure 7. The various contributions to the overall observed hardening in aged FeCu alloy.
PWR vessel wall

FIGURE 2

% of neutrons

Surface 1/4T 3/4T

>0.1 MeV

>1 MeV
% of recoils

- 0.04 - 0.1 keV
- 0.1 - 1.0 keV
- 1.0 - 5.0 keV
- 5.0 - 10 keV
- 10 - 30 keV
- 30 keV

PWR Surface

PWR 1/4 T

FIGURE 3
Defect Yield = \( k \frac{E_{\text{dam}}}{2E_d} \)

Temperature = 100K

FIGURE 4

PKA Energy \( E \) (keV)

Efficiency factor \( k \)

0 Alpha iron

Single and di-SIA

Total

(15) (10) (15) (16) (10) (7) (5) (5)
\[ \Delta T (\text{Cu}) + \Delta T (\text{matrix}) \]

\[ \Delta T (\text{matrix}) \]

**Figure 5**
**Precipitate number density (m^-3 nm^-1)**

**Relative frequency**
FIGURE 7

Measured

Cu-Precipitation

Quenched in Voids

Cu-Solid Solution

Dislocations

Grain Boundaries

Fe Matrix

Ageing time (h)

Hardness Hv (20)
CHAPTER 10

Modeling Irradiation Embrittlement in Reactor Pressure Vessel Steels

G. R. Odette

University of California Santa Barbara
I. INTRODUCTION

Irradiation embrittlement of nuclear reactor pressure vessel (RPV) steel is controlled by interactions between a large number of environmental and metallurgical variables [1]. Hence, purely empirical predictions of embrittlement are unreliable, particularly outside the database. Embrittlement models synthesize experimental and theoretical information from a diverse array of sources to quantitatively predict property changes based on physical understanding. Simple models developed to date have been very useful to developing engineering correlations and rationalizing embrittlement trends [1-9].

Indeed, models have probably had a greater impact on the engineering practice used to forecast and control RPV embrittlement than for any other application of radiation effects research. In spite of this success, however, a number of outstanding issues remain, including:

- Extending embrittlement correlations, such as the US Nuclear Regulatory Commission Regulatory Guide 1.99, to include the effects of flux, spectrum, irradiation temperature, phosphorous and other metallurgical variables, as well as interactions among key variables;

- Interpretation and utilization of accelerated materials test reactor (MTR) data;

- Interpreting outliers;

- Avoiding technical surprises in service regimes that are not well-characterized or have not yet been encountered.

Improved models, coupled with an appropriate engineering data base and fundamental experiments, are needed to address these issues.

The models described in this chapter are restricted to a case when embrittlement can be related to yield stress increases. Specific focus is on Charpy-V notch indexed transition temperature shifts in Mn-Mo steels irradiated at around 290°C. Modeling is divided into the following elements: a) primary defect production of vacancies and interstitial and small defect clusters in displacement cascades; b) evolution of extended microstructural and microchemical features driven by excess defect and solute fluxes; c) increases in the yield stress due to these features; and d) elevation of transition temperatures associated with the yield stress increases. These elements are integrated to model the combined effects of metallurgical and environmental variables on transition temperature shifts.

The emphasis of this Chapter is on microstructural modeling (b). The most salient features of the other elements (a, c, d and integrated models) are summarized briefly. Particular attention is given to the effects of key irradiation and metallurgical variables, including flux, fluence, irradiation temperature and alloy composition. Since the focus is on modeling and modeling techniques, comparisons with experimental observations are only very briefly drawn. Many of the concepts and formulations are presented for the first time.
II. PRIMARY DEFECT PRODUCTION

Vacancies and interstitials generated in displacement cascades are the fundamental source of irradiation embrittlement. The accepted measure of primary damage exposure is displacements per atom (dpa), or the calculated average number of vacancy-interstitial pairs initially created by neutron irradiation [10]. The dpa are determined from tabulated dpa-cross sections, \( \sigma_{dpa}(E) \) [11], the neutron energy (E) spectrum \( \phi(E) \) and the fluence, \( \phi_t \). The dpa-cross sections are evaluated by combining calculated primary-knockon-atom recoil energy \( (T_r) \) cross sections, \( K_r(E,T_r) \), with a model of the average number of displacements generated as a function of the primary recoil energy, \( v_{dpa}(T_r) \) [12]. Computer codes, incorporating appropriate nuclear reaction kinematics models and requisite nuclear data (i.e. from ENDFB-V) are used to calculate \( K_r(E,T_r) \) [13]. The displacement function, \( v_{dpa}(T_r) \), is approximated by \( 0.8T_{dam}/2E_d \), where \( E_d \) is an effective average displacement energy [14] and \( T_{dam}(Z,T_r) \) is the part of the primary recoil energy deposited as kinetic energy in all cascade recoils (versus energy deposited in electron excitation and ionization, which does not cause permanent damage in metals) [15]. Since \( E_d \) is a constant, dpa is fully equivalent to the absorbed kinetic energy or effective dose [16].

The total fast fluence for neutrons with energies greater than 1 MeV, \( \phi_t (E>1 \text{ MeV}) \), is a less useful, but more common measure of exposure. There is no physical or empirical rationale for using this or any other threshold fluence, if a significant component of dpa or dose is created by neutrons with lower energies [10,16]. Calculations of dpa should account for neutrons at all energies. However, the spectral averaged displacement cross sections, \( \sigma_{dpa} \), can be normalized to the fraction of the neutron spectrum above a specified energy. For example, typical materials test reactor (MTR) and surveillance locations have \( \sigma_{dpa} \) of about \( 1500 \pm 200 \) barns \( (10^{-24} \text{ cm}^2) \) for \( E>1 \text{ MeV} \). Since they are equivalent when related by the appropriate cross section, the more familiar fast fluence and fast flux, \( \phi \), notation is used in the following sections.

The best physical measure of primary radiation damage production is the absolute number and configuration of vacancy and interstitial defects following local short term rearrangement within the displacement cascade. This rearrangement, or cascade annealing, results in vacancy-interstitial recombination and clustering. Cascade modeling is an active area of ongoing research and will not be reviewed in detail. Most recent cascade simulations have been carried out with many-bodied molecular dynamics (MD) codes using embedded atom (EA) or equivalent multibody interatomic potentials [17-20]. Earlier work was based on cascade codes using the pair potentials and most often using the binary-collision approximation (BCA) to determine the general configuration of the cascade defects [14]. The predicted defect configurations from BCA and early MD cascade models have been coupled with simulations of short term diffusional cascade annealing (STA) [21,22]. Most studies have simulated cascades in fcc copper; however, very recent research suggests that defect production behavior in bcc iron is generally similar [19]. The key conclusions of both the MD-EA models and BCA-STA studies can be summarized as follows:
The total residual defect production efficiency, $\xi_t$, defined as the ratio of the surviving defects to dpa, decreases from about 1 for very low energy recoils to about 0.25 to 0.45 at energies above about 0.5 to 1 keV; the variation of $\xi_t$ with $T_{\text{dam}}$ is relatively slow at higher energies.

A significant fraction of residual defects are contained in clusters of more than two defects. Maximum cluster sizes are 10 or greater. Clustering can be grossly defined in terms of the fraction of dpa contained in clusters, $\xi_{\text{cl/v}}$. The fractional clustering and average and maximum cluster size increase with increasing recoil energy. Clustering is also a function of the defect type, cluster size and temperature and is sensitive to the details of the MD or BCA/STA model. Various models predict clustering of 40% to more than 90% of the residual defects. A reasonable estimate for $\xi_{\text{cl/v}}$ is in the range of about 0.10 to 0.3.

The corresponding estimate of the fraction of dpa in the form of mobile unclustered or 'freely migrating' residual defects, $\xi_{\text{fi/v}}$, is in the range of about 0.05 to 0.15. Note, while $\xi_t = \xi_{\text{cl/v}} + \xi_{\text{fi/v}}$, denser cascades that produce more recombination also generally result in a higher clustering fraction; hence, $\xi_{\text{fi/v}}$ tends to be less than $\xi_{\text{cl/v}}$.

These conclusions are broadly consistent with observation. Various simulations have also shown that: vacancy-interstitial production is reduced at higher temperatures and by pre-existing lattice defects [19]; and that significant segregation of solutes may take place during STA, leading to the formation of small defect cluster-solute complexes [23].

Essentially all physically plausible measures of defect production attenuate in the pressure vessel less rapidly than $\phi t$ (E > 1 MeV) [24]. Hence, there is a high probability that fast fluence is not only an inappropriate, but is also an unconservative exposure unit for extrapolating embrittlement predictions from surveillance positions to locations deep into the vessel.

As shown in the later sections of this Chapter, the arrangement of primary defects following short term cascade annealing plays a crucial role in subsequent microstructural and microchemical evolution. For example, cascades promote hardening by clusters while retarding precipitation; the balance between these competing effects depends on the copper content of the steel [25]. Hence, continued MD-EA simulations of the production of various types of primary defects are critical to developing improved embrittlement models. The MD-EA cascade results should be combined with rigorous STA simulations to develop realistic defect production cross sections. Finally, it is noted that MD and other advanced modeling methods (i.e. density functional techniques) can be used to characterize the key material properties used in embrittlement models. These properties include a wide variety of defect and solute solution, migration, interaction, interface and free surface energies.

### III. MICROSTRUCTURE-YIELD STRESS RELATIONS

The dominant form of embrittlement in Mn-Mo RPV steels, involving transgranular cleavage fracture, is due to radiation induced increases in the yield stress. Fine scale
precipitates and extended defects formed under irradiation increase the yield stress by acting as dispersed obstacles to dislocation motion. Models of yield stress increases must address two related questions:

How do the features increase the yield stress in isolation?

How do the contributions from the various radiation induced and pre-existing strengthening features combine?

Small obstacles may interact with dislocations by a variety of mechanisms including chemical bond disruption/disordering, surface/interface creation, incommensurate/higher resistance slip systems, stacking faults, image forces, strain fields and modulus mismatches [26]. For coherent copper-rich precipitates (subscript crp) and larger vacancy aggregates, or nanovoids (subscript nv), the modulus mismatch mechanism is dominant. The modulus interaction has been modeled by Russell and Brown [27] based on decrease of the dislocation strain energy in a more compliant obstacle relative to the stiffer iron matrix as:

$$\Delta \sigma_{rb} = S_m \mu_{fe} b \left(1 - \frac{\mu_{rb} \log \left(\frac{r_{rb}}{r_{ic}}\right)}{\mu_{fe} \log \left(\frac{r_{oc}}{r_{ic}}\right)} + \frac{\log \left(\frac{r_{oc}}{r_{ic}}\right)}{\log \left(\frac{r_{oc}}{r_{ic}}\right)} \right) \frac{3/4}{1.77 r_{rb}}$$

where \( f_{rb} \) is the volume fraction of the features, \( S_m \) is the Schmidt factor, \( \mu_{fe} \) and \( \mu_{rb} \) are the shear moduli of iron and the softer feature with a radius \( r_{rb} \), \( b \) (0.248 nm) is the Burger's vector, and \( r_{ic} \) and \( r_{oc} \) are the inner (dislocation core) and outer strain field cut-off radii. Russell and Brown found good agreement between the model predictions and peak hardening data in the iron-copper (subscript cup) alloys for: \( r_{ic} = 2.5 b = 0.62 \) nm; \( r_{oc} = 2500 b \); \( \mu_{cup}/\mu_{fe} = 0.6 \); and \( S_m = 2.5 \). Figure 1 plots \( \Delta \sigma_{cup}/\sqrt{f_{cup}} \) versus \( r_{cup} \) for these parameters; the peak for copper precipitates at about 3750 MPa is at \( r_{cup} = 1.25 \) nm. The corresponding curves for nanovoids (with \( \mu_{nv}/\mu_{fe} = 0.0 \)) and alloyed copper rich precipitates (with \( 0.6 \) Cu, \( 0.25 \) Mn \( 0.15 \) Ni and \( \mu_{crp}/\mu_{fe} = 0.75 \) based on a simple rule of mixtures estimate) with peaks at about 6950 and 2750 MPa respectively are also shown in Figure 1.

Strengthening contributions from other features can be generally represented as:

$$\Delta \sigma_j = S_m \beta_j \mu_{fe} b \sqrt{2 r_j N_j}$$

where \( r_j \), \( N_j \) and \( \beta_j \) are the radius, number density and the strengthening factor of the j’th feature respectively [26]. Strengthening by interstitial clusters (subscript 1) arises from dislocation interactions with the large tetragonal strain fields produced by small loops. Fleisher [28] derived a loop strengthening factor, \( \beta_l \), of 0.27 for fcc crystals. The strengthening factor for loop orientations and strain fields in bcc-iron may not be exactly the same; however a \( \beta_l \) value of 0.3 seems a reasonable approximation. The yield stress increases due to small vacancy clusters generated in cascades (subscript vc) with radii less about 0.6 nm cannot be treated by the Russell-Brown model. A semi-empirical analysis of strengthening due to thermally unstable matrix damage generated at high fluxes, believed to
be small vacancy clusters, is consistent with a strengthening factor $\beta_{vc}$ of about 0.1 [24,25]. Because of their complex chemical bonding and incommensurate crystal structures, higher strengthening factors would be anticipated for other phases (subscript $p$) that may form under irradiation, such as phosphides and carbonitrides. At larger sizes (> 5 nm) such features would act as "strong" Orowan barriers with a $\beta_p$ of about 0.8 [26]. Considering the fine scale of precipitation under irradiation, a provisional value $\beta_p$ of 0.5 seems reasonable.

Modeling of irradiation hardening is also complicated by the need to combine the contributions of various sources of strengthening. The total yield stress $\sigma_y$ can be represented as [26]:

$$\sigma_y = \sigma_{fr}(\dot{\varepsilon}, T) + \sigma_{irs} + \sigma_d + k d^{-1/2}$$

where $\sigma_{fr}(\dot{\varepsilon}, T)$ is a test temperature ($T$) and strain rate ($\dot{\varepsilon}$) sensitive frictional contribution from the inherent lattice resistance to slip and solutes. In these cases, thermal (activation) fluctuations and applied stress combine to mediate the slip process. The remaining terms are essentially athermal (e.g., only weakly dependent on strain rate and temperature). The $kd^{-1/2}$ represents Hall-Petch type grain strengthening. The $\sigma_{irs}$ term is from long range stress fields such as dislocations (about $\mu b\sqrt{\rho}$, where $\rho$ is the dislocation density). Radiation primarily changes the on the athermal dispersed obstacles contribution, $\sigma_d$, thus the yield stress increase is

$$\Delta \sigma_y = \Delta \sigma_d$$

Computer simulations by Foreman and Makin [29] suggest that the net yield stress contribution for two types of dispersed obstacles depend on their relative strengths. For obstacles with similar strengths (weak/weak or strong/strong), the root square sum (RSS) law

$$\Delta \sigma_d = \sqrt{\Sigma \Delta \sigma_j^2}$$

is believed to be most appropriate. Similarly this law applies to the case where the number of weak barriers is comparable to or less than the strong barriers. In this circumstance the weak barriers may have a small or even negligible effect on the yield stress, since they will be penetrated at a stress lower than that required to bow the dislocation around the strong obstacles, as illustrated in Figure 2a. The case of many weak barriers and a few strong obstacles is more complex since both features influence the configuration and effective stress required to bow a dislocation around the strong obstacles. The computer simulations suggest that in this case the net yield stress contribution falls between the linear sum (LS) and RSS extremes. However, in the limit of strengthening from many weak and a few strong obstacles, the contribution from the former can be approximated as an additive LS friction stress, thus

$$\Delta \sigma_d = \Sigma \Delta \sigma_j$$
Prior to irradiation complex RPV steels typically contain a significant density ($> 10^{16}/cm^3$) of fine scale (< 10 nm) phases (i.e., carbonitrides) [30] which produce a significant dispersed obstacle hardening ($\sigma_{do} > 50$ to 100 MPa). Irradiation typically results in a much higher number density of weak-to-medium strength obstacles. In this case the LS law (Equation 5b) is most appropriate. Hence, the net yield stress increase is approximately equal to the net contributions from the features formed during irradiation, $\Delta \sigma_i$,

$$\Delta \sigma_y = \Delta \sigma_i = \sqrt{\Delta \sigma_{crp}^2 + \Delta \sigma_{nv}^2 + \Delta \sigma_{vc}^2 + \Delta \sigma_{p}^2}$$

(6a)

where the contributions of the various features formed under irradiation are combined by the RSS law (Equation 5a). However, for irradiations at high temperatures or following post-irradiation annealing, the number density of the weaker irradiation induced features is much lower. In this case the RSS law is more appropriate, hence

$$\Delta \sigma_y = \sqrt{\sigma_{do}^2 + \Delta \sigma_i^2} - \sigma_{do} < \Delta \sigma_i$$

(6b)

The effect of the alternative superposition models is illustrated in Figure 2b for a $\sigma_{do}$ of 100 MPa.

The large yield stress recovery following post irradiation annealing has been attributed to a change in superposition from the linear to root square sum control [31]. In this case the overall decrease in yield stress significantly exceeds the change in the isolated contribution from the irradiated versus irradiated and annealed features.

There are considerable uncertainties in the hardening models the copper rich precipitates and vacancy clusters particularly at small sizes ($r < 0.8$ nm); and, more generally, for the other features (which have not yet been fully identified). For example, the effects of composition, small size-scales and non-equilibrium structures (e.g., bcc versus fcc) on the properties of copper rich precipitates are not known. Application of detailed MD-EA calculations to dislocation-obstacle interactions could be used to refine strengthening models. Further, computer simulations should be carried out to develop superposition laws appropriate to irradiated and annealed microstructures. In both cases the models must be validated by well-designed experiments.

IV. TRANSITION TEMPERATURE SHIFTS

Modeling the relationship between the microstructure-sensitive basic properties, such as yield stress, and macroscopic measures of fracture resistance, such as of Charpy transition temperatures, involves a combination of micromechanics and macromechanics. Continuum macromechanics requires only an appropriate constitutive description of the material to relate the remote specimen loads and displacements, generally characterized in terms of the elastic stress intensity factor, $K$, or the elastic-plastic energy release rate integral, $J$, to the corresponding stress and strain fields local to the notch (or crack) tip [32]. Sophisticated continuum models are typically based on large deformation finite element methods [33]. Micromechanics models require a set of additional structure-sensitive material properties to
describe the critical notch (or crack) tip fields that lead to fracture. The primary focus of this chapter is on transgranular cleavage fracture in Charpy-V notch (blunt) specimens. However, as briefly noted below, the principles can be extended to modeling various measures of fracture toughness for sharp cracks.

The accepted mechanism for cleavage fracture is that the stress near the notch tip normal to the crack plane, \( \sigma_{22} \), reaches a critical stress level \( \sigma^*_{f} \) needed to trigger the extension of an unstable microcrack [34-36]. The peak \( \sigma_{22} \) stresses are higher than the uni-axial yield stress due to the notch stress concentration and the triaxial plane strain constraint. The stress amplification can be described by the constraint factor, \( Q \), as

\[
\sigma_{22} = Q(P/P_{gy})\sigma_y(T, \dot{\varepsilon})
\]  

(7)

where \( P/P_{gy} \) is the load to general yield load ratio. Near general yielding (\( P/P_{gy} \geq 0.8 \)) \( Q \) is about 2.2 for a Von Mises yield criteria. Hence the elastic cleavage condition requires that

\[
\sigma_{22} = 2.2\sigma_y(T, \dot{\varepsilon}) = \sigma^*_{f}
\]  

(8)

or

\[
T_c = \sigma^{-1}_y(\sigma^*_{f} / 2.2, \dot{\varepsilon})
\]  

(9)

where \( T_c \) is the maximum temperature (minimum \( \sigma_y \)) for elastic cleavage fracture or the cleavage transition temperature.

The microcracks typically propagate from grain boundary carbides at critical stress levels, \( \sigma^*_{f} \), inversely proportional to the square root of the 'largest' carbide size [35]. Since changes on this size scale do not take place, \( \sigma^*_{f} \) is expected to be unaffected by irradiation. Further, microcleavage fracture theories suggest that \( \sigma^*_{f} \) is only weakly dependent or independent of test temperature. This has been confirmed in an extensive study of irradiated Mn-Mo RPV steels [36]. This research showed that the cleavage transition temperature \( T_c \) corresponds to about 10±5 J of absorbed energy in a standard Charpy test. It was further observed that irradiation induced yield stress increases are approximately independent of test temperature and strain rate. Thus the shift in the cleavage transition temperature, \( \Delta T_c \), can be calculated without explicit knowledge of \( \sigma^*_{f} \) as

\[
\Delta T_c = \int \sigma_{yd}(T, \dot{\varepsilon}) \Delta T_d \frac{d \sigma_{yd}}{dT}(T_d) \sigma_{yd}(T, \dot{\varepsilon})^{-1} d \sigma_{yd}
\]  

(10)

where \( T_{co} \) is the unirradiated cleavage transition temperature, \( \sigma_{yd} \) is the dynamic yield stress for the strain rate characteristics of Charpy impact tests (about 140s^{-1}). The slope of the dynamic yield stress versus test temperature (\( d \sigma_{yd}/dT \)) can be determined from a master curve for RPV steels in the unirradiated condition [36]. The procedure for evaluating \( \Delta T_c \) is
schematically illustrated in Figure 3a. The strengthening-cleavage shift relationship can be expressed as

$$\Delta T_c = \Sigma_c(T_{co}, \Delta \sigma_y) \Delta \sigma_y$$  \hspace{1cm} (11)

The cleavage shift coefficient, $\Sigma_c$, increases with increasing $T_{co}$ and $\Delta \sigma_y$ due to the decrease in $(d \sigma_y/d T_c)$ with increasing temperature [36].

