

MATERIALS IRRADIATION SUBPANEL REPORT

to

BESAC NEUTRON SOURCES and RESEARCH PANEL

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PREFACE

Dr. W. Happer, Director, Office of Energy Research of DOE, in a letter dated June 1, 1992 and addressed to Professor L. Silver, Chair of the Basic Energy Sciences Advisory Committee (BESAC), requested formation of a panel to report on key issues concerning possible new neutron sources, emphasizing especially the comparison of steady-state reactors and pulsed spallation sources.

In conjunction with the duly-constituted Panel's meeting in Oak Brook, Illinois, a broad Review of Neutron Sources and Applications was organized. Chairs of the Review Subpanels presented their findings and recommendations to the Panel on September 10, 1992. These presentations will form a companion Proceedings to the Panel Report.

This informal document is the report of the Materials Irradiation Subpanel to the BESAC Panel.

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SUMMARY

The future success of the nuclear power option in the US (fission and fusion) depends critically upon the continued existence of a healthy national materials-irradiation program. Consideration of the requirements for acceptable materials-irradiation systems in a new neutron source has led the subcommittee to identify an advanced steady-state reactor (ANS) as a better choice than a spallation neutron source. However, the subcommittee also hastens to point out that the ANS cannot stand alone as the nation's sole high-flux mixed-spectrum neutron irradiation source in the next century. It must be incorporated in a broader program that includes other currently existing neutron irradiation facilities. Upgrading and continuing support for these facilities must be planned. In particular, serious consideration should be given to converting the HFIR into a dedicated materials test reactor, and long-term support for several university reactors should be established.

I. Introduction

Our objectives in this report are to: 1) describe the broad range of neutron irradiation facilities required to support the Department of Energy's mission to sustain and develop the nuclear energy option and 2) to specifically address the potential roles of high-flux mixed-spectrum reactor and spallation-based neutron sources.

Understanding and optimizing the behavior of materials in neutron radiation environments is a preeminent concern to all nuclear energy technologies ranging from the current generation of fission to future fusion reactors. Ultimately, the safety, economics and, indeed, the basic viability of nuclear energy systems depend on reliable materials performance. The effects of neutron radiation are manifested in diverse, profound and, most often, deleterious ways: gross swelling and other dimensional instabilities, severe reductions in ductility, fracture resistance and creep strength, and increased susceptibility to chemical attack. Significant irradiation damage is experienced by essentially all materials over a wide range of neutron fluxes, fluences, temperatures, and stresses.

II. Response of Materials to Neutron Irradiation

Property changes are controlled by the combination of a large number of environmental and material variables. Hence, predicting and improving the performance of materials in irradiation environments is far from a matter of "testing;" rather, success in meeting these objectives must proceed from a solid, science-based understanding of the fundamental underlying processes. Although they are manifested in different ways, these fundamental processes are generic to all forms of irradiation damage and include:

Production of point defects and small defect aggregates in displacement cascades and transmutants constituting the primary source of damage.

Microstructural and microchemical evolution resulting from long-range diffusion and interactions of the mobile primary defects in the form of various extended defect clusters, regions of non-equilibrium solute segregation, enhanced and induced precipitation and modification of the pre-existing microstructures and precipitate phases.

Microstructurally and microchemically sensitive physical, chemical and mechanical properties changes.

The long-term objective of radiation damage research is to evaluate these fundamental mechanisms based on combinations of theory and experiment and to incorporate them into quantitative predictive models of irradiation damage. Materials-irradiation facilities are needed to develop an understanding of the underlying mechanisms and to validate and calibrate the integrated materials performance models. It might also be noted here, that this fundamental research on defect physics, microstructural and microchemical evolution and the micromechanics of deformation and fracture has impacted, and will continue to impact, the broad field of materials science.

III. Scientific and Technological Needs and Opportunities

In this section we briefly outline some of the significant technological motivations for irradiation damage research; we next provide examples of some key scientific opportunities; and in the following section we describe the irradiation facilities needed to support this research endeavor.

A. Technological motivations

1. Ensure the continued safe and reliable operation of existing light water reactors including, when possible, extending their operating life.
2. Support the development of advanced thermal, fast breeder, space and defense fission reactors.
3. Support the development of fusion reactors with particular attention to low activation materials.
4. Support the development of accelerator-based neutron sources for tritium production, waste transmutation, and scientific applications.

B. Scientific Needs and Opportunities

It is necessary for the nation to maintain a group of neutron sources that provides the diverse neutron spectra required for different reactor environments such as LWR, breeder or fusion reactors. While an understanding of effects peculiar to a given technology are required, it is also necessary to maintain a strong research program on the fundamental processes described in Section II. Such a general information base will allow connection of results to various applications.

