



**ADVANCES IN NUCLEAR FUEL CYCLE
MATERIALS AND CONCEPTS**

A. A. EL SAYED

*Materials Division, Nuclear Research Centre,
Atomic Energy Authority
Cairo, Egypt*

ABSTRACT

This presentation gives an overview of some new trends in the materials used in various steps of the nuclear fuel cycle. This will cover fuel and cladding materials, control rod materials, reactor structural materials as well as those used in the back end of the fuel cycle.

Problems associated with corrosion of fuel cladding materials as well as these in control rod materials (B_4C swelling ...etc.) and approaches for combating these influences are reviewed.

For the case of reactor pressure vessel steels issues related to the influences of alloy composition, design approaches including the use of more forged parts and minimizing, as far as possible, longitudinal welds especially in the central region, are discussed. Furthermore the application of techniques for the recovery of pre-irradiation mechanical properties of PVS components is also covered.

New candidate materials for the construction of high-level waste containers including modified types of stainless steels (high Ni and high Mo), nickel-base alloys and titanium alloys shall also be detailed.

Finally nuclear fuel cycle concepts involving plutonium recycling and actinides recycling shall be reviewed

I - INTRODUCTION

- ***A Focus on Some Developments in Fuel Cycle Materials and Concepts.***

- ***Developments Aiming at Improving***

- * Reliability
- * Economic viability
- * Safety
- * Minimizing environmental and health impacts

Of The Fuel Cycle Activities

- ***The Role of***

- * Increasing accumulated stock of NPP plutonium
- * Weapons plutonium
- * Long-lived actinides

- ***Main Topics Addressed are Advances in :***

- * Fuel and cladding materials
- * Control rod materials
- * Reactor structural materials
- * Materials for high-level waste containers
- * Some new fuel cycle concepts

2. FUEL AND CLADDING MATERIALS

2.1 FUEL

- ***Present World Nuclear Electricity Generating Capacity***

- * 354200 MWe From About 425 Power Reactors

■ *New Reactors*⁽¹⁾

- * Advanced PWRs, BWRs, PHWRs, FBRs, HTGRs.
e.g. AP- 600, SBWR, SIR, PIUS, ISER)
- * Major trend : MOX fuel
 - R & D for MOX to reach burn up of U oxides
 - New LWR designs using 100% MOX

■ *Plutonium and FBR Development*⁽²⁻⁸⁾

- * FBRs in the world⁽²⁾
France , Russia, Britain, Japan & Germany (under construction)
- * Western FBR Policy : and the Shutdown of first prototype
FBR in Britain , end of March 1994.⁽³⁾
- * The FBR strategy in Japan⁽⁴⁻⁶⁾
 - Nuclear electricity in Japan : 40,000 MWe (20%
of installed, 30% of generated capacity)
 - The monju FBR a prototype advanced thermal reactor
(ATR),280 MWe, 5.9 ton MOX, including 1 ton Pu
(operational 1995).
 - Commercial FBR by 2030

■ *World Plutonium Production: Present & Predicted Inventory*

- * (Figs. 1-3)

■ *HTGRs Fuel*⁽⁹⁾

- * First Developed and Demonstrated in the USA
- * Germany
 - AVR, Juelich, 20 MW (operated for 20 years, shutdown
1988)

- Scaled up to 300 MWe (THTR 300, Hamm - Uentrop, operated 16 Nov. 1985).

- Pebble bed system fuel: (Figs. 4,5)

- Fuel elements: Encapsulated ceramic spheres, 6 cm diameter (about 1.2 million, 80% of which are fuel, 20% graphite). Each fuel pin contains low enriched uranium in the form of coated particles = 0.9 mm diameter, two PyC layers and one SiC layer.

■ ***Metal - Alloy Fuel : (Fig. 6)***

- * 90 U - 10 Zr, anticipated to go to about 45 MWd/kg

2.2 CLADDING MATERIALS⁽¹¹⁻¹⁴⁾

■ ***Zircalloys and Zr-Nb Alloys***

■ ***Zircaloy-4 Vs Zr-Nb***

- * Corrosion , hydriding and irradiation effects (Table 1 and Figs 7,8)
- * H₂ content can go up to 600 ppm for a 90 μm thick oxide (20% of H₂ produced by corrosion).
- * Recent french experience on the effect of Sn, (Fig. 9)
low Sn (1.29-1.32%)
high Sn (1.40-1.47%)
- * Russian experience on corrosion and hydriding of Zr-1Nb alloy⁽¹³⁾

■ **Need for a Better Clad to Cope With:**

- * High burn up
- * High temperature
- * Improved operational flexibility

■ ***This Requires :***

- * A Zircaloy-4 with
 - Low Sn (1.29-1.32%)
 - Improved alloy homogeneity
 - Good surface finish
- * Going for new duplex alloys