The Charpy transition temperature shift $\Delta T_{CV}$ is normally indexed at an impact energy of 41J in the transition region rather than at 10 J. In this case there is an additional component of shift due to the effect of embrittlement on the ductile fracture resistance of the steel, manifested in drops as the upper-shelf-energy (USE). While the extra component of shift, $\Delta T_d$, for ductile fracture in Charpy specimens has not been theoretically modeled, it can be estimated based on an empirical observation that the transition region in RPV steels has an approximately constant temperature interval of about 120±25°C [36]. This leads to a relation

$$\Delta T_d \approx \frac{3720}{USE_o} \left( \frac{f_{use}(\Delta \sigma_y)}{1-f_{use}(\Delta \sigma_y)} \right)$$  \hspace{1cm} (12)

where the fractional reduction, $f_{use}(\Delta \sigma_y)$, of the unirradiated upper-shelf-energy, $USE_o$, is a known empirical function of $\Delta \sigma_y$ [36]. The procedure for evaluating $\Delta T_d$ is schematically illustrated in Figure 3b. The overall $\Delta T_{CV}$ at 41J is simply

$$\Delta T_{CV} = \Delta T_d + \Delta T_c = \Sigma_{cv}(USE_o, T_{co}, \Delta \sigma_y) \Delta \sigma_y$$  \hspace{1cm} (13)

For typical values of unirradiated properties ($USE_o$ and $T_{co}$) and yield stress increases, the predicted values of $\Sigma_{cv}$ range from about 0.4 to 0.8°C/MPa, in good agreement with observation. If the unirradiated Charpy curve is represented in terms of a tanh function, similar procedures can be used to estimate changes in the tanh parameters after irradiation, as demonstrated in Figure 4.

Similar theories can be applied to model the effects of irradiation on fracture toughness, $K_{IC}$, in the cleavage regime [8, 37-38]. In this case, however, the highly localized fields in the vicinity of blunting cracks require additional material properties to describe the fracture condition. In the simplest case, this property is a critical microstructural distance that must experience a critical stress level (note this critical stress is not necessarily the same as the $\sigma_f$ for a blunt notch) [39]. Irradiation is not expected to change either the critical stress or the critical distance; thus, cleavage toughness indexed (e.g. at 75 or 100 MPaVm) temperature shifts, $\Delta T_K$, should also depend only on the unirradiated $T_{ko}$ and $\Delta \sigma_y$. The $\Delta T_K$ shifts can be calculated by integrating $(d \sigma_y/d T)$ analogous to Equation 10, subject to appropriate modifications for strain rate and irradiation induced changes in the strain hardening [8, 24]. Predictions of this simple model are generally in reasonable agreement with observed toughness indexed shifts. However, the model predicts that: a) the slope of the $K_{IC}$ versus temperature curve is decreased; and b) that $\Delta T_K$ is much greater than $\Delta T_{CV}$ for large yield
stress increases and/or high $T_{ko}$. Although relevant data are very limited, such effects have generally not been reported.

Future modeling efforts for Charpy tests should be directed at the transition and ductile fracture upper-shelf regimes. More significantly, however, research should be focused on the effect of irradiation on $K_{ic}$ in the cleavage, ductile and transition regimes as well as on other measures of toughness (i.e. resistance curves, crack arrest and dynamic). Particular emphasis should be placed on resolving the questions about a possible layover of the $K_{ic}(T_0)$ curve and the correspondence between toughness and Charpy shifts. These issues must be attacked on the fundamental level; for example, by developing a better understanding of the mechanisms controlling crack tip field conditions leading to cleavage (i.e., temperature effects, microcrack stability and statistical factors [40]). Once again MD-EA modeling techniques, perhaps coupled with finite element methods, could be used to provide new insight into microcrack nucleation and propagation mechanisms. Finally, advanced continuum and micromechanics models can also be used to model the consequences of deviations from small scale yielding fields for realistic geometries (i.e. shallow cracks) and a range of size scales (i.e., small specimens) [41].

V. POINT DEFECT BALANCE EQUATIONS AND RECOMBINATION

Modeling microstructural evolution requires evaluating the fates of diffusing vacancies, interstitials and solutes. These fates includes recombination, annihilation at fixed sinks (that do not evolve) or clustering to form extended defects or precipitates. The excess point defects also accelerate or induce solute diffusion. These phenomena can be approximately modeled using rate theory [42].

The rate theory models used in this work assume three types of sinks characterized by a strength, $S$: network dislocations, $S_p$; point defect clusters, $S_c$; and a surrogate class for "other" microstructural features, $S_o$. The sink strengths for diffusion controlled kinetics are: $S_c = 4\pi r_c N_c$ and $2\pi r_c N_c$ for spherical and disc shaped (loops) clusters, where $r_c$ and $N_c$ are the effective cluster radius and number density; and $S_p = \rho$ for dislocations, where $\rho$ is the dislocation density. The difference between sink strengths for interstitials and vacancies is modeled by an average bias factor, $B$, as $S_i = S_v(1+B)$; the models assume only dislocations (network and loops) are biased. The total sink strength $S_{total}$ is the sum of the contributions from the various features. At around 290°C the defect concentrations typically approach steady state levels in a few seconds [43]. The simplest form of the steady-state defect conservation equations are

$$G_{ii} - D_i C_i S_{ti} - R_{iv} C_i C_v = 0$$

$$G_{iv} - D_v C_v S_{iv} - R_{iv} C_i C_v = 0$$

where

$C_{iv}$ are the interstitial/vacancy concentrations (atom fraction)
are the total interstitial/vacancy production rates: $G_{tv} = G_v + G_{thv}$ and $G_{ti} = G_i$ where $G_i = G_v = G$ is due to irradiation = $\xi_t \sigma_{\text{dpa}}$ and $G_{thv}$ is the thermal vacancy source.

$D_{iv}$ are the interstitial/vacancy diffusion coefficients = $D_{iv}^0 \exp(-H_{iv}^m/RT)$ where $D_{iv}^0$ is the pre-exponential and $H_{iv}^m$ is the interstitial/vacancy migration energy (or more literally enthalpy). Note $D_i \gg D_v$.

$R$ is the interstitial-vacancy recombination coefficient, $R = 4\pi r_r(D_i + D_v)/\Omega_a$ where $r_r$ is the interstitial capture radius surrounding a vacancy and $\Omega_a$ is the atomic volume.

Unless otherwise specified in the text, the 'nominal' material properties used in the models described below are given in Table 1. Only a few of the properties ($\Omega_a$, $b$, $\sigma_{\text{dpa}}$) are well established. While the uncertainties are large, variations in $D_i$ have little effect on the conclusions drawn from the models. Others properties ($D_v$, $r_r$, $B$, $S_p$, $S_o$, $\xi_t$, etc.) are also not well known. However, uncertainties in these properties do not generally compromise the qualitative implications of the models.

It is convenient to express the solutions to Equation 14 in terms of the fraction of primary defects available to produce microstructural changes (e.g., that escape recombination thus reach sinks) $f_s$, as

$$f_s(T_i,\phi) = [D_{iv} C_{iv} S_{iv}/V] / G.$$  \hspace{1cm} (15)

The defect currents needed to model clustering and solute diffusion are

$$D_{iv} C_{iv} = f_s(T_i,\phi) G / S_{iv}/V$$  \hspace{1cm} (16)

where $f_s$ varies with flux, $\phi$, and irradiation temperature, $T_i$, depending on the recombination mechanism.

The long term evolution of the microstructure is a complex function of flux, fluence and irradiation temperature. However, it is reasonable to assume that the embrittlement damage is related to, among other factors, the total time integrated defect current, $D_{iv} C_{iv}/V$, where

$$D_{iv} C_{iv}/V = f_s(T_i,\phi) \xi_t \sigma_{\text{dpa}} / S_{iv}/V.$$  \hspace{1cm} (17)

The fluence, $\phi_{\text{dcl}}$, required to produce a specified ($D_{iv} C_{iv}/V$) is

$$\phi_{\text{dcl}}(\phi,T_i) = [S_{iv}/\xi_t \sigma_{\text{dpa}}] / f_s(\phi,T_i)$$  \hspace{1cm} (18)

Hence, $\phi_{\text{dcl}}$ increases with increasing recombination or decreasing $f_s$.

Matrix recombination (subscript sm) involves random encounters between vacancies and interstitials. Neglecting thermal emission from sinks and bias the solution to Equation 14 and 15 is
Figure 5a shows $f_{sm}(\phi,T_i)$ at irradiation temperatures from 260 to 320°C as a function of flux. Figure 5b plots the flux at 50% recombination ($f_{sm} = 0.5$) versus irradiation temperatures for various sink strengths. These results show that except for low sink strengths ($< 10^{10} \text{cm}^{-2}$) and irradiation temperatures (260°C), matrix recombination rates are minimal for surveillance and most MTR conditions ($\phi < 5 \times 10^{13} \text{n/cm}^2\text{s}$). Recombination is dominant only for very high damage rates, with $f_{sm}$ proportional to $1/\sqrt{\phi}$ in the high flux limit.

Recombination at vacancies strongly bound to a high concentration ($C_t$) of traps (subscript tv) could be significant. The approximate defect balance equations with vacancy trapping are

\begin{align*}
G_v + \tau_t' C_{tv} - RC_t C_v - C_v D_v (S_t + 4\tau_1 C_v/\Omega_a) &= 0 \quad (20a) \\
G_i - C_i (RC_v + R'C_{tv}) - C_i D_i S_t &= 0 \quad (20b) \\
C_v D_v 4\pi r_1 C_v/\Omega_a - \tau_t' C_{iv} - R'C_{iv} - M_{tv} C_{tv} D_v S_t &= 0 \quad (20c) \\
\tau_t = b^2/[D_v \exp(-H_{iv}^t/RT_i)] \quad (20d)
\end{align*}

where $r_t$ is the trap capture radius ($= r_v$), $\tau_t$ is the average trapping time, $H_{iv}^t$ is the trap-vacancy binding energy and $M_{tv} D_v$ is the bound trap-vacancy diffusion coefficient. The trap-vacancy-interstitial recombination coefficient, $R'$, is given by Equation 14 except $M_{tv} D_v$ replaces $D_v$. The fraction of defects reaching sinks, $f_{st}$, is ($M_{tv} D_v C_{tv} + D_v C_v)/G$. The mobility factor $M_{tv}$ for trapped vacancies is not well established. However, for fully mobile traps ($M_{tv} = 1$) there is no trap recombination effect ($f_{st} = f_{sm}$). Conversely, $f_{st}$ is independent of trapped vacancy mobility for values of $M_{iv}$ less than about 0.01.

Solute vacancy binding energies are typically in the range of about 5 to 30 kJ/mole [44]. Figure 6a plots $f_{sm}(\phi,T_i)$ and $f_{st}(\phi,T_i)$ versus flux at 290°C for a large binding energy of 29 kJ/mole and a high immobile trap concentration of $10^{13} \text{n/cm}^2\text{s}$. In this extreme case, significant recombination occurs above a flux of about $10^{11} \text{n/cm}^2\text{s}$. The $f_{st}(\phi,T_i)$ has the same form as $f_{sm}(\phi,T_i)$ (for matrix recombination) but is shifted down by a constant flux increment. Figure 6b plots the flux at 50% recombination versus binding energy for a range of sink strengths and trap concentrations. For lower trap and binding energies, trap recombination is significant only at high fluxes ($> 10^{13} \text{n/cm}^2\text{s}$).

The greatest uncertainty in these calculations is due the vacancy migration energy, with reported values ranging from about 70 to 130 kJ/mole [45]. This treatment of trap
recombination has been superimposed on a high nominal value of the vacancy migration energy of 125.5 kJ/mole. Therefore, the low value of $D_v$ probably implicitly accounts for trapping effects. If the lower values of migration energy were used trap recombination would be negligible for all fluxes of interest. Overall these results suggest that vacancy trapping is probably not important for surveillance and even most MTR flux levels. However, trapping can have a profound effect on the effective solute diffusion coefficients (see below).

As noted previously, a significant fraction of primary cascade defects are in the form of small clusters. The cascade interstitial clusters are expected to have a large interstitial bias, hence, in general would be expected to grow into observable interstitial loops. The fact that such loops are not observed remains something of a puzzle as discussed below.

In contrast, small vacancy-clusters would be expected to be thermally unstable, shrinking by rapid vacancy emission. During their lifetime larger (e.g., around 10 vacancies) cascade vacancy-clusters would act as recombination centers. Following the treatment of Straalsund [46], recombination at cascade vacancy-clusters (subscript sc) can be modeled by neglecting dislocation bias and thermal vacancy sources other than the cascade clusters containing $n$ vacancies with an effective surface energy, $\gamma_{\text{eff}}$, and average radius, $r_{\text{vc}}$, as

$$f_{\text{sc}} = \frac{(1+\alpha)\Omega_3 D_v S_1^2}{8\pi r_{\text{sc}} \xi_{\text{dpa}} \phi} \left\{ 1 - \frac{16\pi r_{\text{sc}} \xi_{\text{dpa}} \phi}{\Omega_3 D_v S_1^2 (1+\alpha)^2} - 1 \right\}$$

$$\alpha = \frac{\xi_{\text{inv}} \xi_{\text{dpa}} \phi}{S_3 D^0_{\text{sd}} \exp(-Q_{\text{sd}}/RT) \exp \left( \frac{2\gamma_{\text{inv}} \Omega_3}{r_{\text{nc}} RT} \right)}$$

where $\Omega_3$ is the molar volume ($\Omega_3 N_\text{a}$ where $N_\text{a}$ is Avogadro's number), $D^0_{\text{sd}}$ and $Q_{\text{sd}}$ are the self-diffusion coefficient ($D_{\text{sd}}$) pre-exponential and activation energies and $\xi_{\text{inv}}$ is the fraction of dpa contained in $n$-vacancy cascade clusters. The self-diffusion activation energy is not well-established; the nominal value is roughly the average of the reported range from about 250 to 290 kJ/mole [47,48]. Figure 7a plots $f_{\text{sm}}(\phi,T_1)$ and $f_{\text{sc}}(\phi,T_1)$ for $\gamma_{\text{eff}} = 1.5$ J/m$^2$, $r_{\text{vc}} = 0.3$ cm (equivalent to 10 vacancies) and $\xi_{10v} = 0.0067$ (which corresponds to a 1 barn cluster cross section) at 290°C. Note this value of $\xi_{10v}$ is much less than the total clustered vacancy factor $\xi_{\text{cv}}$ (about 0.2), since the latter also includes smaller much less stable vacancy aggregates. Figure 7b plots the flux at 50% recombination versus temperature for effective surface energies of both 1.5 and 2 J/m$^2$. Cascade vacancy-cluster recombination becomes increasingly significant starting in the low end of the MTR regime ($>10^{12}$ n/cm$^2$-s) for the smaller effective surface energy but only at a much higher flux ($>5x10^{13}$ n/cm$^2$ at 290°C) for the larger effective surface energy. Uncertainties in the self-diffusion coefficient and cascade cluster production cross sections also have a large effect on cluster recombination. At 290°C a variation of the activation
energy by 20 kJ/mole corresponds to a factor of about 75 in $D_{sd}$. The equivalent range of vacancy emission rates for a 0.3 nm cluster is produced by variations in $\gamma_{eff}$ of about 0.43 J/m$^2$. The fraction of vacancies in large clusters $\xi_{10v}$ is also not well established, but recombination is at most only linearly, rather than exponentially, sensitive to variations in this parameter.

In regimes where matrix recombination is not significant ($f_{sm} = 1$), Equation 21 simplifies to

$$f_{sc} = \frac{1}{1 + \alpha} = \frac{1}{1 + C(T)\phi}$$

At very high flux $\alpha$ is much greater than 1, hence, $f_{sc} = 1/\alpha = 1/C(T)\phi$. In the cascade cluster dominated regime, $\phi_{act}$ increases in direct proportion to flux. A cascade vacancy-cluster recombination model in the form of Equation 22 has been previously applied to modeling MTR and surveillance irradiation data [6]. Recently, experiments involving irradiations over a wide range of fluxes up to about $5 \times 10^{13}$ n/cm$^2$·s coupled with post-irradiation annealing studies, have also provided considerable support for the cascade vacancy cluster recombination model [25].

In summary, the models suggest that matrix recombination is not important and solute trap recombination is probably not significant under normal circumstances. However, cascade vacancy-cluster recombination may be significant for MTR irradiation conditions and in this case would contribute to significant flux and temperature effect on embrittlement. Better experimental and theoretical (i.e., by MD-EA methods) evaluations of the key material properties ($Q_{sd}$, $\gamma_{eff}$, $\xi_{inv}$, etc.) are needed to further refine the recombination models.

**VI. DEFECT AND SOLUTE CURRENTS**

Nucleation and growth of large extended defect clusters is due to imbalances in the vacancy and interstitial currents (DC) resulting from sink bias. Consider the case where network dislocations, $S_p$, with a bias, $B$, and unbiased vacancy clusters, $S_c$, are the only sinks. The net vacancies flow to the clusters normalized by the fraction of the defects that escape recombination ($f_c\xi_c\sigma_{dpa}\phi$) is

$$\frac{(D_vC_v - D_iC_i)S_c}{f_c\xi_c\sigma_{dpa}\phi} = \frac{S_pS_pB}{(S_c + S_p)(S_p(1+B)+ S_c)}$$

For small values of $B$, Equation 23 has a maximum value of about 0.25B for $S_p$ equal to $S_c$. If recombination and thermally stable cascade clusters are neglected, this condition places an approximate upper bound on the number of vacancies or interstitials that can aggregate in larger extended defect clusters of 0.25B$\xi_v\sigma_{dpa}\phi$ (or about $1.6 \times 10^{-22}$B$\phi$). For a nominal $B$ of 0.1 this is equivalent to a vacancy or interstitial cluster volume fractions of about $1.6 \times 10^{-4}$ per 10$^{19}$ n/cm$^2$. The upper bound would be reduced by an imbalance in the
sink strengths, the presence of other sinks and vacancy emission. Equivalent expressions can be developed for net interstitial currents at biased sinks and for other combinations of sink structures.

Excess concentrations of both vacancies and interstitials lead to a significant multiplication of solute diffusion rates or radiation enhanced diffusion (RED). The major consequence of RED is to accelerate the formation of phases that would otherwise form thermally but at a slow rate. In the case where the solutes do not interact with the vacancies or interstitials, the RED coefficient, $D^*$, can be approximated as

$$D^* = f_{ci}D_iC_i + f_{cv}D_vC_v = 2f_s(\phi,T_i)\xi_d\frac{\sigma_{dpa}\phi}{S_t} + D_{th} = K(\phi,T_i,\ldots)\phi + D_{th}$$

where $f_{ci}$ and $f_{cv}$ are correlation coefficients approximately equal to unity and $D_{th}$ is the thermal diffusion coefficient. For the typical case that $D^*$ is much greater than $D_{th}$, the flux and temperature dependence of $\phi_{dct}$ is contained in the $K(\phi,T_i)$ factor. At low damage rates $K$ and, hence, $\phi_{dct}$ are independent of flux; at very high damage rates in the cascade cluster dominated regime, $K$ is inversely proportional and $\phi_{dct}$ is directly proportional to flux. In the very low flux limit where $D^*$ is on-the-order of $D_{th}$, $\phi_{dct}$ is again proportional to flux.

More complicated treatments (e.g. based on Equation 20) are needed in the case of bound defect-solute complexes. However, for relatively weak binding energies $H_{bs}^{SV}$ less than about 20 kJ/mole, the vacancy-solute binding enhancement of $D^*$ can be approximated as

$$D^* = [2f_s(\phi,T)\xi_d\frac{\sigma_{dpa}\phi}{S_d}]E_{sv}$$

(25a)

where

$$E_{sv} = [1+0.5\exp(H_{bs}^{SV}/RT)]$$

(25b)

The maximum value of the $\exp(H_{bs}^{SV}/RT)$ term is $1/C_s$, where $C_s$ is the solute concentration. Based on reported copper-vacancy binding energies of 13.5 kJ/mole [44], $E_{sv}$ is about 10 at 290°C. Neglecting recombination, this yields a nominal $K$ equal to $2.4\times10^{-31}cm^4$ for the properties given in Table 1.

Diffusion of tightly bound interstitial/vacancy-solute trap complexes to sinks also results in radiation induced segregation (RIS) [49]. For RPV conditions diffusion of interstitial-subsize solute atom complexes would probably be the dominant source of RIS. The majority (or in some cases all) of the interstitial-solute complexes arriving at sinks recombine with the nearly equal (or equal) current of vacancies, thereby depositing and enriching solute to a level limited by back diffusion via vacancy exchange. The currents of these species $D_{Si}C_{Si}$ can be approximated from the interstitial equivalent of Equation 20 yielding
where \( H_b^s \) is the solute-interstitial binding energy and \( S_s \) is the solute trap sink strength of about \( 4 \pi r_s C_s / \Omega_a \). For large \( H_b^s \), the lower bound on the fraction of the interstitials arriving at sinks as complexes is \( 1/(S_t/S_s + 1) \); hence, for the typical case of \( S_s \) much greater than \( S_t \), solutes are deposited at sinks approximately at the rate of interstitial generation. Additional inflow and outflow of solutes by vacancies-exchange takes place in the same ratio (but not rate) as for thermal equilibrium conditions. For dilute alloys \( (C_{ss} \ll 1, \text{ where } C_{ss} \text{ is thermal solubility limit of the solute}) \) the solubility limit under irradiation \( C_{ssi} \) is

\[
C_{ssi} = \frac{C_{ss} \Omega_a S_t}{4 \pi r_t} \quad (27)
\]

For any reasonable choices of \( S_s \) and \( r_t \), \( C_{ssi} \) is much less than \( C_{ss} \). Thus a major consequence of RIS may be radiation-induced precipitation of non-equilibrium phases, viz., precipitates that would not form thermally. Extended defect cluster-solute complexes also result from this form of radiation-induced segregation. Such complexes may be the precursors to radiation induced precipitates. Radiation-induced segregation can also take place at fixed sinks such as grain boundaries.