The next generation of neutron sources should provide a range of neutron fluxes and controlled irradiation environments so that it is possible to address and resolve the following radiation-effects phenomena and questions:

1. A detailed description of the number and configuration of defects generated by the primary displacement cascade. These parameters include both stable defect complexes and freely-migrating defects, and should be determined as functions of neutron spectrum, neutron flux, neutron fluence (time-integrated flux), temperature, material type, and existing defect microstructure. The stability of defect structures should be determined as a function of time after their production for different temperatures and microstructure states.
2. Evolution of defect-induced microstructure and microchemistry is a multi-parameter problem. The interaction of migrating defects and transmutation products with each other and with a developing microstructure should be determined. It is important that modelling and experiment interact strongly to allow extrapolation to new situations.

3. Investigation of nonequilibrium phenomenon induced by radiation damage such as metastable phase formation, segregation and amorphization.

4. Investigation of the role of electronic excitation in defect production and microstructure and microchemistry evolution in nonmetals.

5. Understanding of dose-rate effects in microstructure and microchemistry evolution.

6. Connection of property changes to microstructure and microchemistry evolution with the aid of detailed modelling.

7. Interaction between a material and its surrounding chemical environment.

8. Material property changes caused by irradiation-induced alterations in internal interfaces.

IV. Existing Facilities for Controlled Neutron Irradiations and Potentials for Upgrade of Facilities

A. Existing Facilities

The broad definition of radiation damage is the study of the effects of radiation on the structure and properties of materials. This review report is concerned only with neutron radiation damage. With the broad definition of the discipline, any reasonably intense source of neutrons represents a potential resource. This review process was prompted by the need to evaluate two particular types of neutron sources, the ANS, and a spallation neutron source. To complete this evaluation, it is important to understand how these sources fit into the array of currently available neutron irradiation facilities. A brief summary of neutron irradiation facilities currently available in the US has been compiled to provide perspective for these discussions. Historically, both nuclear reactors and accelerator-based charged-particle sources have been employed in the study of irradiation damage.

1. Reactor-Based Facilities

The vast majority of neutron irradiation damage studies have been based on materials irradiations in nuclear reactors. The committee believes that this trend will continue through the foreseeable future. Nuclear reactors offer several inherent advantages:

- a. They allow large irradiation volumes.
- b. They have the potential for reaching high neutron fluences.
- c. Their radiation environment is generally well characterized.

Different reactor designs offer a variety of irradiation environments that can be characterized in terms of their neutron spectrum and the maximum available fast flux (generally defined as $E > 0.1$ MeV). No single reactor irradiation facility can meet all of the needs of the radiation damage community. Comparisons of damage states produced in

various reactor spectra are required to study the processes of radiation damage. Radiation damage poses many significant technological concerns for existing light water reactors and advanced reactor designs (including fast breeder and fusion reactors). The study of these problems requires neutron environments that simulate actual application environments. These considerations help to define the need for reactor irradiation facilities.

A summary of reactors currently available for controlled materials irradiations is included in Table I. For the purposes of this discussion, these reactors have been divided into four basic categories. University research reactors provide a valuable resource for the study of radiation damage in light water reactor pressure vessels. (The committee is aware of four reactors that could be used for this purpose.) Higher flux test reactors, such as the ATR, have provided important sources of information for defense-related reactors and have significant potential for the simulation of LWR core environments. The High Flux Isotope Reactor at Oak Ridge has provided a rich source of data on materials with high He to dpa ratios, which has been useful in the fusion program. HFIR irradiations have also aided in the understanding of material behavior for LWR applications. The breeder reactor test facilities (EBRII and FFTF) are required for breeder reactor alloy development programs.

Table I.

Type	Spectrum	Peak Fast Flux (E > 0.1 MeV)	Technologies Supported
University Research (4 possible)	Mixed	< 10 ¹³	LWR Pressure Vessel
Test Reactor (ATR) (Nat'l Labs, e.g. HFBR)	Mixed	> 10 ¹⁴	Defense, Potential for LWR Core
High Flux (HFIR)	Mixed	> 10 ¹⁵	Fusion, LWR
Fast Breeder (FFTF, EBRII)	Fast	> 10 ¹⁵	Fast Breeder

2. Accelerator-Based Neutron Sources

Spallation neutron sources produce neutrons as a result of the destruction of nuclei within a heavy metal target by high-energy protons. The neutron generation increases with increasing proton energy, and beams with energies over 450 MeV are typically used with the beam delivered in a pulsed mode. The neutrons are produced with the same time structure as the incident protons. Such neutron sources are characterized by neutron spectra resembling a degraded fission spectrum with an additional component of very high energy neutrons. These high energies extend up to the incident proton energy. Neutrons are generated within a fairly small volume determined by proton stopping so that the resulting fluxes have strong spatial gradients. Two spallation neutron sources are operating in the US.