■ ***Cladding & Structural Materials Performance in Relation to Reactor Water Chemistry*** ⁽¹⁴⁾

- * Effect of Li content on Zr O₂
- * Addition of H₂ (220-2000 ppb) to prevent SCC of primary circuit components in BWRs.
- * Addition of Zn (5- 15 ppb) to improve the fracture characteristics of sensitized 304 ss (reduces crack growth by a factor of 5)

■ **Avoid the Use of Cobalt - Base Alloys in Structural Materials to Reduce Man- Sievert.**

3 - CONTROL ROD MATERIALS ASSEMBLIES⁽¹⁵⁾

■ ***Materials***

- * Absorber materials : (mainly B₄C, Ag-In-Cd)
- * Cladding & structural materials (mainly 304 ss)

■ ***Main Problems***

- * Absorber materials :
Swelling (n-α reaction) helium leads to absorber -clad interaction

- * **Cladding & structural materials**
 - Mechanical mechanisms (fretting & wear)
 - Corrosion mechanisms (oxidation & hydriding)
 - Radiation damage mechanisms (enhanced creep & growth)
 - Irradiation assisted stress corrosion cracking (IASCC) involving synergism between two or more mechanisms

■ ***Improvement of Behavior Through***

- * Better materials
- * Better design
- * Clad wear resistant coating
 - Chromium carbide coating
 - Ion nitriding
- * Better control over microstructure
 - Composition of materials at the fabrication stage

■ ***Absorber Materials Changes***

- * Use of Hf instead of Ag-In-Cd
- * Use of compounds based on enriched boron and Hf for reactors using MOX fuel
- * Use of Dy_2O_3 - TiO_2

More R&D is needed : especially on hydrogen pick up by hafnium

■ ***Cladding & Structural Materials Changes***

- * To move from 304 to 304 L stainless steel of better resistance to IASCC

4. REACTOR STRUCTURAL MATERIALS⁽¹⁶⁻²²⁾

■ *Environmental and Operating Conditions*

- * Temperature and cycling
- * Pressure and cycling
- * Neutron flux

■ *Consequently: Irradiation-Assisted Degradation:*

- * Corrosion , hydriding
- * Mechanical

Other combinations of effects

■ *Materials : (16,17)*

High toughness low alloy steels, generally clad with stainless steel

■ *Criteria : (16,18)*

- * American code: impact upper shelf energy (USE) requirement :
 - > 100 J in the belt region
 - > 68 J in all other parts

For a maximum allowable neutron fluence at the inner surface of the pressure vessel of $3 \times 10^{19} \text{ n/cm}^2$ ($E < 1 \text{ MeV}$)

- * German code
USE > 100 J all over the pressure vessel

For a maximum allowable neutron fluence of $\approx 1 \times 10^{19} \text{ n/cm}^2$ ($E < 1 \text{ MeV}$)

- * Both the American and German codes require that the ductile-brittle transition temperature (DBTT) at 41J (T_{41J}) $\leq 0 \text{ }^\circ\text{C}$

■ ***Development***⁽¹⁶⁻²²⁾

- * Design of PVS⁽¹⁶⁾ welds, forged parts (Fig. 10)
- * The role of impurities⁽¹⁷⁻¹⁹⁾ specially Cu and P and their effect on transition temperature (Table 2 and Fig 11)
- * The impact characteristics of some advanced (low Cu and P) PVS (Figs 12-14)
- * Steam generator design (Fig. 15)
- * Pressure tube materials for CANDU reactors (Zircaloy-2 Vs Zr-Nb)
- * Pressure vessel thermal annealing (Fig. 16)

5. MATERIALS FOR HIGH WASTE CONTAINERS⁽²³⁻³⁰⁾

■ ***Constructional Materials Used in the Back End of the Fuel Cycle:***

- * Waste processing plants
- * Containers for high level nuclear waste for deep underground burial for super -long term storage.

■ ***Materials:***

- * Stainless steels (passive alloys)
- * R& D on materials

■ ***Environments***

- * Aqueous electrolytes containing:
 - Chlorides: may be high concentrations
 - Dissolved oxygen or other oxidizers

- * Exposure to :
 - Gamma radiation leading to radiolysis and formation of oxidizing agents, e.g. H_2O_2 and ClO^-
 - High temperature
- **Major Problem: Corrosion**
 - * Most common type : localized corrosion
 - * Crevice corrosion is the most predominant
- **Crevice Corrosion**
 - * Mechanism (Fig. 17)
 - * Parameters related to crevice dimensions
 - Crevice gap and depth
 - Bold/ crevice area ratio
 - * Parameters related to bulk environment
 - Chloride ion concentration
 - Oxygen (H_2O_2 or other oxidizers) content
 - Temperature
 - pH
 - Agitation
 - * Parameters related to crevice environment
 - Hydrolysis
 - Diffusion & mobility of ions
 - Critical crevice solution (dpH)
 - * Parameters related to passive metal:
 - Alloy composition & metallurgical history
 - Minor constituents
 - Passive film properties