**VII. MICROSTRUCTURAL EVOLUTION**

The previous sections form a sound physical basis to model the effects of vacancy and interstitial clustering and accelerated or induced solute diffusion on extended defect formation and precipitation, or microstructural evolution, as a function of the irradiation and metallurgical variables. To date embrittlement models have focused on only two types of features: a) copper rich precipitates (CRP); and b) a general class of unidentified features commonly referred to as 'matrix defects'. Radiation enhanced CRP are well established and form as a consequence of RED [1,3-9,31,50]. The matrix defects may include extended defect clusters (EDC), EDC-solute complexes and other precipitate phases that are enhanced, induced or modified by irradiation.

The rate theory equations described in the previous section can be extended to provide a complete kinetic model of defect and solute cluster evolution by a set of coupled differential equations that account for all generation, growth and shrinkage reactions that produce or remove the cluster of a particular size class. While obtaining numerical solutions to a large number of these often exceedingly stiff equations is not trivial, such detailed formulations are generally computationally feasible for RPV steels, since the clusters are 'relatively' small. Further, in principle, it is possible to generalize the approach to model the formation of defect-solute complexes and multielement precipitates, although the complexity rapidly escalates with the number of species. The general computational scheme is illustrated in...
Figure 8. The rate constants can incorporate an essentially arbitrary level of physical detail describing various thermal and irradiation mediated processes.

a. Extended Defect Clusters (EDC)

Embrittlement models have explicitly or implicitly associated the matrix defects with one or another type of EDC [1,3-9,43,51-54]. For example, early work by Odette modeled the in-situ production and self-annealing kinetics of small cascade vacancy-clusters [53]. This was later modified to include a second matrix defect, postulated to be a growing nanovoid [6-8], in terms of an empirical expression for a contribution to the transition temperature shift, $\Delta T_{md2}$, as

$$\Delta T_{md2} = \theta_{md2}(T_i)(F_{md2}+N_{md2}Ni)\sqrt{\phi t - \theta_{md2}} \quad (28)$$

where the $\theta_{md2}(T)$, $F_{md2}$, $N_{md2}$ and $\phi t$ are fit functions or constants reflecting the increasing importance of this feature with decreasing irradiation temperature and increasing nickel content and fluence. Fisher and co-workers [4, 5] proposed a generally similar model for the matrix defect, postulated to be a dislocation loop, in the form

$$\Delta T_{mdl} = \theta_{mdl}(T_i)\sqrt{\phi t} \quad (29)$$

where the magnitude of $\theta_{mdl}$ depends on the type of steel. Notably, while it might be expected on theoretical grounds, no explicit flux dependence is included in either treatment. More detailed models of EDC are needed.

Since interstitial-emission rates are negligible [43] and thermal vacancy absorption can be neglected, rapid growth of interstitial loops above a critical nucleus size of only a few ($\geq3$) defects would be expected. The corresponding excess vacancies could accumulate at growing vacancy clusters or be annihilated at network dislocations with lower bias. In the simplest approximation, homogeneous loop nucleation rates would scale roughly as $\sqrt{(\phi/D_i)}$ [55]. Hence, due to the large values of the interstitial diffusion coefficient, loop populations might be negligible. However, direct formation of supercritically sized loops as cascade interstitial-clusters (or perhaps heterogeneous nucleation at interstitial-trapping sites) would be much more rapid. For loop number densities between $10^{17}$ and $10^{18}$/cm$^3$ a modest excess loop bias (e.g., $\geq1$) would be expected to yield loops of 2.0 nm or larger at fluences of around $10^{19}$n/cm$^2$-s. However, loops are typically not observed at fluences of less about $5 \times 10^{19}$ n/cm$^2$. Further, even modestly sized interstitial loops would be stable during post irradiation annealing, at least without a corresponding source of excess vacancies. Such stability is inconsistent with the rapid recovery of a matrix defect contribution observed during low temperature post-irradiation annealing (see below). The reasons for the apparent absence of loops are not understood.

One possibility is that there is a very high density of small (subvisible) loops. However, the corresponding ultrahigh sink density ($\gg 10^{11}$/cm$^2$) would be inconsistent with both observed rates of RED and typical levels of hardening. A more plausible reason could be that interstitial solutes (e.g., carbon and nitrogen) atmospheres shield loop strain fields,
reducing their bias and sink strength. Computer simulations suggest that while small loops in iron initially nucleate and grow in the a/2<110> faulted configuration, they convert to perfect prismatic a/2<111> or a<100> loops at cluster sizes above about 10 to 20 interstitials [56]. Thus another hypothesis for the failure to observe these features is that a low probability of a pre-existing jog, or the necessity to nucleate a jog, would make such small prismatic loops inefficient sinks. Other possible 'explanations' include: a) the thermal stability (binding energies) of small interstitial clusters, b) vacancy and interstitial cascade clusters efficiently co-annihilate during STA.

Interstitial clustering models have been applied to loop nucleation and growth [43, 54] and can be extended to include an essentially arbitrary level of physical and mathematical detail. However, meaningful modeling will require a better understanding of the basic properties of small interstitial-cluster-solute complexes. Again, MD-EA modeling could play a key role in determining these properties.

In addition to the small thermally unstable vacancy-clusters that are produced directly in displacement cascades, excess vacancy supersaturations can nucleate growing vacancy-clusters or nanovoids. A unified kinetic model of vacancy-cluster evolution can be based on solving an explicit set of clustering, or master, equations with the general form

\[
\frac{dC_j}{dt} = \eta_{i,j-1}C_{j-1} + (\eta_{i,j+1} + \psi_{j+1})C_{j+1} - (\eta_{i,j} + \psi_j)C_j + G_{vj}, \quad j = 1, j_{\text{max}} \quad (30a)
\]

where \( C_j \) is the concentration of clusters containing \( j \)-vacancies, and \( j_{\text{max}} \) is the maximum cluster size, typically several hundred to a few thousand vacancies (hence, equations). The flows in cluster size space represented by Equation 30a correspond to the positive abcissa in Figure 8. The rate coefficients (\( \eta \) and \( \psi \)) are the absorption rates (\( \eta \)) for vacancies (subscript \( v \)) and interstitials (subscript \( i \))

\[
\eta_{v,j} = 4\pi r_j D_v C_v / \Omega_a \quad \text{and} \quad \eta_{i,j} = 4\pi r_j D_i C_j / \Omega_a \quad (30b)
\]

and vacancy-emission rates (\( \psi \)),

\[
\psi_j = 4\pi r_j D_{sc} \exp\left(\frac{2\gamma_{vf}(\Omega_m/\Omega_a)}{\eta RT}\right) / \Omega_a \quad (30c)
\]

The \( G_{vj} \) is the direct generation rate of cascade vacancy-clusters of size \( j \). Alternate forms of Equation 30a are required for the \( j = 1, j_{\text{max}}-1 \) and \( j_{\text{max}} \) size classes. For the \( j = 1 \), Equation 14, including appropriate cluster vacancy absorption and emission terms, is solved. For \( j_{\text{max}}-1 \), the shrinkage rates from \( j_{\text{max}} \) are set equal to 0; however, the net flow of clusters through \( j_{\text{max}}-1 \) is accumulated in \( j_{\text{max}} \). The set of \( j_{\text{max}} \) equations are integrated using a stiff equation solver (i.e., GEAR [57]). Considerable insight can also be gained from analytical steady-state solutions to Equation 30 obtained by setting \( dC_j/dt = 0 \) and ignoring the clusters as sinks and vacancy-sources.

Primary defect generation processes dominate nanovoid evolution kinetics. Figure 9a shows steady-state vacancy-cluster distributions at 290°C and a flux of \( 5 \times 10^{12} \) n/cm²-s for
\( \gamma_{\text{eff}} \) of 1.2 J/m\(^2\). The dashed curve assumes there is no cascade cluster generation. The solid curve assumes a 1 barn (10\(^{-24}\) cm\(^2\)) cross section for forming cascade clusters containing 10 vacancies. The modest cascade vacancies-cluster generation rate (less than 1% of the primary displacements) increases the nanovoid concentrations at steady-state (and corresponding nucleation rates) by about 4 orders of magnitude. Figure 9b shows the evolution of the cluster size distribution for this case up to a fluence of 10\(^{19}\) n/cm\(^2\). Figures 10a to 10c show the corresponding average nanovoid size, number density and volume fractions for this case as well as irradiations at higher temperature (320°C) and lower flux (1x10\(^{12}\) n/cm\(^2\)-s). The higher temperatures and lower fluxes result in increased nanovoid sizes, slightly smaller volume fractions and much lower number densities.

Predictions of vacancy cluster evolution kinetics are very sensitive to the material properties, particularly \( \gamma_{\text{eff}} \), as illustrated in Figure 11a. Here, the steady-state nanovoid nucleation rate, \( J_{\text{ss}} \), as a function of flux is shown for a range of \( \gamma_{\text{eff}} \) at 290°C. Note this range of \( \gamma_{\text{eff}} \) also acts as a good surrogate for uncertainties in other material properties (e.g. \( D_{\text{sd}} \)) as well. The conditions for 'significant' vacancy clustering, defined (somewhat arbitrarily) as the nucleation rate required to create 10\(^{17}\)/cm\(^3\) nanovoids in 10\(^{19}\) n/cm\(^2\), is shown as a dashed line. Figure 11b plots the flux required for this condition as a function of \( \gamma_{\text{eff}} \) for various temperatures. These results indicate that if \( \gamma_{\text{eff}} \) is sufficiently low (< 1.3 J/m\(^2\)), nanovoids may be significant component of embrittlement microstructures over a large range of MTR irradiation conditions. Nanovoids form at larger \( \gamma_{\text{eff}} \) (< 1.7 J/m\(^2\)) at lower irradiation temperatures or very high fluxes (>>10\(^{13}\) n/cm\(^2\)-s). However, they generally diminish in importance at very low surveillance fluxes or higher irradiation temperatures. Unlike the case of interstitial-loops, there is evidence that vacancies-cluster-complex features develop in irradiated RPV steels and particularly in simple ferritic model steels.

Independent evaluation of \( \gamma_{\text{eff}} \) is difficult. Characteristic values for clean, bulk surfaces are in excess of 2 J/m\(^2\) [43,58] in this case negligible nanovoid evolution would be expected at 290°C even for high flux MTR irradiations. However, these large values may not be appropriate for very small cluster sizes [59] and in impure metals subject to surface segregation [58]. Molecular dynamics, density functional and standard segregation thermodynamics methods could be used to better determine appropriate values of \( \gamma_{\text{eff}} \). A more practical approach to constraining the range of \( \gamma_{\text{eff}} \) could based on modeling the thermal stability of vacancy-cluster microstructures under PIA. This is discussed below.

When nanovoid nucleation and growth rates are negligible, vacancy-feature kinetics are dominated by in-situ annealing rate of cascade vacancy-clusters. Considering only a single vacancies-cluster size class \( ij \) this can be simply modeled as

\[
C_{\text{Vij}} = G_{\text{Vij}} \tau_{\text{Vij}} [1 - \exp(-t/\tau_{\text{Vij}})]
\]

where the effective cascade vacancies-cluster relaxation time, \( \tau_{\text{Vij}} \), is given by
Steady-state is reached at $t \gg \tau_{vj}$ with $C_{vj}^{ss} = G_{vj}\tau_{vj}$.

Evolution of extended defect clusters during PIA annealing can also be modeled using Equation 30 with appropriate modification of the $\eta$ and $\psi$ coefficients. A simpler, albeit less rigorous, approach is to evaluate the average time for the complete dissolution of a individual cluster of size $j$. Assuming equilibrium matrix concentrations of vacancies, the interstitial-loops would be expected to be fairly stable, annealing at self-diffusion controlled rates, given by

$$\tau_{vj} = \sum_{k=1}^{j} \left[ \frac{1}{\eta_{vj} + \psi_{j} - \eta_{vj}} \right]$$  \hspace{1cm} (33)

where $\eta_{vj}$ is given by Equation 30b with $D_vC_v = D_{sd}$. Excess vacancies released by coincident vacancy-cluster annealing would decrease $\tau_{aij}$. Nanovoids are much less stable than loops due to the large effect of the surface energy (e.g., $\exp(2\gamma_{eff}\Omega_m/\gamma_{vj}RT) \gg 1$). The characteristic nanovoid annealing time $\tau_{avj}$ is given by Equation 33. A lower bound on $\tau_{avj}$ is given by neglecting the vacancy absorption term

$$\tau_{avj} = \sum_{k=1}^{j} \left[ \frac{1}{\psi_{j}} \right]$$  \hspace{1cm} (35)

The variation of $\tau_{aij}$ and $\tau_{avj}$ with annealing temperature, $T_a$, for a range of $\gamma_{eff}$ and $j$ are shown in Figure 12. The predicted annealing times for interstitial-loops are much longer than for the vacancies-aggregates. However, the larger nanovoids ($j > 50$) also have very long dissolution times at lower annealing temperatures even for high $\gamma_{eff}$.

Observed matrix defect annealing kinetics are inconsistent with the dissolution times predicted for the loops [25]. Likewise, the annealing times characteristic of even moderately sized nanovoids ($j = 50$) are also inconsistent with observation for low values of $\gamma_{eff}$ that permit the formation of a significant population of these features in the first place. While there are indications that the thermal stability of the matrix features increase at higher fluence [24], the general implication of these results is that large nanovoids and interstitial-loops are not significant at least up to about $10^{19}$ n/cm$^2$; however, these features may evolve at higher fluences. Smaller vacancy-cluster complexes (and possibly small interstitial-loops which could co-anneal) are more likely candidate EDC matrix defects. If loops with large bias eventually do begin to develop, they would provide an additional driving force for a co-evolution of nanovoids, leading to accelerated hardening and embrittlement.

b. Copper Rich Precipitates (CRP)

Early theoretical predictions [4, 50] that small coherent (bcc) copper (rich) precipitates (CRP) are the dominant microstructural feature leading to embrittlement in steels containing significant impurity concentrations of this element have been verified by many experimental studies (see reference 31 for a list of citations). Further, simple precipitation models have
been highly successful in qualitatively rationalizing, and in many cases quantitatively predicting, the effects of embrittlement and annealing variables. A common feature of these models is recognition that typical copper impurity levels greatly exceed the equilibrium solubility limit around 300°C [24, 60-62]. Figure 13 plots the copper solvus, $C_{\text{cusp}}(T)$, for the equilibrium fcc phase as well as for bcc copper with a coherent interface energy of 0.4 J/m$^2$ [24, 63] and precipitate radii from 0.5 nm to infinity (e.g., the bulk bcc phase). The bcc phase increases the copper solubility by about a factor of 2 at around 290°C. The Gibbs-Thompson factor (see below) is somewhat larger for small precipitates (i.e. 11.2 for a precipitate radius of 0.5 nm). Typical copper impurity concentrations are in the range of 5x10$^{-4}$ to 5x10$^{-3}$. Hence, clearly copper precipitation can occur under irradiation. Note, Figure 13 also shows the typical range of heat treatment temperatures, which may limit the copper concentration in solution (due to pre-precipitation) prior to irradiation.

Subject to the considerable uncertainties of extrapolation from higher copper concentrations and aging temperatures, estimates of thermal precipitation times at 290°C range from about 2x10$^5$ to 10$^7$ h [4, 8, 24]. However, under irradiation precipitation is essentially complete within 2000 h or less for typical MTR fluxes of about 4x10$^{12}$n/cm$^2$-s. Indeed, radiation enhanced precipitation has been reported at temperatures as low as 170°C, clearly demonstrating an enormous RED effect [4].

Fisher and co-workers developed a RED model for copper-precipitation based on the following elements [4, 5]. The time to peak strength (yield stress or hardness) under irradiation, $t_p'$, scales with the corresponding time under thermal aging (without irradiation), $t_p$, by the ratio of the thermal equilibrium vacancy-concentration to the vacancy-concentration under irradiation [$C_{v}^{e}/C_v$] as

$$ t_p' = t_p C_{v}^{e}/C_v \tag{36a} $$

The thermal aging time to peak strength is empirically determined by extrapolating a fit to the lower legs of time-temperature copper precipitation/strengthening c-curves constructed from data in the literature. The vacancy concentration under irradiation is evaluated based on Equation 24, neglecting recombination ($f_s = 1$). The peak strengthening is assumed to depend only on the effective copper-content and determined following the analysis of Russell and Brown. The effective copper content is based on a reduction of the nominal concentration by the amount that is located in CuS inclusions, hence, is unavailable for precipitation. The fluence dependence of embrittlement is treated by a simple empirical representation of a generic aging time-strength curve that depends only on the magnitude of and time to peak strength. Thus the RED kinetics are fully represented by

$$ t_p' = t_p \left[ \frac{\Omega_s S_i \exp(-Q_{sd}/RT_i)}{G} \right] \tag{36b} $$

While this formulation has a direct tie to thermal aging data, the RED scaling depends on the validity both of the empirical extrapolation for $t_p$ and the choice of $Q_{sd}$. Good agreement with observed data trends was found using a low value for $Q_{sd}$ of 235.4 kJ/mole which is at "the lower end of the measured range". Further, Equation 36 does not provide any explicit dependence of RED precipitation kinetics (e.g., $\phi_{\text{dec}}$) on flux.
Odette independently developed a RED model [50] based on

\[ D^* = K_\phi = 2\xi C_{dpa}\phi /S_d[D_{uv}/D_{sd}] = 10^{-30}\phi \]  

(37)

where the estimate of \( K \) (10^{-30} \text{ cm}^4) is for a slightly different set of material properties than given in Table 1. The \([D_{uv}/D_{sd}]\) factor approximately accounts for vacancy-copper binding energy; the estimated value of 10 is consistent with Equation 25b. While lacking a direct tie to thermal aging data, this treatment avoids the sensitivity to \( t_p \) and \( Q_{sd} \) found in the Fisher model. This treatment was subsequently modified to account for the effects of cascade vacancy cluster recombination with an empirical representation of Equations 22 and 25 in the form [6]

\[ D^* = \frac{10^{-30}\phi}{1+10^{-13}\phi} \text{ cm}^2/\text{s} \]  

(38)

Note the estimates of \( K \) in Equations 37 and 38 are about 4 times the nominal theoretical value.

Odette's precipitation model is based on the presumption that nucleation occurs early in irradiation and that the kinetics are dominated by diffusion-controlled growth. The model also assumes that: a) copper re-emission from the precipitates can be neglected; and b) CRP contain other elements (initially about 50% iron [50] later modified to consider other constituents [6-8]) arriving at a rate directly proportion to copper. Initially the effective copper content, \( C_{\text{cue}} \), was assumed to be the nominal copper concentration [50]. However, in subsequent treatments, the reduction in the matrix copper due to pre-precipitation during heat treatment as well as CuS inclusions was recognized [6-8]. The revised model also: a) postulates that nickel has a retarding influence on pre-precipitation; and b) adds an effective phosphorous content contribution to \( C_{\text{cue}} \) reflecting the possible irradiation induced or enhanced precipitation of this element.

For a specified precipitate number density, \( N_{crp} \), the fractional of precipitated copper, \( \Theta = f_{crp}/C_{\text{cue}} \), can be found by integrating the diffusional growth equation, yielding [50]

\[ \phi(\Theta) = \frac{0.38x_{\text{cup}}^{1/3}}{KN_{crp}^{2/3}C_{\text{cue}}^{1/3}} \left\{ 0.167^{-3}\left( \frac{\Theta^{2/3}+\Theta^{1/3}+1}{(\Theta^{1/3}-1)^2} \right)+0.577\tan^{-1}\left( \frac{2\Theta^{1/3}+1}{\sqrt{3}} \right)+0.304 \right\} \]  

(39)

where \( x_{\text{cup}} \) is the CRP copper content. Equation 39 cannot be readily inverted to solve for \( \Theta \); however to a good approximation \( \Theta \) is given by

\[ \Theta = 1 - \exp(-10.55K^{3/2}t^{3/2}N_{crp}C_{\text{cue}}^{1/2}x_{\text{cup}}^{-1/2}) \]  

(40)
as illustrated in Figure 14. Note that modified or alternative versions of RED precipitation models have been developed by other workers [9, 64].