ANL/IPNS: In its original configuration, the Intense Pulse Neutron Source (IPNS) had separate targets dedicated to radiation effects and neutron scattering. The Radiation Effects Facility was closed in 1984. Originally, the IPNS Neutron Scattering Facility had a depleted uranium target. This was replaced with highly-enriched uranium resulting in a factor of 2.5 increase in neutron generation. Protons are delivered to the target in 100 nsec pulses at variable repetition rates up to 30 Hz and time average currents of 14 to 16 μ amps. The dedicated Radiation Effects Facility at IPNS had a time average fast neutron flux of 2.8×10^{12} n/cm²s for 14 μ amp of 450 MeV protons. With an enriched uranium target, the flux would be 7×10^{12} n/cm²s.

The irradiation thimble currently in use within the Neutron Scattering Target Assembly is not in an optimized position and has a fast neutron flux of 2×10^{11} n/cm²s with the depleted uranium target. An enriched uranium target would increase the flux to 5×10^{11} n/cm²s.

LANL/LAMPF, LASREF: The Los Alamos Spallation Radiation Effects Facility is located at the beam stop of the Los Alamos Meson Physics Facility. It operates with a time average proton beam of 800 μ amps at 800 MeV. Neutron generation at LASREF depends on the composition of isotope production target and proton irradiation experiments located in front of the copper beam stop. The time-average fast-neutron flux is about 3×10^{13} n/cm²s for 1 ma of 800 MeV protons.

B. Potential Upgrade of Existing Facilities

1. Reactors

The committee is extremely concerned about the erosion in the number of reactor irradiation facilities. We believe that the reactors listed in Table I represent a minimum acceptable level of research activity. In the near term, we recommend that all of these reactors be maintained as irradiation facilities. Further development of the irradiation facilities at these reactors would provide the foundation for a comprehensive radiation damage program.

2. Accelerator-Based Sources

A proposed upgrade of IPNS would utilize a proton energy of 1500 MeV and a proton current of 500 μ amps. This would result in an estimated fast neutron flux of 2.5×10^{14} n/cm²s in an optimized facility used by radiation effects and neutron scattering.

V. Strengths, Weaknesses, and Complementarity of Reactor v. Spallation Neutron Sources

In order to address the relative merits of proposed new sources with respect to their ability to support meaningful materials-irradiation facilities, it is necessary first to define the desired characteristics of such facilities. In general, they can be separated into two classes, those that establish the quality of the facility, and those that are imposed upon the facility by special requirements associated with specific technological needs.

A. Characteristics that Define Facility Quality

The first category consists of those attributes that any dedicated, modern materials-irradiation facility must possess. Among the principle entries on this list, one stands alone as the single most important: the facility must provide a capability to perform controlled, single-variable experiments. An irradiation space that merely permits high levels of sample exposure to neutrons under conditions of uncontrolled neutron flux, spectrum, and experimentally-relevant variables is unacceptable. This imposes upon the facility a set of characteristics/requirements that generically define its quality:

1. First and foremost, the radiation environment experienced by the sample(s) must be well characterized by an adequate, ongoing, documented dosimetry program.
2. Control of the experimental parameters implies a capability to perform single-variable experiments. Temperature measurement and control represent the most important of these capabilities; in broader terms, the facility must be able to support *in situ*, real-time electrical measurements of properties such as electrical resistivity and others that also generate electrical signals (see #6 below).
3. Realistic consideration of the scope of a meaningful irradiation effects program has established the necessity for a large experimental volume in the appropriate-flux region of the source. A typical volume would be 5-20 liters.
4. Access to sample capsules for removal and replacement at arbitrary times would expand the range of feasible experiments.
5. Flux gradients over the sample volume must be minimized in order to limit complications encountered in the subsequent interpretation of the experimental data. Gradients that exceed prescribed values or fluctuate grossly with time are unacceptable.

6. As temperature control is always required, excessive nuclear (gamma) heating cannot be accommodated in an experimental plan. As a general stipulation, the lower the gamma heating rate, the better, although some gamma heating can be helpful.

7. Many of the required irradiations will be of long duration. Interpretation of the data will be simplified if the neutron spectrum remains constant during the radiation exposure. It is difficult to conceive of a realistic program for monitoring spectrum changes that occur frequently. Even those that are documented on a periodic basis over two to three years would introduce serious difficulties in data analysis, and could jeopardize the objectives of the experiments.