■ ***Materials Development***

- * Stainless steels
- * Nickel & copper base alloys
- * Titanium alloys

■ ***Candidate Stainless Steels, Nickel, Copper Base Alloys*** ⁽²⁴⁻²⁸⁾

- * Statistical evaluation of the frequency and depth of crevice attack for type 316 Lss, some duplex ss and nickel base (625) alloys in different pH values up to 1000 hours (Fig. 18)
- * Evaluation of the susceptibility to crevice corrosion of : 304 ss, nickel-base alloys 625, C-276 and titanium alloys grades 2 and 12 in chloride environments ⁽²⁵⁾
 - Tests in bulk & crevice environments
 - Values of depassivation pH (dpH), passive current (I_p) and critical crevice temperature (CCT)
 - Ranking of alloys
- * Evaluation of the effect of fabrication and welding programmes on some candidate materials for the proposed high-level waste containers at the proposed Yucca mountain repository site ⁽²⁶⁾
 - Materials : 304 L ss, 316L ss, nickel-base alloy 825, and copper-base alloys CDA 102, CDA 613, CDA715
 - Studies cover crevice corrosion, stress corrosion cracking (SCC) and enhancement of corrosion by gamma radiation.

- * Investigations of the pitting, crevice and SCC of Mo bearing nickel-base alloys (C-276 and 625) as candidate materials for the Canadian nuclear fuel waste management programme ⁽²⁷⁾ several environmental, electrochemical (pH, temp. Cl⁻, radiolysis...etc.) and materials parameters were studied.
 - Crevice corrosion susceptibility diagrams (Figs. 19,20)
 - Dependence of passivation breakdown potentials on temperature (Fig.21)
 - Pitting & crevice corrosion temperatures in various media (Tables 3 & 4)
 - * An extensive programme was carried out on the Ni-base alloy 825, a candidate material for the Yucca mountain repository site ⁽²⁸⁾. This involved testing in well water from the vicinity of the site in Nevada. A pilot facility was designed to examine the interaction of chlorides, sulfates, nitrates, fluorides and temperature.
- ***Candidate Titanium Alloys*** ⁽²⁹⁻³⁰⁾
- * The crevice corrosion initiation time for commercial (grade1) titanium was found to be remarkably short when the chloride ion concentration ⁽²⁹⁾ is near to a critical value. Pre-passivation was found to prolong the initiation time.
 - * Review of susceptibility of several titanium alloys (Table 5) to crevice attack in relation to some environmental, electrochemical and materials parameters. The resistance of titanium alloys grades 2, 12 and 7 in some salts solutions ⁽³⁰⁾ is shown in Table 6

- * The critical pH values for crevice corrosion was found to be :⁽³⁰⁾
 - 9.5 -10 for grade 2 Titanium
 - 2 for grade 12 Titanium
 - 0.75 for grade 7 Titanium
- * The temperature- pH limits for titanium alloys in NaCl brines are shown in(Fig 22)
- ***The Crevice Corrosion Susceptibility Diagram for Various Alloys***
- * ***(Fig. 23)***

6. SOME NEW FUEL CYCLE CONCEPTS ^(8,31,32)

■ ***Factors Considered***

- * Economics
- * Availability of technology
- * The desired level of self sufficiency in nuclear power operation
- * Issues related to waste management and disposal

■ ***The Uranium - Plutonium Fuel Cycle (Fig. 24)***

■ ***The Thorium - Uranium Fuel Cycle (Fig. 25)***

■ ***Advanced Fuel Recycling (AFR) ⁽³¹⁾***

- * A new concept of fast reactor fuel cycle comprising
 - Simplification of the cycle and improvement of waste generation
 - Advancement of the cycle by the application of new fuel types
- * Primary concepts of actinide recycling (Fig. 26)
 - Strengthen utilization of fissionable nuclides
 - Improvement of generation of alpha- emitting wastes

- Strengthen recovery of actinides like Pu, and minor actinides (MAs) including Am, Cm and higher products
- Change the concept of reprocessing into one based on rough-purification, where imperfectly purified Pu with minor actinides (MAs) is recycled as a fast reactor fuel for burning
- Development of reprocessing technology, primary target being the recovery of actinides and cost reduction of secondary waste generation (Fig. 27)
- Fuel development involving less expensive, simple and easy

fabrication processes and modification of MOX fuel design, by

which recycling of all actinides (Pu and MAs) becomes possible (Fig. 28)