The RED factor K (hence, D*) can be evaluated by fitting Θ(φt) data using Equation 40 along with measured values of C_{Cu}, N_{CrP} and \( x_{Cu} \). Data from small angle neutron scattering (SANS) measurements on several commercial Mn-Mo bainitic steels irradiated at 288°C at a flux of about \( 4 \times 10^{12} \) n/cm\(^2\)-s to a fluence of about \( 1 \times 10^{19} \) n/cm\(^2\), yields a K averaging about \( 5.5 \pm 2 \times 10^{-31} \) cm\(^4\) [24]. These estimates are approximate due to uncertainties in the measurements (\( \Theta, C_{Cu}, N_{CrP} \) and \( x_{Cu} \)) as well as assumptions in the model. However, the measured radiation enhanced diffusion factor values compare well with the nominal value predicted by Equation 25 of \( 2.4 \times 10^{-31} \) cm\(^4\). The differences can be readily explained by lower effective sink densities. At a flux of \( 4 \times 10^{12} \) n/cm\(^2\) the corresponding D* is \( 2.2 \times 10^{-18} \) cm\(^2\)/s compared to an extrapolated thermal diffusion coefficient \( D_{th} \) at 300°C of about \( 2.0 \times 10^{-23} \) cm\(^2\)/s [65]. While the \( D_{th} \) estimates are also highly uncertain at such low temperatures, with plausible values ranging from about \( 10^{-21} \) to \( 2 \times 10^{-25} \) m\(^2\)/s, an enormous RED factor is indicated. Similar analysis for different materials and irradiation conditions shows that K[24]: a) is higher for the simple ferritic (Fe-Cu-C-Ni) model steel, consistent with the lower dislocation sink density in this alloy; b) decreases with increasing fluence, consistent with a buildup in the sink density; and c) increases with decreasing flux and increasing irradiation temperature, consistent with predictions of the recombination models described previously.

Figure 15 shows transition temperature shifts predicted by combining Equations 1 (the Russell-Brown model), 4 (linear superposition), 14 (using \( \Sigma_{cv} \) of 0.65°C/MPa) and 40 (with \( x_{Cu} \) of 0.6). The shifts are plotted as a function of Kφt for a range of effective copper contents and CRP densities. Note the specified N_{CrP}, C_{Cu}, x_{Cu} scale the non-dimensional fluence \( 2.63 N_{CrP}^{2/3} C_{Cu}^{1/3} x_{Cu}^{-1/3} K \) in Figure 15. The dashed curves show empirical estimates of CRP contribution to embrittlement for welds found by subtracting a matrix damage term (Equation 28 with \( \Delta T_{md2} = (8+30Ni) \sqrt{\phi t} \) (°C), where Ni is in percent and \( \phi t \) in units of \( 10^{19} \) n/cm\(^2\)) from the surveillance data base correlation for welds [3]. The empirical curves have been scaled assuming a K of \( 10^{-30} \) cm\(^4\). This simple formulation has been highly successful in describing many of the key features of CRP evolution and the corresponding mechanical property changes. However, the diffusion controlled growth model is clearly overly simplified and cannot be used to predict precipitate densities or accurately model the effect of low copper contents.

A more rigorous treatment can be based on a full nucleation, growth and coarsening clustering model (Equation 30). In the simplest case the \( \eta \) and \( \psi \) absorption and emission terms are modified to account for RED and thermal emission of copper atoms. Assuming diffusion controlled kinetics, the clustering model requires only D*, the equilibrium solubility of copper, C_{CuS}, and the energy of the coherent precipitate-iron interface, \( \gamma_{coh} \). The C_{CuS} and \( \gamma_{coh} \) parameters mediate the balance of mechanisms (nucleation, growth and coarsening) controlling the precipitate evolution kinetics while K fixes absolute fluence scale. Elaborations of the clustering model include: interface or mixed rate control; alloyed precipitates (which effects \( N_{CrP}, C_{CuS} \) and \( \gamma_{coh} \)); coherency strain free energy contributions; heterogeneous nucleation and dynamic effects of radiation such as resolutioning and
copper vacancy cluster-complexes (the latter may be another form of heterogeneous nucleation).

Figure 16a to d show predictions of the full clustering model for a copper content of 0.3% at 290°C and a flux of $4 \times 10^{12}$ n/cm$^2$-s assuming a $\gamma_{coh}$ of 0.4 J/m$^2$ and a K of $10^{-30}$ cm$^4$. The cluster size distribution develops a minimum (pinches off) at about $10^{18}$ n/cm$^2$ signaling the end of the nucleation regime (Figure 16a); at higher fluences the maximum size increases due to growth and coarsening (Figure 16a). The corresponding precipitate densities increase up to a broad maximum of about $2 \times 10^{18}$/cm$^3$ between about 0.075 to 2 $\times 10^{19}$n/cm$^2$-s, declining slowly at higher fluences (Figure 16b). The CRP volume fraction, $f_{crp}$, begins to increase rapidly at about $10^{17}$ n/cm$^2$ approaching the limit at about $5 \times 10^{18}$ n/cm$^2$ (Figure 16c). The precipitate radius also increases rapidly in this regime, transforming from growth to non-equilibrium coarsening dominated kinetics following nearly full precipitation.

The yield stress increases (using Equations 1 and 4) rapidly between about 1 and $5 \times 10^{18}$/n/cm$^2$, as shown in Figure 17. At higher fluences the strengthening rate decreases approaching an asymptotic fluence exponent ($\Delta \sigma_{Y} \propto \phi^p$) with p of about 0.2. These predictions are qualitatively consistent with observation [1]. However, data trends suggest that the entire curve should be shifted down in fluence by a factor of about 2 (the dashed line). This could be attributed to underestimate of K which may be higher than $10^{-30}$ cm$^4$ for low flux (surveillance) and low fluence conditions. Further, the precipitate number densities are somewhat larger (by about a factor of 4) than typically observed [24]. Overall, however, the results are encouraging and generally resemble the predictions of the simple diffusional growth model.

Evaluation of the effects of copper content and irradiation temperature with the full clustering model are also in qualitative agreement with expected increase (or decrease) in the precipitate number density with higher (or lower) copper content and lower (or higher) temperature. As the precipitate densities increase, the kinetics in general, and the strengthening in particular, become more and more dominated by the coarsening mechanism. In contrast, at low copper concentrations or high temperatures the kinetics become increasingly dominated by nucleation. Overall, the clustering model shows that precipitation and strengthening kinetics manifest a complex interplay between nucleation, growth and coarsening.

To date no effort has been made to fine tune the clustering model parameters or to incorporate many of the refinements noted above. A key element in developing more rigorous CRP models is an accurate treatment of the precipitate composition, which includes significant quantities of manganese and nickel (and to a lesser extent other elements such as silicon and phosphorous). As noted in the previous section such alloying may have a significant effect on a number of fundamental CRP properties.

Manganese and nickel enrichment in the CRP can be understood and modeled based on quasi-equilibrium thermodynamics treatment [24]. Rigorous formulation requires a rather complex set of relations and a large number of thermophysical parameters; hence, a
detailed description of these models is beyond the scope of this paper. However, a simplified description illustrates the key physical factors controlling precipitate compositions. Perhaps the most significant factor is associated with the very small precipitate sizes ($r_p < 2 \text{ nm}$). In this circumstance, the precipitate-matrix interface adds a significant contribution to the total molar free energy, $F_t$, as

$$F_t = \sum_i x_i (\mu_i + \Delta F_{si}) + \Delta F_{mix} + 2\gamma_{coh}Q_{crp}$$

(41)

where $x_i$ is the mole fraction of constituent $i$, $\mu_i$ is the free energy of the pure $i$ in the reference state, $\Delta F_{si}$ is the structural free energy change between the reference and solution states, $\Delta F_{mix}$ is the free energy of mixing, $r_{crp}$ is the precipitate radius and $\gamma_{coh}$ is the coherent (chemical) interface energy. For a given amount of copper precipitation both $r_{crp}$ and $\gamma_{coh}$ are functions of composition. The chemical interface energy can be approximated as [66]

$$\gamma_{coh} (\text{Cu}, \text{Mn}, \text{Ni}) = \sum_i \Delta x_{ip}^2 H_{si} n_s z_s N_s z$$

(42)

where $\Delta x_{ip}$ is the difference between the precipitate and matrix concentration and $H_{si}$ is the enthalpy of solution of the $i$th constituent, $n_s$ is the number of bonds across the interface (= 2), $n_s$ is the number of atoms per unit area of interface and $z$ is the lattice coordination (63). Since the enthalpy of solution for copper in iron is much greater than for either manganese or nickel, $\gamma_{coh} = \gamma_{coh}(\text{Cu})x_{crp}^2$. Hence, these solutes may decrease the total free energy of small precipitates, even if they increase the corresponding bulk free energy of the copper rich phase. The precise precipitate composition is controlled by both thermodynamic and kinetic factors. However, it is reasonable to assume that the precipitates grow at a rate of controlled by the arrival of their primary constituent, copper, and that manganese and nickel contents are at near quasi-equilibrium levels. Quasi-equilibrium is established when $\mu_i^{crp} = \mu_i^{fe}$ and $\mu_{crp} = \mu_{mn}^{fe}$, where the $\mu$'s are the constituent chemical potentials, defined from the standard relation

$$\mu_i = \langle \partial F_i / \partial n_i \rangle_{T,p,n_j=n_j}$$

(43)

where $\partial F_i$ is the change in total extensive free energy with a change in $\partial n_i$ are the moles of $i$. Based on thermochemical data from the literature [67-70], an iterative minimization procedure is used to find the compositions. A subregular solution model is used to calculate $\Delta F_{mix}$. The model predicts CRP compositions as a function of temperature, precipitate radius and alloy composition. Salient predictions of the CRP composition model include:

- Manganese is enriched in the bulk fcc-copper phase in Cu-Mn-Fe alloys. The non-equilibrium enrichment is enhanced by both the lower structural free energy of manganese in the bcc phase and by its effect on $\gamma_{coh}$.
- Nickel is not enriched in either the bulk fcc or bcc copper phases in Cu-Ni-Fe alloys. However, due to reductions in $\gamma_{coh}$, modest non-equilibrium enrichment of nickel to bcc-copper is predicted at very small precipitate sizes.
Co-enrichment of manganese and nickel is predicted for fcc-copper rich phases in Cu-Ni-Mn-Fe alloys, due to the interaction between manganese and nickel, increased entropy of mixing and their combined effects on $\gamma_{coh}$. Unfortunately, thermodynamic parameters are not available for bcc-Cu-Ni-Mn phases, but similar or enhanced co-enrichment is expected.

Nickel and manganese concentrations increase with decreasing precipitate size, temperature and nickel and manganese content.

At high nickel and manganese contents and low temperatures nickel-manganese rich phases may form.

Segregation of manganese and nickel to the CRP interface would be expected and could further increase enrichment in very small precipitates.

Figure 18 shows the estimated manganese plus nickel content of CRP as a function of the precipitate radius for available matrix solute compositions of $C_{mn} = 0.01$ and $C_{ni} = 0.005$. Partitioning is shown for temperatures of 250°C near the low end of the pressure vessel range, at 300°C typical of most operating vessels and 450°C characteristic of high temperature post-irradiation anneals. At 300°C small ($= 0.75 \text{ nm}$) CRP contain about 77% copper, 20% manganese and 3% nickel. The solute concentrations increase rapidly at smaller sizes and fall of more slowly to near 'bulk' values at 4 nm. Enrichments are much higher at the low temperatures. Similar, albeit less pronounced, enrichment in other solutes such as phosphorous and silicon would also be anticipated.

These predictions are generally consistent with experimental observations. There is also experimental confirmation of the model prediction that post irradiation annealing (PIA) reduces the manganese and nickel contents markedly due to the higher temperatures and larger (coarsened) precipitate sizes [24,31].

Solute enrichment directly increases the volume fraction of CRP relative the available copper content. However, indirect effects could also be significant. Even relatively small concentrations of solutes that reduce $\gamma_{coh}$ would result in significant reductions the local copper solubility, hence, corresponding increases the nucleation rates and precipitate number densities. The potential of form nickel-manganese rich phases (containing lesser amounts of copper and iron) may also be significant. While quasi-equilibrium thermodynamics suggests that the copper rich phases are compact clusters, the nickel manganese rich features may constitute the ghostly 'clouds' or 'atmospheres' reported in some atom probe studies. Finally, recent Monte Carlo simulations have suggested that of nickel and manganese may both: a) form ordered regions within CRP; and b) segregate to the iron-precipitate interface. These simulations also point to the possible existence of very small copper-nickel-manganese clusters (e.g. less than about 10 to 20 atoms) with complex, non-compact, shapes. While such features may not produce mechanical property changes per-se, they may influence the interpretation of small angle neutron scattering data.
The PIA kinetics of CRP depend on their as-irradiated characteristics and the matrix composition as well as annealing temperature and time. Clustering models (Equation 30) can also be applied to CRP evolution during PIA. The PIA models show that CRP undergo both dissolution and more rapid coarsening and property changes during PIA than might be predicted by simple treatments due to:

- The 'large' non-linear Gibbs-Thompson term \( \exp(2\gamma_{coh}Q_m/\kappa_{crp}RT) \) for small CRP
- The decrease in the CRP manganese and nickel contents noted previously. This composition change also results in a higher value of \( \gamma_{coh} \), further accelerating coarsening.
- A large effect of the decrease in the density of CRP on strengthening due to superposition with pre-existing dispersed obstacle strengthening contributions.
- Possible release of vacancies from vacancies-clusters during annealing enhancing copper diffusion rates.

In spite of the limited dissolution and enhanced coarsening kinetics, the CRP features are relatively stable under PIA. Notably, the copper contained in coarsened precipitates is not available for precipitation during subsequent re-irradiations. Outstanding questions about CRP annealing include the co-evolution with vacancy clusters, noted above, and the possibility of alternate evolution mechanisms, such as accelerated coarsening of precipitates attached to dislocations. Finally, the CRP undergo a complex transition from coherent bcc to incoherent fcc structures via a complex semi-coherent R9 transition phase [71]; the properties of this phase are not well known.

In summary, more rigorous clustering-thermodynamic models suggest that CRP evolution involves a complex interplay between nucleation, growth and coarsening that is sensitive to precipitate, hence, alloy composition. Better experimental and theoretical (i.e. by MD-EA methods) evaluations of the key material properties (e.g., \( D^* \), \( \gamma_{coh} \), \( C_{cus} \), \( \Delta F_{mix} \)) are needed to further refine the clustering models. Further, interactions between the evolution of CRP and other irradiation enhanced or induced features during both irradiation and PIA may be significant. Such interactions are discussed briefly below.

c. Extended Defect Cluster (EDC) Complexes and Other Radiation Induced and Modified Phases

It is almost certain that extended defect clusters are complexed with solute atoms. Radiation induced segregation (RIS; due to point defect-solute complex migration described previously is implicitly associated with defect clustering. Clustering also drives a second RIS mechanism associated with preferential vacancy exchange (this mechanism is probably not significant for RPV conditions). Complexing may also arise from equilibrium thermal segregation (equal chemical potentials) due to:
- Surface/interface active elements (i.e., carbon, nitrogen, nickel and copper and, particularly, phosphorous) concentrating at the vacancy clusters and dislocation loop stacking faults.

- Strain-energy-induced enrichment of subsized (e.g., carbon and nitrogen and phosphorous) atoms in impurity atmospheres of dislocation loops, and at coherent interfaces and free surfaces.

Thermal and RIS segregation mechanisms can work in tandem. Segregation probably alters the thermal stability and sink strengths of EDC and as well as their effect on the yield stress. However, a more profound consequence of segregation at cluster-complexes may be radiation induced precipitation, at solute concentrations below the thermodynamic solubility limit (Equations 26 and 27).

A large variety of carbide, nitride (or more precisely carbonitride), phosphide and other compound phases have been observed in unirradiated as well as irradiated RPV alloys [30]. In iron the quasi-equilibrium carbon rich phase is orthorombic Fe3C. In low alloy RPV the carbides are highly alloyed (FeMn)3C and (FeMo)2C. Typical atomic carbon concentrations range from about 2 x 10^{-3} to 10^{-2}. However, in heat treated RPV steels most of the carbon is likely to be bound in relatively stable coarse precipitate structures due to its high mobility and low solubility. Hence, carbon may have a modest direct role in embrittlement microstructures. However, this may not be the case for phosphorous and nitrogen.

The temperature and composition dependence of phosphorous and nitrogen solubility can be used to evaluate the propensity to form phosphide and nitride phases under irradiation. The solubility limits \( C_{\text{phns}} \) for pure Fe\(_{a}X_{b}\) (e.g., Fe3P or Fe4N) are given by [24, 72]

\[
C_{xs} = \exp[\Delta^\circ F(\text{Fe}_aX_b)(a+b)/bRT] \quad (44a)
\]

\[
\Delta^\circ F(\text{Fe}_aX_b) = [\varphi^\circ F(\text{Fe}_aX_b) - (a\varphi^\circ F_a - b\varphi^\circ F_b)/(a+b)] \quad (44b)
\]

Here the \( \varphi^\circ \) are the pure compound/element free energies. In the case of alloyed (mixed) compound phases (Fe\(_{1-y}M_y\)_3Xb the expression for \( C_{xs} \) is modified as

\[
C_{xs} = \exp(\Delta^\circ F(a+b)/bRT)/[1+\exp(-\Delta^\circ F(a+b)/aRT)-1]a_m^{a/b} \quad (45a)
\]

where \( a_m \) is the activity of alloying element \( M \) in the iron matrix and

\[
\Delta^\circ F' = \Delta^\circ F(M_aX_b) - \Delta^\circ F(\text{Fe}_aX_b) \quad (45b)
\]

For a pure alloy phase \( M_aX_b \), \( C_{xs} \) is given by is given by
\[ C_{xs} = \exp(\Delta^{0}F(M_{a}X_{b})(a+b)/bRT)/a_{m}^{a/b} \]  

(46)

The assumptions and approximations in these models will be presented elsewhere [24]. However, it is noted that the results are particularly sensitive to the metal atom activities, \( a_{m} \), and thermochemical data (i.e., compound formation energies \( \Delta^{0}F \)) [73-75]. Figure 19 shows solubility limits of phosphorous and nitrogen in equilibrium with for the Fe\(_{3}\)P (Figure 19a) and Fe\(_{4}\)N (Figure 19b) phases along with the typical ranges of impurity concentrations and heat treatment temperatures (600 to 675°C). Both phosphorous and nitrogen fall well below the concentration limit for thermal precipitation during heat treatment. This is also the case for phosphorous at normal service temperatures of about 290°C. However, radiation induced segregation (Equation 27) might reduce the phosphorous solubility limits to negligible levels (dashed line). Iron nitrides could thermally precipitate at 290°C at concentrations near the upper end of the typical range of nitrogen contents. While RIS is unlikely to have a direct effect on Fe\(_{4}\)N formation, segregation of nitrogen to dislocation loops would promote the formation of these precipitates.

A major active alloying element in RPV steels is molybdenum. Figure 19 shows the effect of a small concentration of \( 5 \times 10^{-4} \) free molybdenum on the phosphorous and nitrogen in equilibrium with mixed (MoFe)\(_{3}\)P (Figure 19a) and pure Mo\(_{2}\)N (Figure 19b) respectively. The actual effective free molybdenum could be higher or lower, depending on the composition and volume fraction of the competing carbide phases. While the computed solubility limits may not be very accurate, they qualitatively demonstrate that even small quantities of free molybdenum could induce the precipitation of fine scale alloy carbonitrides and phosphides at 290°C. The formation of iron and alloy carbonitride and phosphide phases prior to irradiation would depend on the alloy's thermal history. Precipitation of most of the residual fraction of these elements left in solution could take place under irradiation.

A plausible sequence of events for phosphide evolution is as follows: a) segregation of phosphorous to unstable EDC-complexes; b) re-arrangement of the complexes into metastable radiation induced (by RIS) Fe\(_{3}\)P phases; and c) subsequent formation of thermodynamically stable (FeMo)\(_{3}\)P at accelerated rates due to RED infiltration of molybdenum. Alternately, alloy phosphides may form directly at molybdenum clusters promoted by RED. The morphology of these phases might resemble thermal phosphides that first precipitate as fine rods (matrix) and platelets (near dislocations) with face-centered-orthorombic habit plates close to \( \{100\} \alpha \) [76]. The upper-bound for the radiation-induced or enhanced phosphide volume fraction is about 4 times the available phosphorous concentration or about \( 2 \times 10^{-3} \).

In iron the equilibrium nitride is fcc \( \gamma \)-Fe\(_{4}\)N, which forms via a series of metastable transition phases [76]. Owing to its high intrinsic mobility, interstitial nitrogen is not a candidate for RIS or direct radiation induced precipitation. However, nitrogen would segregate to EDC and could form iron nitrides. The RED of molybdenum to these sites could result in the formation of more stable Mo\(_{2}\)N. Alternately alloy nitrides may also form
directly at molybdenum clusters formed by RED. The maximum radiation enhanced alloy nitride volume fraction is about 3 times the available nitrogen concentration, or about $1.5 \times 10^{-3}$. Note the actual sequence of transition phases and the participation of carbon in nitrogen rich phases may be somewhat more complex than the simple picture outlined here.

The formation of stable alloy phosphide and nitride phases requires diffusional clustering of molybdenum. Thus, RED precipitation/clustering models based on Equations 30 or 40 might be developed to treat the evolution of these features. A key unknown is the availability of the phosphorous, nitrogen and molybdenum prior to and during irradiation. Solutes located in relatively stable pre-existing phases, or strongly bound at defects and interfaces, initially would be unavailable for fine scale precipitation under irradiation. However, thermal emission and cascade resolutioning (either direct ejection or a disordering mechanism) could provide a continuous source of matrix solutes. Thus over an extended period of time, the microchemistry could evolve towards a lower energy state as the solutes re-partition to more stable alloy phases. This suggests that fine scale alloy phosphides and carbonitrides may emerge as significant features of irradiation microstructures only at high to very high fluence.