B. Characteristics Required by Specific Technology Questions

The second category of facility characteristics consists of those most closely associated with special technological demands. While such a list cannot be exhaustive, it is likely to include at a minimum those discussed below.

1. The necessity to reach significant total exposures (fluences) in reasonable times implies a high rate of atomic displacements. The fusion and breeder reactor technologies each require this feature. Significant microstructural and microchemical changes do not develop or are undetectable at low fluences.

2. Fusion and accelerator materials development requires irradiations that ensure the appropriate high ratios of He to dpa. These ratios must be attainable in the facility.

3. For reasons related once again to the fusion program, it must be possible to supply high-energy neutrons (14 MeV). This capability would span the range of interest that the materials-irradiation community is likely to require for the foreseeable future.

4. An ability to match required spectra including methods for spectral tailoring has become a desirable feature for future experiments. It enhances the utility of a given source, and permits the pursuit of certain experiments that otherwise could not be conducted.

5. Recent experience with light water reactor technology has led to the realization that a relatively low-dose-rate facility is needed in addition to facilities already mentioned.

6. Certain components in fusion reactors will be exposed to neutrons while operating at cryogenic temperatures, viz., superconducting magnets. Proper investigation of the consequences of these exposure conditions requires a cryogenic-temperature irradiation facility of modest flux. Such a facility also would serve as a welcome adjunct to a program in basic radiation effects whereby primary radiation-induced defects can be immobilized and subjected to subsequent thermal annealing studies.

C. Summary

This analysis of the characteristics of an "ideal" materials-irradiation facility made it possible to evaluate each type of source, recognizing that some present design uncertainties have not been resolved.

It should be apparent that no single neutron source will be capable of meeting all of the foregoing criteria. Thus, it is already clear that the only viable solution to a national strategy for maintaining a healthy materials-irradiation program is one that embodies more than one source. It is stated here and will be reiterated later, that it is in the national interest to maintain at least some of the remaining irradiation capabilities still available at universities, industrial sites, and National Laboratories.

VI. Conclusions and Recommendations

A. Conclusions

The conclusions drawn by this committee are based in part upon operating experiences drawn from existing reactors and spallation neutron sources, and upon the characteristics of the proposed Advanced Neutron Source (ORNL) and of a spallation source capable of providing a 1-2 MW proton beam on target. A detailed design of the spallation source was not provided; the assumption was made that the proton energy would lie between 800 and 1600 MeV, and that the neutron yield increases linearly with proton energy.

In addition to the assumptions of a technical nature outlined above, the committee also assumed that the US will continue to depend upon nuclear power systems far into the next century. Nuclear power for the purposes of this discussion includes fission and fusion technology, and applies to applications in space and for defense purposes as well as for meeting conventional civilian power demands.

Advanced concepts associated with the fission and fusion power options will require a broad materials-irradiation program that must be based upon the availability of diverse neutron sources. It is clear that an Advanced Neutron Source whose primary purpose is to support research with thermal neutron beams will not suffice as the sole high-flux source for national materials-irradiation program needs. Table I of this subcommittee report lists currently available options for additional neutron sources in this country. Some of these sources are scheduled for shutdown or are already shutdown in a standby mode.

Finally, these facts together with those stated in Section V have led to several conclusions:

1. The relatively low time-averaged fast flux and limited irradiation volume of a spallation neutron source are major negative factors.

2. Conclusion 1 leads to the choice of a steady-state reactor option such as the proposed Advanced Neutron Source, but with reservations.

3. Reservations concerning the ANS can be addressed by further design considerations in order to establish more firmly its utility to a comprehensive materials-irradiation program. These design considerations would affect core and reflector regions of the reactor.

4. Regardless of the final configurations of ANS irradiation facilities, it should be reiterated that the ANS will not be adequate as the sole future materials-irradiation facility for the nation.

5. Although a higher priority is assigned to the construction of the ANS, materials-irradiation experiments of limited scope could be performed in a high-flux spallation neutron source.

B. Recommendations

1. Move ahead with the construction of the ANS taking additional time to optimize its design and operating parameters as best as possible for the many research communities it will serve.

2. Develop a strategy for maintaining or, if possible, upgrading some number of the nation's existing irradiation facilities (see Table I) for long-term use.

3. Consider funding a design study for converting the HFIR into a dedicated materials test reactor. This would involve replacement of its core but not its pressure vessel. A rough estimate of the cost of this conversion is 150 million FY 92 dollars.

4. Include plans for materials-irradiation facilities in any strategy for constructing a new spallation neutron source.