■ ***Thorium Based Fuel Cycles in CANDU Reactors***⁽³²⁾

- * The driver fuel can be:
 - Natural uranium
 - U²³³ in spent thorium fuel “ self sufficient thorium cycle, SSTC”
- * Reprocessing LWR fuel, 1.6 % U²³⁵ compared to 0.73 % (direct use of spent PWR fuel in CANDU, DUPIC)
- * Plutonium- Thorium cycles
 - LWR Pu
 - FBR Pu
 - Weapons Pu

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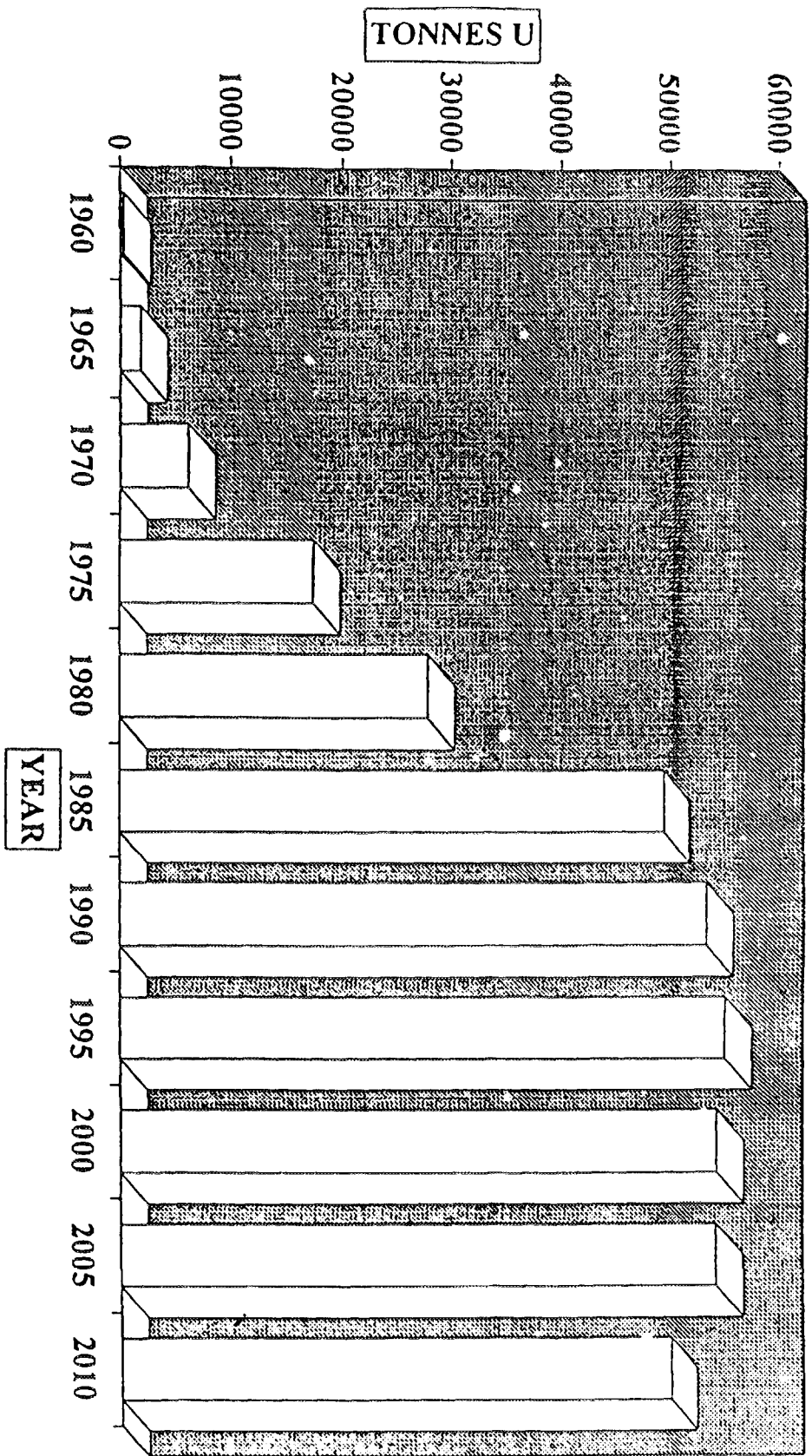


Figure 1 : Annual Plutonium Production Worldwide (7).