There are a number of other chemically active elements in RPV steels which would compete for the available phosphorous and nitrogen. Calculations indicate that manganese produces even larger reductions in the nitrogen solubility than molybdenum. However, the reliability of these calculations is open to serious question. The thermodynamics evaluations also show that copper strongly interacts with phosphorous. This is consistent with observed 'gettering' of phosphorous at CRP and explains the minimal effect of this element at higher copper levels. The effects of additions of other elements (chromium, vanadium and titanium) or higher concentrations of molybdenum have not been considered, but can be treated in a similar manner. Finally, another possible 'animal' in the menagerie of potential low temperature phases could derive from the clustering of free matrix molybdenum (not in carbides) or even low temperate intermetallic laves-phases (e.g., Fe$_2$Mo).

Nickel has been empirically linked with the development of the matrix defects, presumably in the form of EDC-complexes and/or radiation enhanced or induced phases other than CRP [1,6-8, 77]. Nickel may also undergo either thermal or radiation induced segregation to EDC, altering their thermal stability (i.e., $\gamma_{eff}$) and other basic properties. Thermodynamic evaluations suggest that nickel does not play a direct role the evolution of phosphides and carbonitrides. However as noted above, nickel-manganese rich phases may form under some conditions (e.g., low temperature and high solute concentrations). In principle, nickel may also produce indirect effects by its influence on the activities of various elements in solution. Kinetic effects of nickel may also be manifested during thermal heat treatment, (i.e., analogous to the proposed retardation of copper pre-precipitation). Nickel-silicon rich phases (e.g. M$_6$C or G-phase [24]) have been implicated in irradiation hardening of iron based alloys at much higher temperatures and fluences. However, a significant role for of such features seems less likely for RPV conditions. Further, nickel has a significant effect on the overall microstructure which may have an indirect influence on the evolution of fine scale irradiation induced features. Finally, nickel
effects may be partially due to the influence of this element on a wide range of mechanical properties.

Although EDC complexes would be more stable than bare EDC, very small aggregates would still recover rapidly during PIA even at relatively low temperatures. The annealing sequence might first involve reduction in the EDC solute content followed by dissolution by vacancy emission (vacancy-clusters) or absorption (interstitial-clusters). Small radiation induced Fe$_3$P, or radiation enhanced molybdenum and nickel-manganese clusters (or atmospheres) would be relatively stable but would re-dissolve at rates governed by constituent solubility and self-diffusion rates, both depending on the annealing temperature. However, alloy phosphides and nitrides and intermetallic phases would be expected to be thermally stable, coarsening at rates roughly comparable to those for CRP.

In summary, kinetic and thermodynamic models indicate that phosphorous and nitrogen remaining in solution prior to irradiation would be available for: a) segregation to vacancy-clusters and interstitial-loops by thermal and/or radiation induced mechanisms; and b) precipitation of radiation-induced Fe$_3$P (by RIS) or enhanced (FeMo)$_3$P and (FeMo)$_4$N (by RED) phases. Formation kinetics of alloy carbonitride and phosphide phases are probably controlled by the RED of molybdenum. Radiation enhanced diffusion would also promote the formation of nickel-manganese and molybdenum rich phases in some circumstances. A key unknown is the 'availability' of the solutes. However, initially bound solutes may re-partition from metastable sites to the EDC and fine scale radiation enhanced phases at high fluence.

Clearly additional kinetic and thermodynamic modeling of EDC-complexes and a wide array of potential precipitates is needed. Accurate specification solute-defect and solute-interaction and compound formation energies is the key element in developing improved models. Once again advanced MD-EA and other simulation techniques (e.g., Monte Carlo) can contribute to better understanding. Comprehensive characterization of both pre- and post-irradiation microstructures and microchemistries, including the measurements of matrix concentrations of key elements (C, N, P, Mo, Cu, Ni, Mn), is also needed.

Finally, it is important to note that some alloys undergo an alternate form of embrittlement related to thermal segregation of phosphorous. Temper embrittlement may be manifested as a severe weakening of grain boundaries, leading to a change from a transgranular to intergranular fracture. Such effects have not been reported for typical Mn-Mo RPV steels. However, RED/RIS segregation may produce more subtle effects related interface and boundary weakening. Conditions where both hardening and segregation occur would lead to unusually severe embrittlement. A key factor in understanding such radiation enhanced temper embrittlement may be the competition between matrix precipitation and boundary segregation of phosphorous.
d. Summary of the Microstructural Features and Interactions the Evolution of Various Evolutions

Table 2 summarizes the putative characteristics, formation mechanisms and thermal stability of the various irradiation-induced and enhanced features discussed in the preceding sections. A number of cases where the evolution of one feature (i.e. CRP) during irradiation or PIA might be influenced by the coincident evolution of other features (i.e., vacancies-clusters). Figure 20 summarizes some of these key interactions and suggests how they might be treated based on simultaneous modeling of defect and solute clustering. While these co-evolution simulations have not been implemented, they are an important next step in developing more realistic embrittlement models.

VIII. COMPOSITE MODELS OF EMBRITTLEMENT

The various elements described in the previous section can be readily combined into integrated embrittlement model, as demonstrated in Figures 15 and 17. While an extensive discussion of the implications of the integrated models is beyond the scope of this Chapter a few key points should be noted. First, the copper precipitation models have been highly successful in qualitatively rationalizing, and in many cases quantitatively predicting, the separate and coupled effects of many embrittlement variables, including:

- Copper effects on the magnitudes and fluence dependence of RED controlled precipitation strengthening and embrittlement.
- Heat treatment effects on pre-precipitation.
- Copper-phosphorous interactions.
- Nickel effects on CRP volume fractions and number densities and on copper pre-precipitation.
- Manganese and phosphorous effects on CRP volume fractions and number densities.
- Pre-irradiation microstructural effects on RED (sink strengths) and on structure property relations.
- Irradiation temperature effects on precipitate number densities and RED (recombination).
- Flux effects on RED (recombination).
- Combined effects of copper, irradiation temperature, flux and fluence on RED and precipitation kinetics.
- Spectrum effects on free and clustered defect production and RED (recombination).
Effects of dissolution and accelerated coarsening, composition changes and differences in strengthening superposition laws on PIA annealing kinetics.

Effects of copper removal in coarsened precipitates on PIA-re-irradiation kinetics.

The models described in this chapter have also been useful in clarifying the role of extended defect clusters and the effect of key variables on EDC contributions to matrix defect embrittlement. In particular it seems clear that at low-to-intermediate fluences the matrix defects are neither large interstitial loops nor nanovoids. Small vacancy-cluster-complexes, perhaps primarily formed in displacement cascades, are the more likely candidates [25]. However, at least the models (and limited data) also suggest that larger clusters may evolve at high fluences at least under MTR (high flux) conditions. The significance of these features would be expected to diminish for surveillance and vessel service (low flux) conditions. These models also provide a framework to evaluate the effects of various alloying (i.e. nickel) and impurity (i.e., nitrogen) elements on matrix defect cluster complexes and the pivotal role played by cascade clusters on the evolution of matrix features.

One important conclusion derived from considerations of the interactions between CRP and EDC evolutions concerns the effect of flux. Increasing embrittlement (at low fluence) with decreasing flux has been observed in high copper containing steels, while the opposite effect has been found in low copper alloys. This complex behavior can be rationalized and quantitatively modeled [25] based on a dual role of cascade vacancy clusters. In low copper steels the clusters contribute directly to a small increment of strengthening that increases with flux. However, in high copper steels the this effect is off-set by recombination at the cascade clusters. Recombination retards RED and delays CRP precipitation strengthening. These concepts can also be extended to model the complex interactions between copper content, flux, fluence (crossovers) and temperature (inverted sensitivities). However, the most important conclusion is that these particular rate effects do not persist down to very low fluxes.

Note there is experimental evidence that the contributions of some matrix defect features do not decrease at low flux as predicted by the unstable vacancy-cluster models [4-9]. Indeed, theory suggests that interstitial loop evolution might be relatively flux-independent (although loops are empirically known to be sensitive to temperature). A flux independent matrix defect contribution would lead to a continued increase in embrittlement (roughly proportional to \( V \phi t \)) following the completion of flux dependent copper precipitation.

In the either case (flux dependent or independent matrix contribution) a rate effect would not be observed at sufficiently high fluence for low flux. However, other mechanisms may give rise to previously unobserved flux/fluence effects at very high fluences (i.e., new phases) or extremely long times even at very low fluxes and fluences (i.e. thermal aging contributions). For example, the conceptual models developed in this work suggest that new alloy nitride and phosphide phases may evolve under irradiation (perhaps, along with
loops and nanovoids) could increase in importance at very high fluences. The minimum fluence to enter this regime would be expected to decrease with decreasing flux.

IV. CONCLUDING REMARKS

This Chapter has emphasized the role of modeling in advancing the state-of-the-art of predicting irradiation embrittlement of RPV steels. However, it should be made absolutely clear that further development of physically-based models will require well designed fundamental experiments. Further, the ultimate application of these models will be to facilitate the utilization of the engineering data base.

The fundamental experiments include those aimed at particular mechanisms, such as strengthening superposition and recombination. Integrated embrittlement models must also be evaluated and validated by controlled single variable (or single variable combination) microstructural and mechanical property experiments. One key aspect of experimental validation is assurance that the models correctly predict annealing and re-irradiation behavior as well as development of embrittlement during the initial irradiation. This approach to comprehensive model validation is schematically illustrated in Figure 21. Well controlled tests of the models for a wide range of conditions can be used to severely restrict the uncertainties in the model parameters (i.e. \( \gamma_{eff} \)), which may be difficult to evaluate independently.

Perhaps the most important role of modeling is to help to avoid technical surprises. The results of this study point to at least two possible candidate surprises: a) the development of additional embrittlement mechanisms (i.e., new phases, accelerated defect development or even completely new embrittlement phenomena) at high fluences; and b) large shifts in fracture toughness that are not well characterized by surveillance based on Charpy-V notch impact tests. Modeling can play a key role in addressing these and other outstanding embrittlement issues. In particular models can guide the design of critical experiments and in the optimal use of accelerated MTR irradiations needed to obtain answers in a timely manner.

Acknowledgments

The author wishes to thank a number of individuals who have contributed to this work. The creative and untiring efforts by my collaborator in numerical modeling, Dr. B.L. Chao, deserve particular thanks and recognition. Further, the numerical and experimental research of UCSB PhD candidate E. Mader has made a major contribution to our fundamental understanding of embrittlement mechanisms. I also appreciate and acknowledge numerous helpful discussions over the years with many valued collaborators and colleagues particularly G. Lucas as well as W. Phythian, C. English, P. Beaven and T. Williams. Finally, I wish to thank A. Taboada and C. Serpan of the US Nuclear Regulatory Commission (NRC) for their encouragement and guidance. This research was supported by the US NRC under Grant: - G04-088-139 and Contract -04-90-046.
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Figure Captions

Figure 1. Russell-Brown model predictions of the radius dependence of the yield stress contributions of copper and copper rich precipitates and nanovoids normalized by the square root of their volume fraction.

Figure 2. a) Schematic illustration of a dislocation penetrating an approximately equal number of strong and weak dispersed obstacles; and b) the net yield stress change (irradiated or irradiated and annealed) for linear sum (LS) and root sum square (RSS) superposition models for a pre-irradiation contribution from strong dispersed obstacles of 100 MPa.

Figure 3. a) The basis for evaluating $\Delta T_C$ illustrated for the Maine Yankee Weld [36]. The intersection of the maximum load ($P_m$) and general yield ($P_{gy}$) lines specifies the microcleavage fracture stress, $\sigma_f^*$, which is seen to be independent of both test temperature and irradiation. Hence the shift in the Charpy curve at 10 J ($\Delta T_C$) is entirely due to the yield stress increase, $\Delta \sigma_Y$; b) illustration of the method to estimate the extra increment of shift at 41 J due to the upper-shelf energy (USE) decrease, based on the observation of an approximately constant 120°C transition temperature interval.

Figure 4. Tanh Charpy-v notch impact energy curves following irradiation (solid lines and circles) predicted by the strengthening-shift model based on the increase in the yield stress (static room temperature) and unirradiated Charpy curves (dashed lines; data not shown).

Figure 5. a) The fraction of primary defects reaching sinks for matrix recombination as a function of flux for various temperatures; and b) the flux at 50% matrix recombination as a function of temperature and total sink strength.
Figure 6.  a) The fraction of primary defects reaching sinks at 290°C as a function of flux for matrix and vacancy trap recombination; b) the flux at 50% recombination at 290°C as a function of trap binding energy for various trap concentrations and the total sink strengths.

Figure 7.  The fraction of primary defects reaching sinks at 290° as a function of flux for matrix and cascade recombination; b) the flux at 50% matrix recombination as a function of temperature for various surface energies and with and without cascade clusters.

Figure 8.  Illustration a full clustering model of the evolution of vacancy and interstitial aggregates, complexes and precipitates.

Figure 9.  a) Steady-state distributions (jmax=1500) of vacancy clusters at 290°C and a flux of 5x10^{12} n/cm^2-s for \gamma_{\text{eff}}=1.2 J/m^2, with and without the production (1 barn) of 10 vacancy cascade clusters; and b) evolution of the cluster distribution with fluence.

Figure 10.  Variation of nanovoid parameters with fluence for various irradiation conditions: a) number density; b) average radius; and c) volume fraction.

Figure 11.  a) Variation in the steady-state nanovoid nucleation rate at 290°C with flux and \gamma_{\text{eff}}; and b) the flux required to nucleate 10^{17} nanovoids/cm^3 at a fluence of 10^{19} n/cm^2 as a function of \gamma_{\text{eff}} for various irradiation temperatures.

Figure 12.  Vacancy (nanovoid) and interstitial (loop) cluster annealing times as a function of temperature for various \gamma_{\text{eff}} and cluster sizes.
Figure 13. Copper solvus as a function of temperature for various crystal structure and cluster radii. The typical impurity range and heat treatment temperatures are also shown.

Figure 14. Fractional precipitation, \( \Theta \), as a function of non-dimensional fluence, 
\[ \Phi t[2.63N^{2/3}_\text{crp} K_c^{1/3} \text{cup}^{-1/3}] \] from integration of the growth curve (Equation 39, open circles) and the approximate analytic expression (Equation 40, solid line).

Figure 15. The transition temperature shift predicted by the diffusion controlled growth model as a function of the RED factor normalized fluence, \( K\Phi t \), for various copper concentrations and precipitate number densities. Estimates of the copper contribution to embrittlement from the surveillance data base are shown as dashed lines for \( K=10^{-30} \text{ cm}^4 \) (Note, the empirical estimates predict a peak copper contribution, followed by a gradual decline at higher fluences. While this effect could result from overaging, it is actually an artifact of the simple estimation procedure, hence, is not shown).

Figure 16. Predictions of the copper clustering model at 290°C and 4x10^{12} n/cm²-s: a) size distribution for various fluences; b) number density as a function of fluence; c) volume fraction as a function of fluence; and d) average size as a function of fluence.

Figure 17. The yield stress increase predicted by the clustering model as a function of fluence. The solid curve is for a nominal radiation enhanced diffusion factor \( K \) derived using the diffusion controlled growth model. The dashed curve is for \( K \) increased by a factor of 2.
Figure 18. The manganese and nickel content of copper as a function of the precipitate radius, predicted by the non-equilibrium thermodynamic model.

Figure 19. The predicted solubility limits for iron and alloy phases as a function of temperature for: a) phosphorous in equilibrium with Fe₃P and mixed (MoFe)₃P; and b) nitrogen in equilibrium with Fe₄N and Mo₂N.

Figure 20. Illustration some of the interactions between evolutions of various features under irradiation. This flow chart provides the framework for developing an integrated model of microstructural evolution using simultaneous clustering and defect and solute partitioning conservation equations. Sub-models relate the cluster size and composition to their basic properties (i.e. $\gamma_{\text{coh}}$ and $\gamma_{\text{eff}}$) and evaluate defect concentrations and solute diffusion rates.

Figure 21. Schematic illustration of how experiments on microstructural and mechanical property changes during both irradiation and post-irradiation annealing for controlled variations in embrittlement variables (and variable combinations) would be used to validate and calibrate embrittlement models. This illustration is for a two-defect (i.e., CRP and UMD) model.
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<td>copper-vacancy binding energy</td>
<td>( H_{Cu}^{b} )</td>
<td>13.5 kJ/mole</td>
<td>[44]</td>
</tr>
<tr>
<td>Effective surface binding energy</td>
<td>( Y_{eff} )</td>
<td>1.2 J/m²</td>
<td>not known, variable [24,43,58,59]</td>
</tr>
<tr>
<td>Cu-Fe coherent interface energy</td>
<td>( Y_{coh} )</td>
<td>0.4 J/m²</td>
<td>[24,63]</td>
</tr>
<tr>
<td>Copper solubility</td>
<td>( C_{cus} )</td>
<td>( \exp(1.049 - 6057/T) )</td>
<td>At around 300°C for bcc copper [24, 60-62]</td>
</tr>
<tr>
<td>Type</td>
<td>Characteristics</td>
<td>( r (\text{nm}) )</td>
<td>( N_p (\text{}/\text{cm}^3) )</td>
</tr>
<tr>
<td>---------</td>
<td>------------------------------------------------------</td>
<td>----------------------</td>
<td>-------------------------------</td>
</tr>
<tr>
<td>CRP</td>
<td>BCC/Coherent Cu-Mn-Ni ...</td>
<td>~1</td>
<td>( &gt;10^{17} )</td>
</tr>
<tr>
<td>UMD</td>
<td>small ( V_n/V_{nSm} ) vacancy clusters/solute complexes-C,N,Ni,Cu,P</td>
<td>&lt;0.4</td>
<td>( &gt;10^{17} )</td>
</tr>
<tr>
<td>SMD</td>
<td>small ( V_n/V_{nSm} ) loops/segregation atmospheres-C,N,...</td>
<td>&lt;1</td>
<td>( &gt;10^{17} )</td>
</tr>
<tr>
<td>Microvoids</td>
<td>Compact clusters, surface segregation</td>
<td>&gt;0.5</td>
<td>( &gt;&gt;10^{16} )</td>
</tr>
<tr>
<td>Large I-loops</td>
<td>a&lt;100&gt; atmospheres?</td>
<td>&gt;2</td>
<td>( &gt;&gt;10^{16} )</td>
</tr>
<tr>
<td>Carbonitrides</td>
<td>alloyed w/Mo</td>
<td>&gt;1</td>
<td>( &gt;&gt;10^{16} )</td>
</tr>
<tr>
<td>Phosphides</td>
<td>Fe_3P M_3P alloyed w/Mo</td>
<td>&gt;1</td>
<td>( &gt;&gt;10^{16} )</td>
</tr>
<tr>
<td>Other</td>
<td>Mn-Ni-clusters/atmospheres Mo clusters, MoFe_2</td>
<td>&lt;1</td>
<td>( &gt;&gt;10^{16} )</td>
</tr>
</tbody>
</table>
FIGURE 1

The graph shows the relationship between nanovoids and Cu-precipitates versus the radius (r) in nanometers (nm). The y-axis represents the change in yield stress (Δσy/Nf) in MPa. The curves indicate different concentrations of Cu-precipitates and nanovoids, with the Cu-0.25Mn-0.15Ni precipitates showing the least change in yield stress compared to nanovoids.
FIGURE 2

(a) 'Strong' Obstacle 'Weak' Obstacle Pinned Dislocation Moving Dislocation

(b) 

\[
\Delta \sigma_n (\text{MPa})
\]

\[
\Delta \sigma_i, \Delta \sigma_{ia} (\text{MPa})
\]
FIGURE 3

(a) Charpy Energy (J)

Unirr.  
\[
\Delta T_c
\]
Irrad.

(b) Load (kN)

Unirr.  
\[
\frac{d\sigma_y}{dT_t}
\]
Irrad.

(c) CVN Energy (J)

Unirr.  
\[
\Delta T_{41}
\]
Irrad.

\[\Delta U_{SE} = f(\Delta \sigma_y, U_{SE_0})\]
FIGURE 4

Test Temperature (°C)

Chappy Energy (J)

GINNA-W

JOSE CABREIA-W

EP24-W

ZION-2-W

MAINE YANKEE-W

EPRI EP23-W

A533 HIGH SENSITIVITY-W

QUAD CIT-1 43-W

FIGURE 4
FIGURE 5
Cluster Evolution in Vacancy-Interstitial-Solute Space

Interstitial (i) Number \( \rightarrow \) Increasing Vacancy (v) Number \( \rightarrow \) Increasing Solute (s) Number

\( v + i \) Annihilation Reactions

Reactions at Sinks
- Mobile Species
- Discrete or Average Cluster Classes Represented by Concentrations

\( J, K, L \) Maximum Sizes

Clustering Reactions
- \( v \) Emission + i Absorption \( \rightarrow \) \( v \) Emission + i Absorption
- \( s \) Emission
- \( s \) Absorption
- \( v-s \) Absorption / Emission

**FIGURE 8**
Figure 10
FIGURE 13

The diagram shows a plot of temperature (°C) against concentration of Cu (C_{Cu}) on a log scale. The x-axis represents the concentration of Cu ranging from $10^{-6}$ to $10^{-1}$, and the y-axis represents the temperature ranging from 100°C to 800°C. The graph includes lines for different concentrations: 2nm, 1nm, and 0.5nm, indicating the phase transitions of BCC Cu. Additionally, there is a shaded area representing the heat treatment/impurity range.
Figure 14

\[ \theta = \frac{[2.63K(t)N_{crp}]^{2/3}}{[C_{cue}/x_{cup}]^{1/3}} \]
FIGURE 18

Mn
1.0Mn - 0.5Ni

Mn+Ni

250°C
300°C
450°C

IN_X+uMN_X/UN_X

r (nm)
FIGURE 19
Microvoids
v-clusters

Loops
i-clusters

Copper Rich
Precipitates

Other
Precipitates

Segregation/
Nucleation sites

Defect Concentrations,
Defect Fluxes,
and Diffusion Rates
$S_j, D_v C_v, D_i C_i, D^*$

$\gamma_{eff}$

$\gamma_{coh}$

Solute Partitioning
Thermal and Radiation Induced
Segregation, Precipitation

FIGURE 20
FIGURE 21

Mechanical Properties

Microstructure

Embrittlement Models
- Microstructural Evolution
- Structure/Property
CHAPTER 11

THERMAL ANNEALING OF AN EMBRITTLED REACTOR PRESSURE VESSEL

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1. INTRODUCTION

Radiation embrittlement of ferritic pressure vessel steels i.e. usually manifested by a shift (increase) in the ductile-brittle transition temperature (DBTT), a drop in upper shelf energy as measured in the conventional Charpy V-notch test, and a decrease in fracture toughness ($K_C$, $J_C$). A thermal anneal cycle above the normal operating temperature of the vessel can restore most of shift in the ductile-brittle transition temperature, the original Charpy V-notch energy properties and the fracture toughness. While the possibility of restoring material toughness levels through thermal annealing treatments of reactor pressure vessels received little attention, it was not without precedent. Because of sensitive material and a low operating temperature of 239°C (430°F), the U.S. Army SM-1A reactor reached a point where thermal annealing of the reactor pressure vessel was desirable after only a few years of operation. Interest increased in the late nineteen-seventies when the Electric Power Research Institute (EPRI) sponsored a program to assess the feasibility of and to develop a methodology for thermal annealing an embrittled reactor pressure vessel. Under the sponsorship of EPRI, Westinghouse clearly demonstrated the benefits of annealing in terms of mechanical property recovery. In 1984, the BR-3 reactor vessel in Mol, Belgium underwent a "wet" anneal similar to the "wet anneal" of the SM-1A reactor vessel. About 15 commercial power reactor pressure vessels of the WWER-440 type have been successfully "dry annealed." (See Section 4 of this chapter.)

2. THERMAL ANNEALING RESPONSE OF MATERIALS

The irradiation sensitivity and thermal annealing behaviour of reactor pressure vessels materials has been assessed by many investigators. Spitznagel et al studied the post irradiation annealing response of A302 grade B plate and high copper weld metal irradiated at 288°C and annealed in the temperature range 316°C to 454°C. Recovery of pre-irradiation microhardness was followed for all three steels. The measured annealing response of these steels agreed well with the values predicted by a previously developed theoretical model based on the dissolution of copper-vacancy aggregates. A number of authors presented the results of their studies at the 1979 IAEA specialist meeting in Vienna, Austria. Steele presented a review of the IAEA specialist meeting. Sivaramakrishnan carried out post irradiation annealing studies on the two types of steels utilized in the TAPS and RAPP reactors in India and reported that post irradiation thermal treatment can recover irradiation damage. Hawthorne reported on post irradiation heat treatment (annealing) as a method for reversal of detrimental radiation effects for reactor vessel steels. Petrequin and Soulat reported on thermal annealing studies carried out in France. Annealing of irradiation embrittlement was examined on a 508 class 2 reactor pressure steel by heat treatment in the range 300° to 500°C. As with other published data recovery was a function of annealing temperature. Popp et al presented a set of experimental data regarding the influence of sampling depth, neutron irradiation and post-irradiation annealing treatment on properties derived from instrumented Charpy V-notch impact testing, tensile and hardness tests. Complete recovery was reported for Charpy impact energy transition temperatures, hardness, and dynamic yield stress. The most effective recovery took place during short-time anneals of about one hour.
In a programme sponsored by the United States Nuclear Regulatory Commission (NRC) and conducted by Hawthorne et al.[36,37,38] post irradiation annealing and re-irradiation behaviour of plate and weld were investigated. The influence of factors such as copper, nickel and phosphorus contents, weld flux and annealing temperature (399 and 454°C) were studied. Phosphorus had no influence on annealing, but copper and copper plus nickel reduced the annealing affect.

In Germany, post irradiation annealing and re-irradiation experiments on a NiCrMo type weld containing 0.27% Cu (on average) were conducted in the nineteen seventies but published later.[39,40,41] Hardness and Charpy-V tests were conducted after annealing at several temperatures, drop weight and fracture mechanics tests after annealing at 450°C. They showed that annealing at 450°C for 60 hours produced significant irradiation recovery.

A number of papers reported on the development of a model of annealing and reembrittlement consistent with available experimental data that can be applied to commercial reactor pressure vessels. A model based on copper precipitation was reported by Lott, et al.[10] which concluded that the model underpredicted the reembrittlement rate for weldments. Originally, Lott et al.[10] considered two possible mechanisms for annealing; dissolution of copper precipitates and over-aging by an Ostwald Ripening process. The copper concentrations in reactor pressure vessel steels appear to be well in excess of the solubility limits in the temperature range 300°C to 400°C. Therefore, dissolution appeared to be the least likely mechanism. In Ostwald ripening, the larger precipitates grow at the expense of the smaller ones. Pachur[11] identified four distinct annealing stages. Pachur's work has several important implications for the interpretation of the power reactor annealing data. First, the existence of two and possibly three mechanisms of embrittlement indicates that a simple single mechanism model based on copper precipitation will not adequately describe annealing. The existence of a mechanism that is marginally unstable at 290°C helps to explain why the damage produced at lower irradiation temperatures is more easily recovered than damage produced at reactor pressure vessel operating temperatures. Extrapolations based on experience with lower temperature anneals may produce erroneous conclusions. Pachur also indicates that the drop in Charpy upper shelf is caused by type 3 defects and not by type 4 defects. This would explain the results of a previous annealing study, where it was found that the Charpy upper shelf was completely recovered after a 450°C anneal while the Charpy transition temperature was not. Earlier, Nichols[12] published a model for the recovery (annealing) of radiation damage in pressure vessel materials and suggested that annealing of the damage occurs by the thermally activated motion and coalescence of the damage zones as entities through surface diffusion of atoms on the zone. Fisher and Buswell[13] extended and adapted the "Magnox model"[14] to the interpretation of data from PWR pressure vessel steels. The extended Magnox model of Fisher and Buswell[13] can be very helpful in assessing the response of irradiated thermal treatment (annealing). The extended Magnox model suggests that as the irradiation temperature decreases the contribution of displacement damage increases and as the irradiation temperature increases the contribution of copper precipitation increases. More recently, Macdonald[15] developed a correlation to describe the residual shift in the 41 joule Charpy transition temperature after thermal annealing of reactor pressure vessel steels. Macdonald[15] expanded on the previous work of Powers[16] and resulted in a correlation reasonably current for residual post-annealing shifts after annealing of less than 1:1°C.
The annealing and re-irradiation response of typical reactor pressure vessels is given in Figures 1, 2, and 3. Conclusions that can be drawn from references 2 and 17 are:

- The ductile-brittle transition temperature shift recovery is between 80 and 100% after annealing at 454°C for 168 hours.
- The Charpy upper-shelf impact energy recovery is a 100% after annealing at 454°C.
- The re-irradiation after annealing is a function of the annealing temperature. At high annealing temperature (454°C) the ductile-brittle transition temperature appears to continue at the lower rate which would have been expected if no anneal had been performed. At lower temperatures (<400°C) the ductile-brittle transition temperature appears to have a lateral shift.

The results of Leitz et al. confirm the above statements. In addition, they found that hardness also (as upper shelf energy) recovers completely at 450°C for 60 or 168 hours. Transition temperatures, even after 500°C annealing, are not completely recovered but after a 550°C anneal the total transition curve is an improvement on that before annealing.

A. D. Amaev, et al. reported test results of specimens removed from operating VVER-type reactors. Analysis of 40 sets of vessel material specimens were conducted to assess the role of copper and phosphorus. The results of the investigation of annealing effect on recovering the brittle transition temperature included those of material properties after the repeated cycle of radiation following annealing. Figure 4 shows the change of the transition temperature of the vessel materials depending on the neutron fluence with periodic phases of radiation and annealing at 420°C for 144 hours. It is seen in Figure 4 that the degree of the steel radiation embrittlement after annealing did not depend on the material history and was only determined by the condition of the final anneal.

3. THERMAL ANNEAL PROCEDURE DEVELOPMENT

3.1 U.S. Developments

There are two methods of thermal annealing an embrittled reactor pressure vessel: wet and dry. The U.S. Army SM-1A and BR-3 reactor vessel used "wet" annealing for their thermal annealing programme. "Wet" annealing restricts the temperature of the thermal anneal to the design temperature of the nuclear steam supply system (NSSS). At this relative low temperature, full recovery of the mechanical properties is not possible and upon re-irradiation after annealing the increase in the DBTT shift will continue at a similar rate which would have been expected if no anneal had been performed. "Dry" annealing permits selection of an elevated temperature to obtain full recovery of mechanical properties. Therefore "dry" annealing is recommended and a thermal anneal procedure was developed to utilize "dry" annealing.
To assess the feasibility of and to develop a procedure for dry thermal annealing, the following objectives must be met:

- Determine the thermal annealing temperature and holding time required to maximize fracture toughness recovery, minimize re-exposure sensitivity, and minimize reactor downtime.

- Determine the areas of the reactor vessel that may require thermal annealing.

- Determine if the following restrictions would hamper the feasibility of applying a thermal annealing operation:
  - Reactor vessel design and metallurgical limitations
  - Reactor vessel insulation limitations
  - Primary shield concrete limitations
  - RCS piping and associated equipment support restrictions
  - Reactor vessel internals and fuel storage limitations
  - Health physics considerations

- Determine the most efficient method of producing and maintaining the thermal annealing parameters. Possible methods are as follows:
  - Spent fuel assemblies used as heating elements
  - Circulating externally (outside vessel) heated fluid (inert gas)
  - Induction heating elements
  - Circulating internally (inside vessel) heated fluid (hot air)
  - Resistance heating elements

- Develop a conceptual apparatus to be used to thermally anneal a reactor vessel.

- Develop an in-situ thermal annealing procedure.

- Develop technical specifications related to the thermal annealing procedure.

In Germany, as a provision, annealing equipment for annealing the central circumferential weld seam at the beltline of a reactor pressure vessel at 450°C was designed, constructed, and tested on a dummy vessel. Later on, it was shown, however, that the annealing procedure was not needed.

The results of the EPRI work indicated that a thermal annealing method based on dry, localized heating could be used to apply the annealing temperature of 454°C for 168 h in an effort to restore the fracture toughness properties of ferritic materials of the reactor vessel as required by 10CFR Part 50 Appendix G. The method would require an apparatus designed so that it could be lowered on the reactor vessel. After a watertight seal is formed between the inside of the reactor vessel and the
refueling cavity, the primary water inside the reactor vessel would then be pumped out into the cavity. The resistance type heating elements system, designed in a manner so that the area requiring thermal anneal would be heated, could then be cycled on and off to achieve the specified thermal annealing parameters. This method could be adapted to the standard two-, three-, and four-loop plants. Removal of the reactor vessel internals and the fuel assemblies would be required. The maximum volume of a reactor vessel that may require a thermal annealing treatment would comprise that material from some point below the beltline region up to and including the intermediate-to-upper shell girth seam. This volume is described in Figure 5.

3.2 Thermal Annealing Equipment

In developing the EPRI conceptual thermal annealing equipment, the following factors were considered:

- ability to produce and maintain a temperature of 454°C for a holding time of 168 hours.
- ability to control the heatup rate to less than 28°C/hour.
- power requirements
- costs
- logistics

The annealing apparatus (Figure 6) would be lowered on the reactor vessel with the assistance of the overhead polar crane. A watertight seal would be made between the support plate (4) and the reactor vessel internals support ledge (18) to prevent leakage between the inner confines of the vessel (24) and the refueling cavity (16). This would be a secondary water seal. The primary water seal would be made between the water shield barrel dam (8) and the bottom of the refueling cavity (16) by the use of a new design cavity seal ring (5).

3.3 Generic Thermal Annealing

The following would be the general thermal annealing procedure on a generic basis.

(1) All unassembled thermal annealing equipment would be moved into the containment building through the equipment hatch. This equipment would then be transferred to the operating deck.

(2) The missile shield, cavity gates, seismic restraints, control rod drive mechanism, vent air ducts, and closure head areas insulation would be removed and stored per the standard operations procedure.
(3) The reactor vessel studs would be detensioned, unthreaded, and then removed per the standard operations procedure.

(4) The new design cavity seal ring for adapting the thermal annealing water shield barrel dam would be installed.

(5) The closure head would be removed and the refueling cavity flooded. Then the upper internals would be lifted and placed in storage in the reactor vessel internals storage area. In parallel with this procedure, the thermal annealing equipment would be prepared for assembly.

(6) All fuel assemblies would be removed and transferred to the spent fuel pit. In parallel with this procedure, the thermal annealing equipment would be assembled.

(7) After all fuel assemblies have been transferred, the lower internals would be lifted and placed in storage in the reactor vessel internals storage area.

(8) The annealing water shield barrel dam would be lowered by the overhead polar crane and sealed to the floor of the refueling cavity.

(9) The reactor vessel would be drained to just below the vessel flange. The water would be pumped out of the inner shield barrel dam volume into the remainder of the refueling cavity.

(10) The seals on the water shield barrel dam would be checked for leaks and a segment of the cavity seal ring would be removed thereby operating the air path between the lower reactor vessel cavity and upper containment.

(11) The reactor vessel would be drained to a water level just below the reactor vessel nozzles.

(12) The nozzle plugs would be installed.

(13) The thermal annealing apparatus package would be lowered onto the reactor vessel by the overhead polar crane. All vent and drain lines would be checked.

(14) The remaining water would be pumped out of the vessel into the refueling cavity.

(15) The resistance heaters would be energized using a low voltage power source to dry out the heating elements. All electrical connections, heater controllers, X-Y recorders, and thermocouple functions would be checked.

(16) The resistance heaters would be cycled on and off using a high voltage power source to achieve and maintain the annealing temperature (454°C) for the required period of time (168 hours).

(17) The vessel would be permitted to cool to ambient temperature.
(18) The cavity seal ring segment, removed previously, would be installed.

(19) Water would be transferred from the refueling cavity to the reactor vessel, to a level just below the nozzles.

(20) The thermal annealing apparatus package would be removed from the vessel by the polar crane, and the package would be decontaminated.

(21) The nozzle plugs would be removed and decontaminated.

(22) The inner confines of the water shield barrel dam would be filled with water to equalize the water levels with the refueling cavity.

(23) The water shield barrel dam would be removed, decontaminated, and disassembled.

(24) All equipment would be packaged and removed from the containment building.

(25) The lower internals, all fuel assemblies, and the upper internals would be installed to standard operations procedure.

(26) After draining of the refuelling cavity, the closure head would be installed to the standard operations procedure.

(27) The cavity seal ring would be removed.

(28) The reactor vessel studs would be installed and tensioned to the standard operations procedure.

(29) The closure head insulation, control rod drive mechanism vent air duct, seismic restraints, cavity gates, and missile shield would be installed.

4. ANNEALING OF WWER PRESSURE VESSELS

Background studies on the heat treatment of WWER-440 reactor vessels and the development of processes and hardware by the leading organizations in the then USSR led to the pilot operation annealing of Novo-Voronezhskaya NPS, Unit III by "dry" method (430 ± 20°C, 150 hours). In 1988 the Armenian NPS, Unit I, was annealed by a revised procedure (450 + 40°C, 150 hours). The experience of performing these two first recovery heat treatments of operating power reactors verified the procedures adopted. The annealing of eight more WWER-440 vessels of reactors has been carried out: NPS "Bruno Leuschner," Unit I, GDR (1988); Kolskaya NPS, Units I and II, USSR (1989), NPS Kozlodui, Units I, II and III, Bulgaria (1989 and 1992), NPS "Bruno Leuschner," Units II and III, GDR (1990).
The investigations showed that for the assurance of safe operation and life extension of radiation of WWER-440 reactor vessel with regard to the brittle failure strength criterion would be met by annealing the weld in the high fluence area of the pressure vessel which was the controlling region with regard to irradiation embrittlement. This feature eliminated many technical difficulties if annealing of the entire reactor vessel had been needed and made the process of restoration of properties of vessel metal practicable.

The heating equipment was developed and fabricated by NPO "TZNITMASH" and PO "Izhorskyj zavod." The heating equipment (Figure 7) is a welded construction incorporating a heating unit, a cover, being both a biological and thermal protection, and a cross-piece connecting the heating unit and the cover. The heating unit has 54 panels of one-sided heaters of radiative heaters arranged into three circular rows, each row has 18 electrical heaters. Each row of electrical heaters is divided into 3 independently controlled areas having two paired thermoelectric temperature transducers (thermocouples) to monitor the vessel temperature in the area of annealing and the total number of thermocouples is eighteen.

The heating equipment also includes a distributing board, power cabinets, and transformers (one for each heating zone). The transformer power is 90 Kv. Also included are cabinets to control the heaters; a system of power supply communications, monitoring and control circuits, a system for automatic maintaining the annealing conditions by the assigned programme (Remicont), a system for recording thermocouple readings during the anneal, a system for remote control and monitoring thermal and electrical conditions. a jig for assembling and adjusting and temporary storage of the equipment.

Before the start of vessel annealing the following procedures are followed, emptying the reactor vessel, its drainage, cleaning and drying, checking the equipment cooling system of the reactor pit to prevent overheating of structure surrounding the vessel during annealing. Also measures are taken to exclude water, cold air and other materials into the reactor vessel during heat-up, hold-time during annealing and cooling down:

- work on steam generators, reactor coolant pumps (RCP), pressurizer is forbidden;
- closure of all main gate valves (MGV) is checked;
- work on non-disconnected sections of the main coolant circuit is prohibited;
- electrical circuits of the RCP and MGV are dismantled;
- the hydroseal of the cooling pond and refueling channel are leak tested;
- closing valves of pulse tubes of inspection of the reactor main joint leakages is checked;
After erecting the jig, assembling the heating equipment, connection of electrical equipment and unit-by-unit inspection of thyristor controls at the nominal current load, the heating apparatus is put into the reactor vessel and located on its main joint. The equipment design provides location at the middle ring of heaters opposite the axis of the weld being annealed (the annealing zone covered by the heating equipment is not less than ± 0.7 m with respect to the weld axis).

The following temperature-time conditions were used to anneal the WWER-440 welds:

- annealing temperature of 475 ± 15°C;
- holding time of 150 hours.

Annealing conditions are monitored with thermocouples. After the equipment has been inserted into the reactor vessel, a special mechanism moves the thermocouples and presses them to the internal surface of the vessel wall. The same mechanism presses the thermocouples tight against the vessel wall during its expansion during annealing. Additional thermocouples are installed outside the reactor vessel (nozzle area, supporting shoulder, cylindrical part and bottom), in the channels of ionization chambers of the tank of biological shielding, on the concrete of the reactor pit and on the thermal insulation of the vessel (Fig. 8). The number of additional thermocouples is not less than twenty. Thermocouple readings are continuously recorded.

The following restrictions apply during annealing:

- the heating up rate is not higher than 20°C/h;
- the cooling down rate is not higher than 30°C/h;
- the temperature of the vessel supporting shoulder is not to exceed 300°C;
- the temperature of the biological shielding tank water is not to exceed 90°C;
- concrete temperature of the reactor pit in the area of tank fastening – not higher than 90°C;
- stratification of temperature by the readings of thermocouples of one positions is not to exceed 50°C.

During annealing routine calculations of the thermal state of the vessel and pit equipment components are carried out. Temperature trends are analyzed and evaluated and any corrections needed are made.
Annealing conditions are at temperatures above the operating temperature (annealing temperature is 490°C). Heat flows from the region being annealed to the top and bottom regions of the vessel. Because of this, a major fraction of energy which the vessel receives from heaters is spent in heating the vessel. Some fraction of energy is lost through the thermal insulation and there is a heat sink through the supporting structure.

Local heating of the annealing area produces stresses and these result in deformation of the reactor vessel. Maximum stresses act at the edge of the annealing area at when the highest temperature gradients occur both along the height and through the thickness of the vessel wall. At the hold stage as the vessel is being warmed up, stresses decrease. Stresses arising in the vessel during annealing should not be at a level to cause metal creep, residual strains and cause stresses which could prevent reactor vessel safe operation. Implementation of annealing conditions must not result in overheating of structures surrounding the vessel. Mathematical models and corresponding computer programmes were developed to analyze the thermal-stress state of the reactor vessel during annealing.

In all actual vessel anneal cases thermal conditions of the vessel and reactor pit equipment as a whole corresponded to the design one and temperature of separate components of the vessel and pit equipment did not exceed that permitted. Check calculations of vessels strength performed using temperatures actually measured during annealing verified the fact that stresses did not exceed the design values.

Prior to and subsequent to annealing the vessel was inspected nondestructively. The results show that annealing does not initiate new, or indeed develop flaws.

When the hold stage (150 hours at a temperature of 475± 15°C) is completed then the heating equipment is turned off and the vessel is gradually-cooled down to temperature not more than 70°C and then the vessel is filled with water.

The evaluation of the vessel metal properties and quality control of the annealing process are matters of great importance. This activity encompasses the determination of variation of mechanical properties, and the investigation of the degree of recovery of mechanical properties by means of metal sampling and also the measurement of hardness (for vessels without cladding).