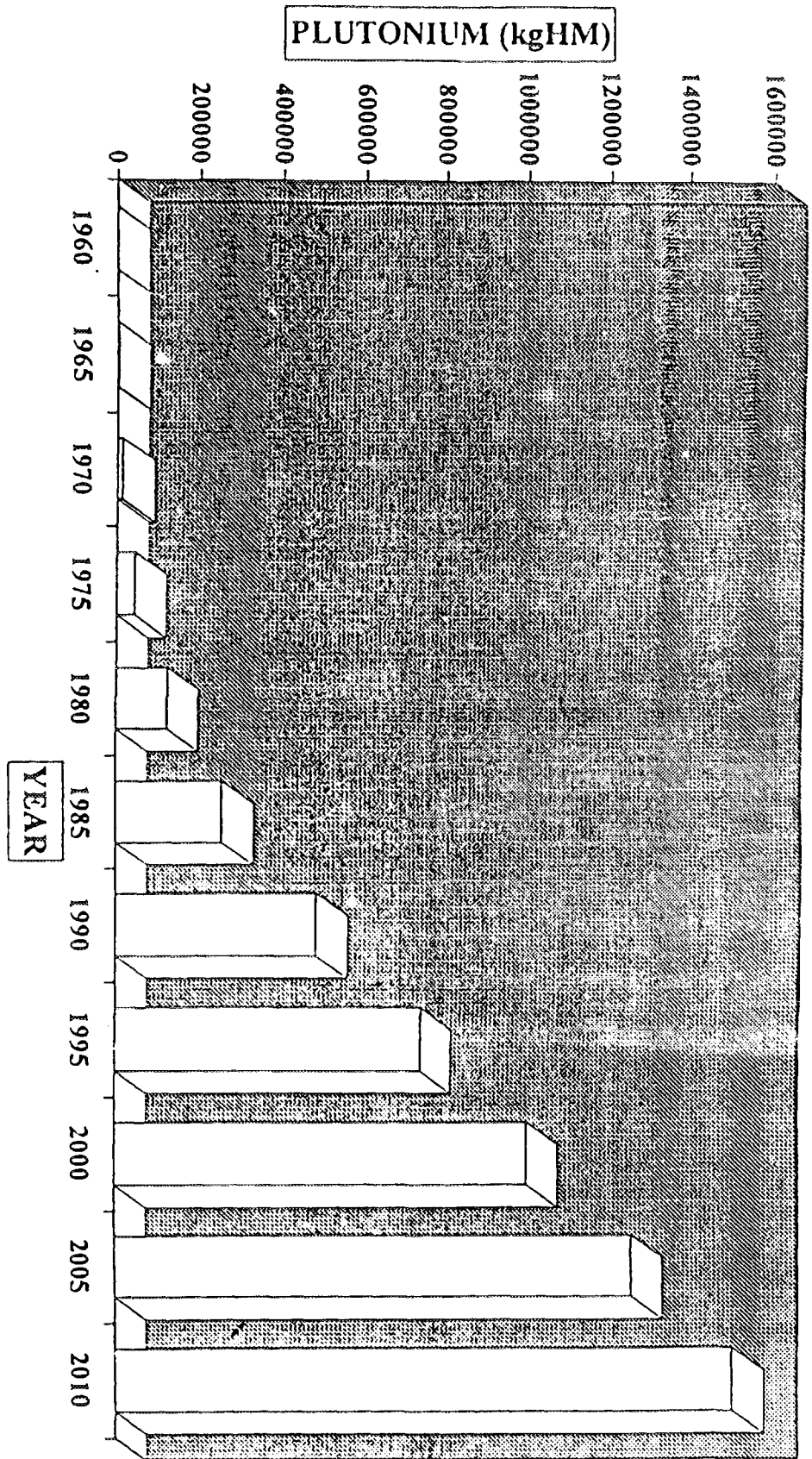


Figure 2 : Plutonium Accumulated Worldwide (7).