The VNIIAES Institute has developed and fabricated equipment for sampling weld metal from the reactor vessel inner surface in the form of a 'chip' to conduct chemical analysis from the first 5 mm of the vessel wall and then grind the locations of sampling. The equipment includes a milling cutter device, a chip collector, grinding device, control cabinet, and a rig for simulating and adjustment of facilities and devices.

To perform the work the milling cutter and grinding devices are hung on the multipurpose maintenance cabin (MMC) or on the protective container (PC). Metal sampling is carried out along the weld axis in three places at an angle 120° to one another.
Chemical analysis of the sampled chip allows the determination of phosphorus and copper in the weld metal and to check the brittle strength of the heat treated reactor vessel. Table 1 represents values of phosphorus and copper content for the reactor vessels subjected to annealing.

Hardness is measured prior to and subsequent to heat-treatment with the help of an automatic hardness meter T-4M (designed and fabricated by VNIIAES to determine the variation of mechanical properties of reactor vessel base and weld metal. The hardness meter is a multipurpose machine for establishing values of Brinell (HB) hardness of metal of the reactor vessel linear surface. The apparatus has a measuring head, a moveable carriage with magnetic fastening, a control desk, a unit for processing the results, and for operation it is hung onto the multipurpose maintenance cabin or protective cabin in a similar way to the metal sampling equipment.

The machine is fixed on the internal surface of the reactor vessel with a magnetic frame (pressure force 900-1200 kgf), and after that the moveable carriage with the hardness meter head attached on it is set with respect to the selected measurement point. The entire process of loading and load relief is carried out in an automatic way with simultaneous recording using a two-coordinate recorder and a minicomputer memory. The range of hardness measurements is 10 - 500 HB. The results are processed on the computer immediately after finishing the complete cycle of measurement.

Hardness measurements of weld and base metal of adjacent shells is carried out on non-ground surfaces at three points in close vicinity to the metal sampling positions above and below the weld axis. Measurements are made at each location before and after annealing for 10 loading cycles and with a distance between measurement points of 10 - 15 mm.

Hardness results (See Table 2) clearly demonstrate the decrease of hardness values after annealing and weld metal hardness is reduced to a greater extent.

After annealing the NPP Bruno Leuschner Unit I, and before and after annealing the NPP Bruno Leuschner Unit II and Kosloduy Unit II, Samples were removed from the inner surface of the RPV and these were big enough to provide subsize V-notch impact specimens. The results of been reported elsewhere (25) and they showed that the transition temperature after annealing was lower than predicted. (Testing of samples of Bruno Leuschner was discontinued after the German Authorities decided not to continue with the operation of the plant. It is proposed to complete the tests as part of the general watch background for WWER 440/230 reactors). The Kosloduy II samples are being tested as part of a cooperative effort by Electricite de France, the Kurchatov Institute and Prometey Institute, and Siemens. The results are not yet available for publication.

The work schedule for reactor vessel annealing including metal sampling and measurements of hardness before and after heat treatment amounts to about 15 days which is included in the maintenance schedule of the unit.
At present there are two sets of equipment for annealing WWER-440 reactor vessels. This factor allows up to 4 annealings per year. The heating equipment is collapsible in design and consists of separate transportable sections permitting thorough decontamination and, fits into containers that can be transported to NPS by road, rail or water.

5. RECOMMENDED GUIDE FOR THERMAL ANNEALING

The American Society for Testing Materials (ASTM) has issued a recommended guide which covers the general procedures to be considered for conducting an in-service thermal anneal of a light-water-cooled nuclear reactor vessel and demonstrating the effectiveness of the procedure. The guide, ASTM E509, "Standard Recommended Guide for Inservice Annealing of Light-Water Cooled Nuclear Reactor Vessels," is designed to accommodate the variable response of reactor-vessel materials to post-irradiation heat treatment at various temperatures and different time periods. The guide describes certain inherent limiting factors which must be considered in developing an annealing procedure; these factors include (a) system design limitations, (b) physical constraints resulting from attached piping, support structures, and the primary system shielding, and (c) the mechanical and thermal stresses in the components and the system as a whole. The guide also provides guidance for developing a post-annealing surveillance vessel radiation surveillance programme to monitor the effects of subsequent irradiation of the annealed-vessel beltelline materials.

Appendix B, "Guidance for Verifying Recovery and Re-Irradiation Embrittlement," to ASTM E509 (21) addresses the new rate of re-embrittlement after annealing. The guide introduces the concept of horizontal or lateral shift of the initial irradiation embrittlement path to become the post-anneal re-irradiation trend curve and vertical shift (vertically downward). The generally accepted concept or model for the rate of re-embrittlement is the lateral shift.

The U.S. Nuclear Regulatory Commission (NRC) is in the process of preparing a NUREG that will address reactor pressure vessel annealing. It is expected that NRC will incorporate the basis and concepts identified in ASTM E509 into the NUREG.

6. SUMMARY

The use of a thermal heat treatment to recover mechanical properties which were degraded by neutron radiation exposure is a potential method for assuring reactor pressure vessel compliance with regulatory licensing rules and for license renewal. Research programmes worldwide have clearly demonstrated the recovery effects on annealing irradiated steels and weldments. "Wet anneals" at temperatures less than 343°C have been conducted in both the United States and Belgium (SM-1A reactor and BR3 reactor). The Russians and other countries in eastern Europe have performed "dry anneals" at higher temperatures (450°C) than design temperatures (343°C) for a number of commercial reactor vessels.
In the United States, a standard recommended guide for inservice annealing of reactor pressure vessels has been published by ASTM\textsuperscript{(21)}. This recommended guide is expected to serve as the basis for a NUREG to be issued by the US NRC on inservice annealing of reactor pressure vessels in the United States. Technical and economic uncertainties have made utilities in the United States reluctant to seriously consider thermal annealing to date of their reactor pressure vessels. However, as utilities begin to experience significant radiation embrittlement or consider extending the operating licensing life of the vessel, thermal annealing of the reactor pressure vessel will take place. Griesbach and Server\textsuperscript{(22)} have shown that thermal annealing of an embrittled reactor pressure vessel is of economic benefit.
REFERENCES


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<th>Content of Elements in Metal of Weld No. 4, %</th>
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<td>Used in Design Verification</td>
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HARDNESS RESULTS FROM THE REACTOR VESSEL OF KOLSKAYA NPS, UNIT I, PRIOR AND SUBSEQUENT TO ANNEALING

<table>
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<tr>
<th>Place of Measurement</th>
<th>Hardness Values Prior to Annealing ( \text{HB} ) (irrad.)</th>
<th>Hardness Values Subsequent to Annealing ( \text{HB} ) (anneal.)</th>
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<td>( \text{HB}_{\text{irrad.}} )</td>
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Figure 1. The Ductile-Brittle Transition Temperature Shift Recovery Between 80 and 100°F After Annealing at 454°C for 168 Hours.
After annealing at 371°C for 168 hours, the ductile-brittle transition temperature shift recovery between 20 and 40%.

Figure 2.
Figure 3. Re-Irradiation After Annealing is a Function of the Annealing Temperature.
Figure 4. Diagram of Change of Critical Brittle Temperature of 15x2 M9A Steel (a) and its Weld Metal (b) Depending on the Neutron Fluence at Periodical Alternation of Irradiation and Annealing at 420°C (During 144 Hours).
Figure 5. Thermal Annealing Zone-Cases 1 to 3.
Figure 6. Design of Conceptual Thermal Annealing Apparatus.
Figure 7. Cross Section of the Heating Element Installation
Figure 8. Arrangement of Thermocouples on the Vessel and Reactor Pit Equipment.

Pos. 1-8 of the Figure Correspond to Additionally Installed Thermocouples; No. 9-II - to Thermocouples of the Heating Apparatus.
12 SAFETY ASSESSMENTS USING SURVEILLANCE PROGRAMMES
AND DATA BASE

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1. BACKGROUND

Generally speaking the licensing authority is required to ensure that all the requirements provided by law to assure the safe operation of Nuclear Power Plants (NPP's) are met.

The safe performance of important components in a nuclear power plant during its operational life is ensured in principle by two complementary sets of actions. These can be defined as safety assurance and assessment of the structural integrity.

- safety assurance is based primarily on the safety concepts such as defense in depth, redundancy where feasible and so forth. These are concepts which have been implemented and safety related measures and actions taken during the design, construction and operation of the plant. These include the safety systems to cope with accident conditions. For the "structural systems", in particular the components of the primary and secondary systems, this requires appropriate design, selection of materials and manufacturing processes, and procedures. The effectiveness of the quality assurance system plays a decisive role.

- the assessment of plant safety during their operational life means primarily evaluating the integrity of the important "structural systems". All components relevant to safety have to maintain their structural integrity and perform their intended functions under all expected operational conditions, thereby preventing failures of these components.

Therefore after the NPP has received the operating permit, the main activities of the licensing authority and the owner of the NPP are the assessments of the plant safety and consequently the taking of all necessary measures for its assurance.

2. THE ASSESSMENT OF SAFETY

The assessment of safety or structural integrity of a structure or component, in principle, has to consider all modes of failure they can experience beginning from intolerable deformation to leakage and eventually fracture of the structure or component. Although for the "structural system" all modes of failure that render the system nonfunctional have in principle the same "failure status", their effects in relation to safety are not the same. Of all the failure modes that can occur, the fast fracture of a component can be considered the "worst case" which has the highest potential of catastrophic consequences. Therefore assessments made on components relevant to safety are primarily focused on preventing fracture, especially brittle or
nonductile fracture of these components. The appropriate method to assess their structural integrity with respect to fracture involves the use of fracture mechanics analysis.

The assessment of safety in the various fields in connection with the structural integrity of primary system components are usually based on the following:

- the state of the art in science and technology of the subject in question.
- status of the worldwide experience on the subject.
- regulatory position in different countries on the subject.

Since the state of the art in science and technology in the various fields is continuously progressing the bases for the assessment also have to be adjusted to comply with the state of the art and as considered necessary by the regulators.

The normal procedure is that the utilities have to present their safety case (safety evaluations) to the licensing authority as required by the regulations of the country where the nuclear power plants (NPPs) are located. The licensing authority then has to critically review and assess the adequacy of these evaluations and their compliance to the regulations.

From experience to make the reviewing process straightforward and expeditious, the submittals to the licensing authority of these evaluations should be correct, comprehensive, clear and complete as well as that the data and references cited are readily verifiable and respectively easily traceable.

3. THE EFFECT OF NEUTRON IRRADIATION ON THE SAFETY OF REACTOR PRESSURE VESSEL (RPV)

Materials exposed to neutron irradiation will experience a certain loss of ductility and fracture toughness as can be measured by conventional and fracture mechanics specimens. This loss of ductility or embrittlement, primarily in the core belt region of the RPV, will make the material more susceptible to nonductile fracture. This change in the fracture resistance of the material has to be taken into account when assessing the structural integrity of the RPV.

In principle there are two ways of monitoring the change of material properties by neutron irradiation: the so-called "direct" method and the "indirect" method.

In the USA and all countries in western Europe and Asia having Light Water Reactors (LWR) NPPs originating or derived from the "USA-Type LWR", normally use the "direct" method to monitor the irradiation damage. That is the change of the ductility or fracture resistance of the material is monitored "directly" by means of a surveillance programme according to ASTM standard E 185 [1]. Capsules containing Charpy V-notch specimens, tensile and sometimes fracture mechanics specimens (prepared from the specific RPV materials to be monitored) are placed in the RPV at the level of the core belt region. Due mainly to the space available in the RPV a sufficient number of fracture mechanics specimens of appropriate type and size cannot be included in the surveillance programme for the determination of the $K_{IR} = f$
(temperature) curve in the irradiated condition. Therefore present irradiation surveillance programmes are based mainly on Charpy V-notch impact bend specimens. The reference nil-ductility transition temperature (RT_{NDT}) (according to ASME code Section III article NB 2331) shift as a relative measure of embrittlement is determined from the Charpy-V transition temperature curves, usually at a fixed level of Charpy energy (i.e. 30 ft.lbs. or 41 J respectively 50 ft.lbs or 68 J). Under the assumption of identical shifts of the Charpy V-notch transition curves and the pressure vessel material fracture behaviour, as expressed by the K_{IR} = f(T - RT_{NDT}) curve in the ASME Code [2], the RPV integrity is evaluated and the pressure-temperature limits for the RPV are derived.

A surveillance programme containing fracture mechanics specimens can therefore additionally provide "valid" (applicable to real structure) specific fracture toughness values provided the type and size of the specimens can fulfill the validity and transferability criteria. These measured fracture toughnesses in the irradiated condition can be used to check or confirm the applicability of the K_{IR} curve mentioned above.

This present practice for determining the RT_{NDT} temperature shift and the loss of ductility stems from the US and has been used in most countries with LWR-NPP. It is to be noted, however, that this practice is based on the following "assumptions":

- there exists for the ferritic low alloy steels used for RPVs a certain correlation between the NDT-temperature as determined by the Pellini Test according to ASTM E 208 [3] and the Charpy V-notch test according to ASTM E 23 [4], hence the RT_{NDT} concept according to ASME code Section III article NB 2331 to assure a sufficient increase (steepness of the Charpy-V curve) in ductility of the material with increasing temperature (>50 ft.lb.energy level and 35 mils lateral expansion at NDT + 60°F)
- this certain correlation (definition) holds after neutron irradiation.
- the reference K_{IR}-temperature curve, when properly indexed to the RT_{NDT} describes the fracture toughness properties of that heat of material. The reference curve, which applies to all ASME Code Section III ferritic materials having a minimum specified yield strength of 50 ksi or less, is construed (intended) to be a lower bounding function of all available measures of fracture toughness of such materials. It includes both static and dynamic fracture toughness as well as crack arrest data (K_{ji}, K_{id} and K_{ia}), but the basis is K_{id} and K_{ia}. Research activities are currently underway to reassess and if indicated further validate or adjust this reference curve.
- the shift of the Charpy V transition curves indexed at a certain Charpy-energy level of 50 ft.lb(68J) later changed to 30 ft.lb (41 J) is identical to the shift of the pressure vessel material behaviour as expressed by the reference K_{IR} = f(T - RT_{NDT}) curve in the ASME code [2].
- the shape of the reference K_{IR} curve remains the same after irradiation.

The above assumptions have not all been verified to be justified as either correct (i.e. complying to the present state of the art in science and technology as well as experience) or conservative (although not strictly scientifically correct the results obtained are conservative). Regarding the assumption on the change of measuring the
shift at the 30ft.lb energy level, although stated as based on surveillance data still seems not yet to be justified since no confirmation has been given that at the new obtained (shifted) RT_{NDT} value + 60 °F, a minimum Charpy V energy level of 50 ft.lb. and 35 mils lateral expansion (LE) (of at least three Charpy V specimens) are attained. RT_{NDT} always has to be understood together with 50 ft.lb. Charpy V energy level and 35 mils LE according to the definition in the ASME code [2]. Research activities in the USA and Europe e.g. Finland are currently underway to reassess and consequently confirm or change/adjust these assumptions.

It has to be noted that the USNRC has played and is still playing a leading role in the regulatory aspects of this field, has issued extensive essential documentation dealing with several aspects of neutron irradiation embrittlement that can be considered as unique in the world and which has been used in one way or another worldwide, i.e. regulatory requirements, regulatory guides and other relevant documents of which the most important are the following:

  - Section 50.61: "Fracture Toughness Requirements for Protection Against Pressurized Thermal Shock Events".
  - Appendix A: "General Design Criteria for NPP, Criterion 31, Fracture Prevention of Reactor Pressure Boundary".
  - Appendix G: "Fracture Toughness Requirements".
  - Appendix H: "Reactor Vessel Material Surveillance Program Requirements".
- Regulatory Guide 1.99, Revision 2: "Radiation Embrittlement of Reactor Vessel Material".
- Regulatory Guide 1.154: "Format and Content of Plant- Specific Pressurized Thermal Shock Analysis Reports for Pressurized Water Reactors".
  - Chapter 5.3.1 "Reactor Vessel Materials".
  - Chapter 5.3.2 "Pressure Temperature Limits"
  - Chapter 5.3.3 "Reactor Vessel Integrity"

In most east European countries with WWERs, it seems that the "indirect" method is the normal practice. The shift is solely determined by a formula based on generic data of the materials used for the RPV. According to information obtained also the definition of NDT-Temperature differs from the US-practice. There is no similar test such as the ASTM E 208 (Pellini) test [3] to determine the NDT-temperature.

In the following we will primarily deal with relevant aspects related to the US-practice, which in principle also form the bases for the practice in most of the European and Asian countries having NPPs.

4. CONSIDERATIONS CONCERNING THE APPLICATION OF SURVEILLANCE PROGRAMME RESULTS

The surveillance programme results, in principle, should be such that they can...
be used in the integrity assessment of the RPV. This means that the results must realistically represent the material behaviour of the RPV materials being monitored. The ASTM E 185 standard [1] first published in 1961 was actually conceived to provide the necessary elements for this realisation. However, because of several boundary conditions at hand (e.g. space limitations and geometrical conditions in the RPV due to the specific design, possible locations of the capsules, available number and type of specimens, the progress in the state of the art etc.), ideal surveillance conditions from current viewpoint have rarely existed.

Important aspects to be considered in connection with the usefulness of surveillance programme results are the following:

- the representativeness of the materials used in the surveillance programme (e.g. the same heat, manufacturing processes, chemical composition including impurities and heat treatments matching the corresponding RPV materials)
- type and number of specimens available in each set of withdrawal and the number of sets
- the knowledge of the exact removal location in the thickness direction and the orientation of the specimens
- the representativeness of irradiation-conditions such as temperature history, fluence and fluence rate experienced by the specimens compared to the corresponding RPV materials being monitored (as well as the use of appropriate thermal monitors)
- adequate calculational and dosimetry methods for determining RPV and specimen neutron fluence.

4.1 Representativeness of surveillance materials

The selection of the materials of the RPV to be monitored is in principle described in ASTM E 185 standard [1]. The materials used shall be from the same heats and if possible also from the same plates or forgings as are used for the RPV (e.g. produced with extra length). In any case it shall be ensured that all the heat treatments and manufacturing processes seen by the materials used in the surveillance programme reflect as closely as possible the RPV materials to be monitored. Also detailed chemical compositions of the surveillance and RPV materials including all alloy and residual elements as well as impurities shall be given for comparisons. It is imperative to ensure that the materials chosen for the surveillance programme exactly or as close as possible represent the RPV materials to be monitored.

4.2 Capsule availability and withdrawal plan

The present practice shows up to 6 sets (withdrawals) for PWR and 3 sets for BWR in the surveillance programme. The difference stems from the expected end of life fluence (usually after 40 years) of the NPP. If in the future an extended plant life (> 40 years) is anticipated, the required number of sets has to be reevaluated. This has to be based primarily on the experiences we have with present practice and their adequacy in fulfilling the intended function. Caution dictates planning for this exigency at the initial plant project.
Present surveillance programmes contain mainly Charpy V-notch and tensile specimens. This corresponds to ASTM E185 requirements and concerns also the number of specimens for each set (minimum requirements). However some reactor vendors already have included a few WOL or other fracture mechanics specimens in their surveillance programme. Also ASTM recommends the inclusion of fracture mechanics but does not require them specifically. However, a new standard is in the making which addresses this question more specifically. This is ASTM standard E636 [5]. Anyhow the type and number of specimens to be included in future surveillance programmes should be reevaluated. The present practice mostly containing only tensile and 9 - 12 Charpy specimens and no fracture mechanics specimens has proven to be unsatisfactory. In some European countries attempts to improve the surveillance programme have been initiated e.g. in Switzerland, where additional precracked Charpy-type specimens and three point bend specimens have been included in the surveillance programmes of the Gösgen and Leibstadt NPP [6].

The basic considerations for fracture toughness specimen inclusion should be (see also [7]):

- adequate and reliable results (information); i.e. sufficient number of specimens to facilitate the assessment of uncertainties
- versatility; the specimens should permit the testing in the measuring range of interest (from the elastic to the elastic-plastic or if possible, the fully plastic region) and should permit the determination of the most important fracture mechanics characteristics, such as $K_c$, $K_{ic}$, $J_{lc}$ and $d_i$.
- the greatest possible measuring capacity (for a given capsule cross-section) for the determination of the fracture mechanic material property of interest.

Last but not least, the results of the Coordinated Research Program (CRP) of the IAEA should also be considered.

4.3 Selection of surveillance specimens orientation from base materials

In forgings and plates used for RPV’s there is always a gradient in the material properties in the thickness direction. Depending on the degree of rolling or forging, the thickness, the heat treatment, and chemical composition (sulphur and alloying elements that influence hardenability), the differences observed can be quite significant. This information has to be duly recorded and shall be considered in the evaluations. This observation reflects more the first and second generation RPVs but for current RPV-steels with state of the art manufacturing technique the differences should be small. However, to provide sound bases for the analysis of the test results, especially when unexpected results are obtained, the exact removal locations have to be known. ASTM and ASME practices require specimens to be taken from the 1/4 T location.