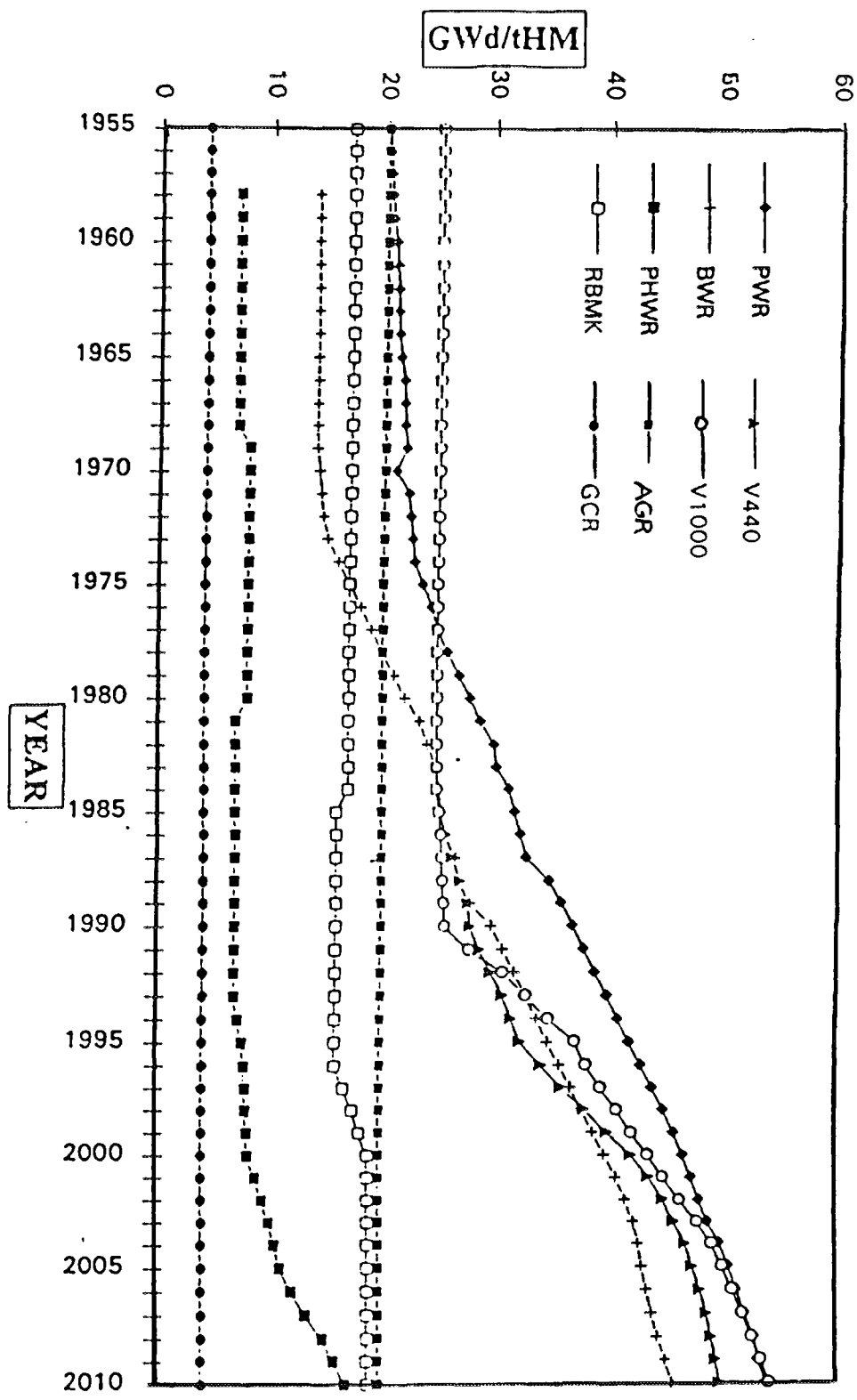


Figure 3 : Burnup Evolution (7).

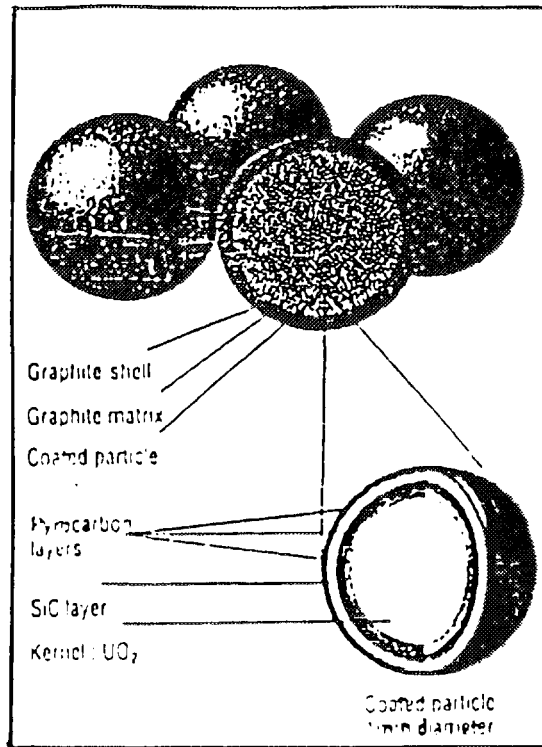


Figure 4 : HRT Fuel Elements, 60 mm dia. (9)

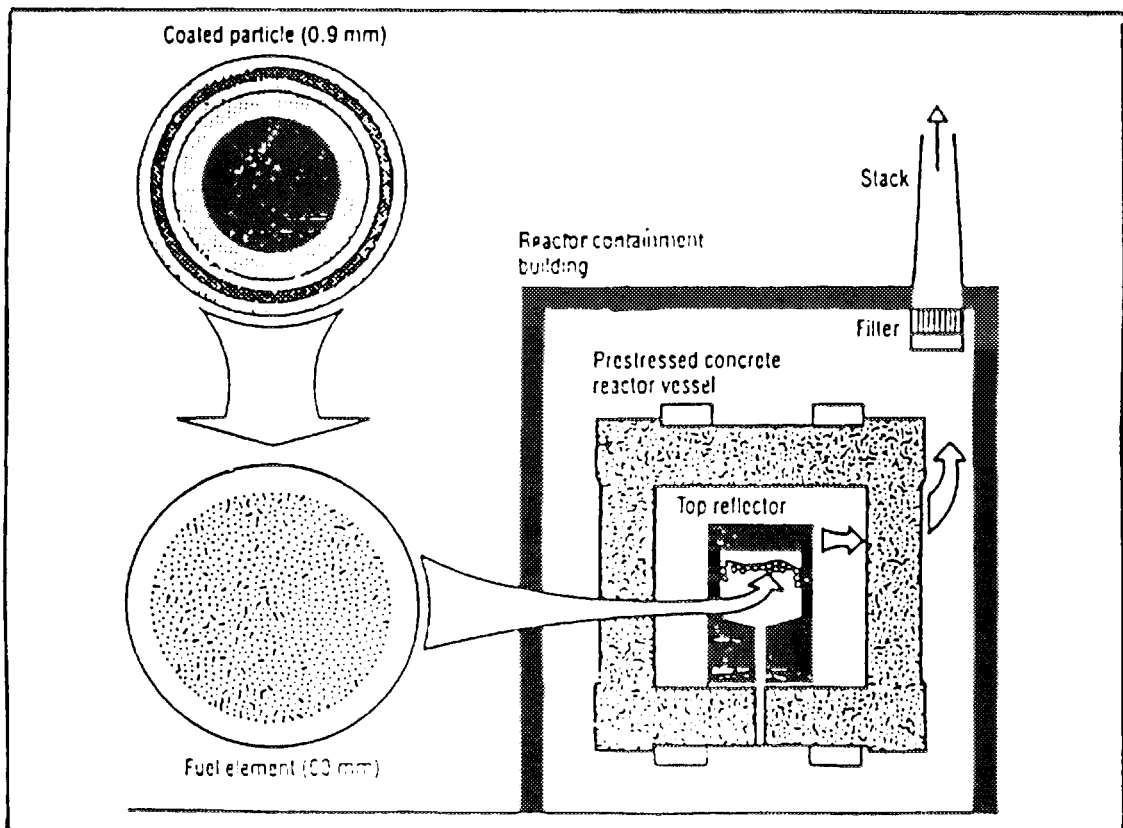


Figure 5 : Multiple Barriers for Fission Product Retention in a High Temperature Reactor (9).

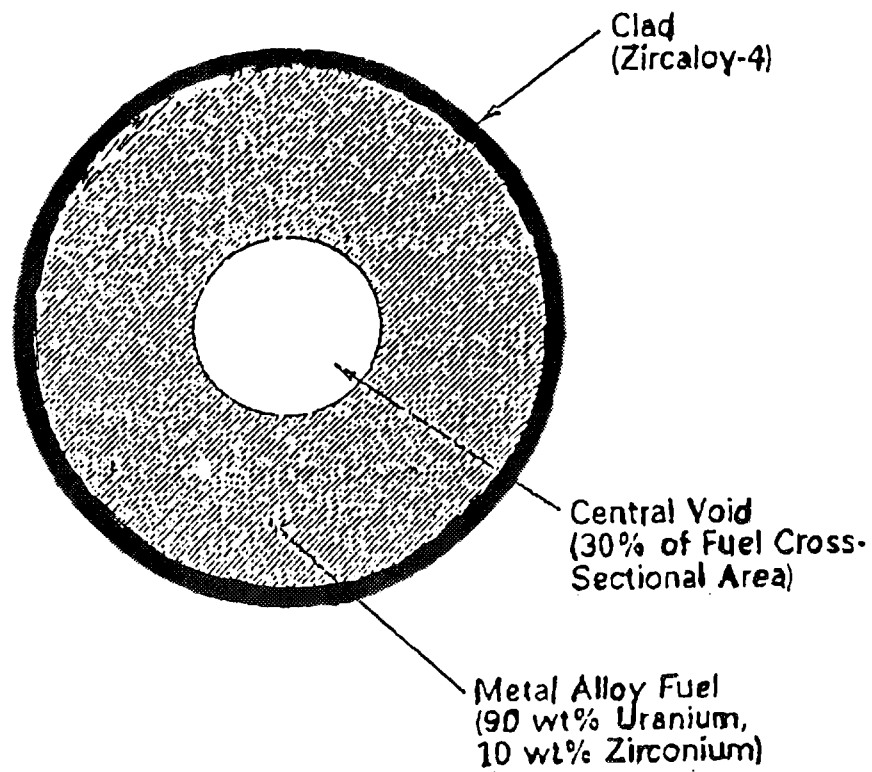


Figure 6 : Schematic of a Metal Alloy Fuel Pin (9).