The orientation, that is the crack plane orientation of the specimens, is a decisive point that has to be considered. There are quite significant differences in fracture resistance in the different orientations. Definitions and abbreviated terminology on this subject can be found in ASTM E 399 standard [8]. In safety assessments, results...
obtained from the so-called weak orientation of the specimens normally are used, T-L for plates (Fig. 1. in ASTM E 399) respectively C-L orientation for bar and hollow cylinders (Fig. 3 in ASTM E 399) or transverse orientation for Charpy V Notch-Bend specimen. Here L denotes the maximum grain flow (working direction). However for RPV made from forgings the maximum grain flow (working direction) of these forgings is normally the circumferential direction. Therefore for RPV made of forgings the weakest direction is normally the L-C orientation (transverse orientation) and not the C-L as shown in Fig. 3 of ASTM E 399.

Figure 1. illustrates a typical shell course and excess material section and Figure 2. a base metal section for reactor surveillance programme (RSVP) test specimens, as well as test specimen orientations.

4.4 Adequacy and determination of exposure conditions

Important parameters for describing the exposure conditions are the following:

- temperature history i.e. it should be ensured that the temperature history during irradiation seen by the surveillance specimens should be representative of that seen by the corresponding RPV materials. The location of the specimens in the RPV and the thermohydraulic conditions at that location should be verified with respect to the effective prevailing temperature at that location. Also the influence of gamma heating in the specimens should be considered. Currently the thermal monitors in the surveillance programme only indicate the highest temperature the specimens have seen, with no knowledge of how long this highest temperature has been effective. Besides that the lower temperature exposure is actually more of concern.

- the fluence seen by the accelerated surveillance specimens especially for PWR’s are normally more than three times the fluence seen by the RPV material. The so-called fluence rate factor (lead factor) is therefore > 3. Present practice consider this fact acceptable because it is "assumed" to give conservative or similar results.

- the neutron spectrum seen by the accelerated surveillance specimens is also different from that seen by the RPV wall. Presently it seems possible to correct the error if the spectra at both locations are known. These could be obtained from the surveillance specimens and from "scratch specimens" (shavings of the cladding) taken from the RPV inner wall.

Therefore to ensure the representativeness of the irradiation conditions the above-mentioned points should be evaluated.

4.5 Neutron measuring methodology (and test results relationship)

The correct determination of the embrittlement as a function of the fluence for projection purposes is only possible if both the fluence calculations as well as the tests on the specimens are correctly done. Normally we can assume that for conventional tests such as tensile and Charpy V performed in the hot cells applying ASTM standards or their equivalents and using well qualified test machines and procedures as well as having effective quality assurance programmes will give correct results. However, for fracture mechanics tests
in the hot cells such as $K_{ic}$, $J_{ic}$, CTOD ($\delta$), and $K_{td}$ (using precracked Charpy-type specimen) tests, careful planning and qualification of test standards and procedures, as well as test machines have to be duly carried out to ensure well qualified and verifiable test results.

Current damage fluence determination for the RPV consists of:

**Neutron fluence calculations** coupled with **Neutron fluence measurements**

4.4.1 Neutron fluence calculations

Calculations of the RPV damage fluence in principle consist of the following steps:

- determination of the geometrical and material input data
- the determination of the core neutron source
- the propagation of the neutron fluence from the core to beyond the vessel
- the qualification of the calculational procedure.

Current fluence calculations cover the entire damage fluence spectrum (0.1 to 15 MeV) and normally use 2 or 3 D computer Codes to obtain the 3 D fluence distribution.

The commonly used unit for the fluence is still $n/m^2$, although recently more and more scientists are advocating the use of dpa (displacement per atom) to characterize the neutron embrittlement damage.

4.4.2 Neutron fluence measurements

Experimental dosimetry provides an alternate neutron fluence that may be used to confirm neutron transport calculations. Integral fission detectors placed in surveillance capsules and, more recently, in the ex-vessel cavity provide the information needed. The choice of the "threshold" detectors for this RPV dosimetry must be carefully evaluated to optimally fulfill their intended functions. Also the use of established, applicable ASTM standards or their equivalents should give adequate and reliable results.

For the fluence determination it is imperative to know the maximum fluence seen by the RPV wall and its exact location as well as the fluences at the surveillance capsules. Therefore normally 3 D neutron flux distributions are necessary and the codes used to determine these must be capable to take into account all the relevant parameters such as geometry, material density changes, capsule representation, required mesh fineness, and related factors of substance.

Also the fluence calculational uncertainties shall be duly verified.

5. DATA BASES AND THEIR APPLICATIONS

As a general rule integrity assessments of RPVs should be done based on a sufficient
amount of well qualified specific data. In the case of the embrittlement of materials caused by neutron irradiation and its influence on their fracture toughness, normally the only specific data available stems from the surveillance programme of the RPV in question. The fact that normally only a limited number of specimens can be included in the surveillance programmes has forced us to look for additional supporting data for integrity assessments of RPVs. Therefore data bases on material embrittlement have an important role to play in "broadening" the basis of available specific data for RPV integrity assessments.

To be useful data bases preferably consisting of surveillance programme results from various LWR-NPPs should at least fulfill the following requirements:

- the material is fully characterized (i.e. information on manufacturer, manufacturing process, product form, chemical composition including impurities, heat treatments etc).

- for weldments also the welding process, procedures, type and shape of the weldments.

- well documented information on tests performed and the standards and procedures used as well as the implemented quality assurance programme. The variabilities due to procedures and interlaboratory differences have to be determined in order to form a sound basis for factoring heat-to-heat variability and to assist in evaluating results from nonstandard tests.

- all available unirradiated as well as irradiated material properties including fracture toughnesses \( K_{ic}, K_{ld}, K_{is}, J_{lc} \) and \( d_l \) shall be given.

It has to be stressed that only well qualified data shall be included in the data base. Therefore so-called validity criteria must be established and followed to ensure the qualification of the data. Furthermore uncertainties inherent or otherwise shall be included in the evaluations of the data to provide a sound and rational basis for establishing statistically meaningful data.

A considerable amount of data has been generated in research programmes using research reactors. The results of these research programmes are invaluable for our understanding of the mechanisms of the embrittlement by neutron irradiation, as well as the dominating roles certain elements play and their interrelationship. Generic trends can be derived but the applicability of the results for a specific RPV still have to be evaluated with respect to the representativeness of the data for that specific RPV. Especially the very high fluence rate in research reactors as compared to RPV conditions is currently being discussed and investigated. The issue here is whether the fact accepted to date that the same fluence in a shorter time is more damaging, is still generally valid. In other words are accelerated tests results conservative compared to more realistic test results. Recent data seem to indicate this could well not be the case (fluence-rate effect). Also other aspects such as e.g. the neutron spectrum should also be duly considered.
5.1 Data base sources [9, 10, 11, 12]

The sources of data fall into two categories:

- RPV-Surveillance programme results (termed power reactor data) and,
- Research programme results using research reactors (termed test reactor data).

The main body of the data base will primarily consist of data of the first category, because these in general are more realistic respectively representative due to the boundary conditions (in NPP) under which they are obtained. Data of the second category are important for understanding the mechanism and the role specific alloying elements and impurities play, as well as to be used for anticipating possible embrittlement effects for the preventive maintenance aspects and also aid in material selection. The intended data base will primarily be used for improving the bases of safety assessments in particular to increase the confidence level. However, this is presently limited to LWR-RPV materials. Information required for data to qualify their adequacy for inclusion in data bases are given in the following subchapters.

5.2 Power reactor data

General information
- NPP-name, rated power and country
- date of construction and commissioning of the NPP
- LWR-type, vendor and manufacturer, as well as country of origin
- a short description of the RPV with the main specific features
- number of years in operation

Design information
- design pressure and temperature
- load cycle diagrams used and environmental conditions specified
- design code used and date of its issuance including addendas considered
- equipment specifications and data sheets

Data base information

Material identification and characterization (initial unirradiated condition)
- material type code
- product form i.e. forging, plate or weld
- heat number
- RPV manufacturer and the material supplier, fabrication process(es) used, as well as all the heat treatments the RPV material has seen.
- the specified, as well as the effective chemical composition of the material (including impurities) with the range of uncertainties in the latter.
- mechanical properties and fracture toughnesses determined, as well as the specimen types used.
- the orientation of the specimens tested i.e. longitudinal or transverse (L-T, T-L, L-C, C-L etc.)
- the removal location of the specimens with respect to the thickness i.e. 1/4T, 1/2T etc.
- the standards used to test the specimens and the degree of compliance with the criteria required for a valid test.
- methods which have been effectively used to determine the initial NDT- resp. RT_{NDT}-temperature.
Surveillance programme information.

- the requirements respectively the standard used to design the surveillance programme (e.g. 10 CFR 50 Appendix H, ASTM E 185, etc.).
- the number of capsules and number and type of specimens in each capsule for each material monitored.
- the placement of the capsules in the RPV with respect to the radial, axial and azimuthal location (lead factors).
- the expected and effective fluence rate (lead factor) the capsules experienced.
- neutron fluence calculation method (1D, 2D, 3D or others) and neutron fluence measurements (dosimetry etc.) used.
- the irradiation temperature and the temperature monitors and methods used for its determination.
- the number of available Charpy-specimens (test results) and method used to construct the transition temperature curve e.g. "eye ball", sigmoidal (tanh.) or polynomial curve fit etc.
- determined RTNDT-shift resp. RTNDT-temperature should be reported with an uncertainty tolerance.
- the number of capsules withdrawn and the date(s) of withdrawal(s)
- the accumulated fluence at the highest irradiated location of the RPV and the maximum RTNDT-shift determined.
- DRTNDT = f (Fluence) - curve

Qualification and verification/validation of the data

Availability of documentations pertaining to:

- material identification and characterization of the initial unirradiated condition
- quality assurance audits and reports
- identification of the laboratory performing the test and tests reports on the irradiated condition including QA performed.
- updates of reports and additional tests performed in the framework of the surveillance programme or related activities.

Confidence level, e.g. "very high", "high", "medium", "low", or "very low" of the data source based on the following:

- age of the documentation
- changes identified in superseding documentation
- the standards or test techniques used to generate the data and whether such standards have been superseded
- the pedigree of material source (whether obtained from original archive material or reconstituted)
- the history of updates or errata associated with the original source data
- whether the data source documentation was QA'ed or independently verified
- the origin or publication type of the source documentation
- the amount of testing performed and reported in the source document
Adequacy of the data source based on the completeness, verifiability and accuracy of the information contained in the documentation.

Reporting format

The standard format for data collection and reporting are given in tables 1 - 5 as proposed by Griesbach (EPRI) [12] and supplemented by the additional information required in the previous chapters.

5.3 Test reactor data

General information

- Research reactor name and type, rated power and country
- Flux level applied, fluence rate to typical LWR flux level
- Irradiation temperature control possibility
- Neutron spectrum tailoring availability

Data base information

Appropriate information similar to those of power reactor data is required on material identification and characterization, test reactor capsules and test standards/procedures, qualification and verification/validation, as well as reporting format.

6. CURRENT ISSUES RELATED TO THE ASSESSMENT OF IRRADIATION EMBRITTLEMENT OF RPV

With respect to the state-of-the-art, research results in the last few years indicate the necessity to reevaluate the adequacy of the bases used in present assessment of embrittlement by neutron irradiation as described in sections 3, 4 and 5.

The most relevant issues are the following:

- Fluence rate (lead factor) effect

Presently the perception prevails that for RPV materials, the same fluence attained by neutron irradiation in a shorter time period is more damaging than if this is attained in a longer time period, i.e. research reactors results and surveillance programmes results showing fluence-rate factors (lead factors) > 1 will be conservative. Results from investigations on trepans from the decommissioned Gundremmingen RPV [13, 14] and from the same materials in research reactors seem to indicate the existence of a fluence rate (formerly called lead factor or flux factor) effect that seems to contradict this assumption. It has to be noted, however, that the effect of flux on embrittlement seems also to be strongly dependent on other irradiation variables, most notably fluence level, temperature and material chemistry. Since this fluence rate effect observed in the Gundremmingen investigations has only manifested in the weak orientation of the material and not in the strong orientation and presently also further research is still ongoing, controversial opinions as to the
real existence of this fluence rate effect that could have major impact on present practice prevails.

From a safety point of view, however, it is imperative that this controversial phenomenon be incontestibly clarified as soon as possible. This is important mainly because of the following reasons:

- presently it is considered to be a fact that for a given fluence the shorter the irradiation time the more damaging the embrittlement effect will be. Therefore accelerated specimens for the surveillance program are acceptable because they are considered to give conservative results. If the Gundremmingen results, which show the opposite effect proved to be correct then the basic concept of present assessment practice of irradiation embrittlement is questionable and must be critically reassessed, possibly has to be abandoned and eventually a new concept be put into its place.
- research efforts must be set up to find methods to resolve the issue of possible nonconservatism of accelerated surveillance programme results, as well as the existing data base.
- new integrity assessments must be performed on all RPVs which have used accelerated data base for integrity assessments in their safety cases.
- the ASTM E 185 standard [1] must be revised to reflect the new state-of-the-art. But even now before this issue is resolved it seems warranted to have surveillance programmes capsules located at the RPV innerwall with fluence rate factor of about 1. In this way the issue can be resolved for future RPVs. An even better solution would be if both accelerated and vessel wall capsules are applied. In this way fluence projection for operating purposes can be obtained with accelerated capsules and this projection can be checked with the vessel wall capsules (fluence rate near 1) results at a later date.
- Fluence "threshold".

The presently still accepted fact that only when the expected fluence is greater than $10^{21}$n.m$^{-2}$ is of relevance i.e. causes relevant embrittlement of the material and therefore needs to be monitored seems not to be correct. At low irradiation temperatures, much lower fluence has already been shown to have caused relevant damage. The temperature especially the lower temperature during irradiation has been identified to be one of the dominant factor. In some plants with RPV-support columns made of irradiation sensitive materials and traversing the core belt region at a short distance from the RPV and seeing a significant lower temperature could be affected. The failure of these support structures will have direct impact to the safety of the RPV.

In this context it has also been suggested that possibly the neutrons with energy between 0.1 and 0.5 MeV could also play the primary role.

- The analytical assessment procedure.
- The "assumptions" on which the procedure for the determination of the $RT_{NDT-stiff}$ and fracture toughness in section 3 is based, have not all been
verified yet to be justified as either correct or conservative. Recent results from Finland [15, 16] seem to indicate that the fourth "assumption" (same shift of the Charpy transition curve and the fracture toughness $K_{IC}$-curve respectively the $J$-curve after neutron irradiation) is not conservative i.e. is too optimistic. On the other hand ORNL studies seem to indicate that there is good agreement between the shift of the $K_{IC}$-curve and the 30 ft.lb shift. It is however not clear whether the type of materials that have been studied can represent the RPV materials used worldwide. Thus this is still a controversial issue that should be clarified also because of the possible influence of other factors such as material type and its chemical composition, material structure etc.

- The shift determination only based at the 41 J energy level without confirmation that at the new (shifted) $RT_{NDT} + 60 \, ^\circ F$, Charpy V energy level of \ 50 ft.lb and 35 mils LE are attained does not comply to the definition of $RT_{NDT}$ and is probably nonconservative. This is because after irradiation the upper shelf energy (USE) is also reduced and the Charpy V transition curve tends to be less steep (more flat).

- For cases where the USE fall below the 50 ft.lb level, 10 CFR Part 50, § 50.60 and Appendix G "Fracture Toughness Requirements" apply. There, as alternatives, full detailed fracture mechanics analyses using appropriate specific fracture toughnesses or annealing of the RPV which can demonstrate compliance with the requirements are considered acceptable.

- For LWR-RPVs of the first and second generations there are the following additional "issues":
  - the initial NDT temperature determinations were not all done according the ASTM 208 standard [3]. Some of them were "conservatively" determined by a so-called "confirmation tests" using two Pellini specimens. And some used only Charpy V-notch specimens to "conservatively" determine the NDT temperature by correlation. At that time these practices were considered conservative, but recent knowledge (see also the following point below) and the introduction of the $RT_{NDT}$ concept strongly question the "conservatism" respectively adequacy of these practices.
  - the ASTM E 208 standard [3] used before 1984 does not comply to the present valid edition defining the requirements for the preparation of the crack starter weld.

The practice used up to the 1981 edition is by depositing the crack starter weld from both ends and ending in the middle where the notch is located. Investigations from Japan [17] and Germany [18] have shown that this old practice can result in too optimistic a value of the NDT temperature by as much as 25 \, ^\circ C. To avoid this shortcoming, since the 1984 edition of the ASTM E 208 standard the crack starter weld has to be deposited in one pass without interruption.

The issue now is how can we reasonably take into account this possible nonconservatism in initial NDT temperature determination in our safety assessment?

- many of the first and second generation RPV surveillance programmes only contain longitudinal (strong orientation) Charpy V-notch specimens to
determine the RT<sub>NDT</sub> shift instead of the presently required transverse (weak orientation) specimens according to the ASME code. Since 1972 these transverse specimens per definition are part of the RT<sub>NDT</sub> concept.

Besides the above mentioned items many of the first and second generation RPV surveillance programmes contain very few (about 9 to 12) specimens to construct the transition temperature curve. If we proceed according to the Code by testing 3 specimens for each temperature, we will only have 3 to 4 points for the curve (if all goes well)! In addition to that, the large scatter normally observed in the results makes the task even more difficult. This is certainly an unsatisfactory situation and makes a reliable and adequate assessment very difficult to achieve. Therefore ways and means have to be found to improve this situation. The so-called "reconstituted" specimens could be of great value here.

The USNRC has addressed some of the above mentioned issues in their Branch Technical Position of the Materials Engineering Branch (MTEB) - No. 5-2 "Fracture Toughness Requirements". However this BTP-MTEB is given for the following material types manufactured mostly in the US and according to ASME specifications:

- SA-533 Grade B, Class I, Plate and
- SA-508, Class II, Forging

Therefore for similar materials not produced in the US and/or not complying to the corresponding ASME specifications, the applicability of this BTP-MTEB should first be evaluated.

However, the BTP-MTEB give a method for converting longitudinal values of the USE to transverse values (using a factor of 0.65) but does not give rules as to what the corresponding temperature shift (conversion) must be. That is the corresponding temperature at which the transverse USE is attained. This can have significant consequences for the RT<sub>NDT</sub> shift determination whether at the 68 J energy level or 41 J energy level. This issue also has to be clarified.

The construction of the transition curves through the Charpy V energy values of the initial unirradiated conditions and of the surveillance programme specimens, after irradiation, to obtain reliable and reproducible RT<sub>NDT</sub> shift determination is still a matter of debate. There is presently no generally agreed upon method or standard to do this. Some use the so-called "eye ball" method and others use some analytical mathematical expressions to trace the transition curves e.g. sigmoidal (tanh) curve fits, polynomial curve fits etc.. The problem mainly lies in the limited number of data points available and the large scatter observed in the data as has already been mentioned.

The use of analytical mathematical expression can avoid subjectivity and can be expected to give a better reproducible basis for the transition curves. On the other hand one can ask whether the mathematical expression can really always represent the real material behaviour influenced by many parameters. And by using this analytical expression one does in a way "force" the material to behave as prescribed. In so
doing one can actually miss an anomalous behaviour that could be very important, which would probably not have been missed by using the "eye ball" method and realising that the data in question are not outliers.

A logical approach would be to combine both methods in an appropriate way. Some would even propose to rely on the so-called "engineering judgement". Experience unfortunately shows that this term has often been misused, is subjective and should thus only be used with due reservation.

Therefore it is very desirable if this issue could also be resolved to ensure reliable and reproducible as well as comparable results. This aspect should also be considered in selecting the data to be used in a data base.

- Irradiation assisted stress corrosion cracking (IASCC)

It should be noted that material embrittlement by neutron irradiation is but one of the many phenomena in material degradation processes that have to be taken into account in the safety assessment of the RPV. Possible synergistic effects of all the relevant phenomena have not as yet been systematically and comprehensively investigated in research programmes. However, for many years one of those possible synergistic effects i.e. IASCC has already received appropriate attention.

Stress corrosion cracking (SCC) has long been recognized to be a relevant material degradation process for austenitic stainless steels but recently also for ferritic low alloy steels. The question in what way and how damaging the neutron irradiation effect can have on the material behaviour with respect to SCC is presently still under investigations. An international group called International Cooperative Group on Irradiation Assisted Stress Corrosion Cracking (ICG-IASCC) exists since 1986 to systematically study this phenomenon. Here primarily the internals and core support structures are affected but also RPV-materials at the core beltline region such as cladding, pads welded to the vessel wall and the vessel wall proper.

7. CONCLUDING REMARKS

The neutron irradiation embrittlement, i.e. the reduction of the fracture toughness of the RPV materials due to neutron irradiation has to be taken into account in the integrity assessment of the RPV. This assessment is usually performed periodically during the NPP's life, i.e. at the beginning of operation and at certain intervals in the life of the plant, in particular after a surveillance programme capsule has been withdrawn or due to other causes, which require new assessments to be done. Depending on the so-called fluence rate or lead factor of the surveillance programme capsules, for assuring the integrity for the next operating interval (before the next periodical assessment) one has to either interpolate (e.g. for PWRs) or extrapolate (e.g. for BWRs) the rate of neutron irradiation embrittlement. The commonly used approach of the RT_{NDT} concept and its shift determination are based on several assumptions and still have shortcomings, also with respect to the adequacy of understanding the factors influencing the embrittlement mechanism as have been described in the previous chapters. Therefore, one should be aware of these and is
cautioned to consider them appropriately when taking neutron irradiation embrittlement into account in the integrity assessment of the RPV.

Acknowledgments

Thanks are due to Tim Griesbach (EPRI) and William Server (ATI) for reviewing the first draft and the material/reference they have provided and used in particular in chapter 5, and Lendell E. Steele for his review and comments of the first draft.
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