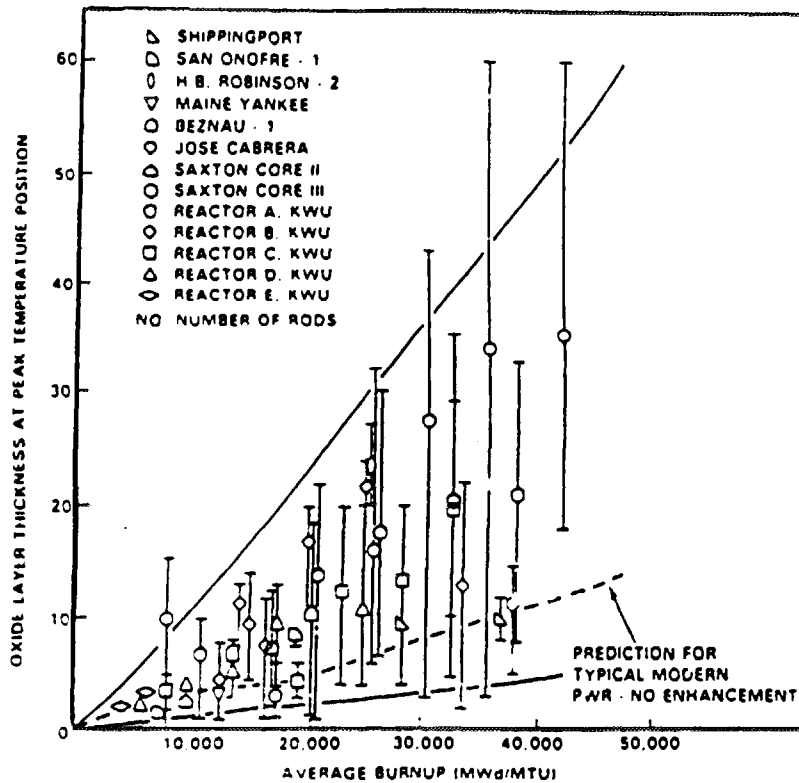


Figure 7 : Maximum Oxide Layer Thickness of PWR Fuel Rods Versus Burnup (11).

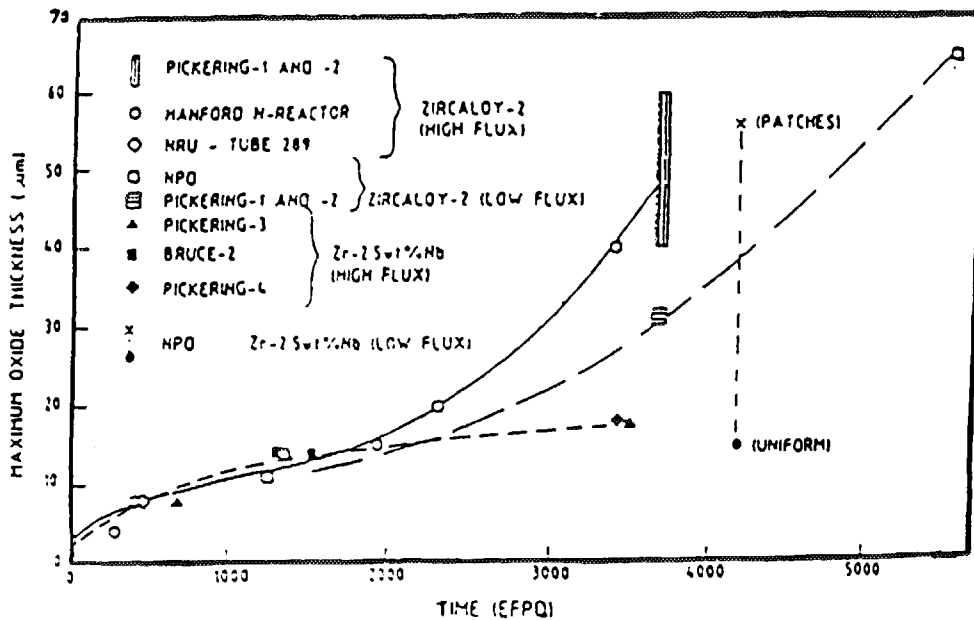


Figure 8 : Long Term Corrosion of Cold Worked Zircaloy-2 and Zr-2.5 Nb Pressure Tubes (11).

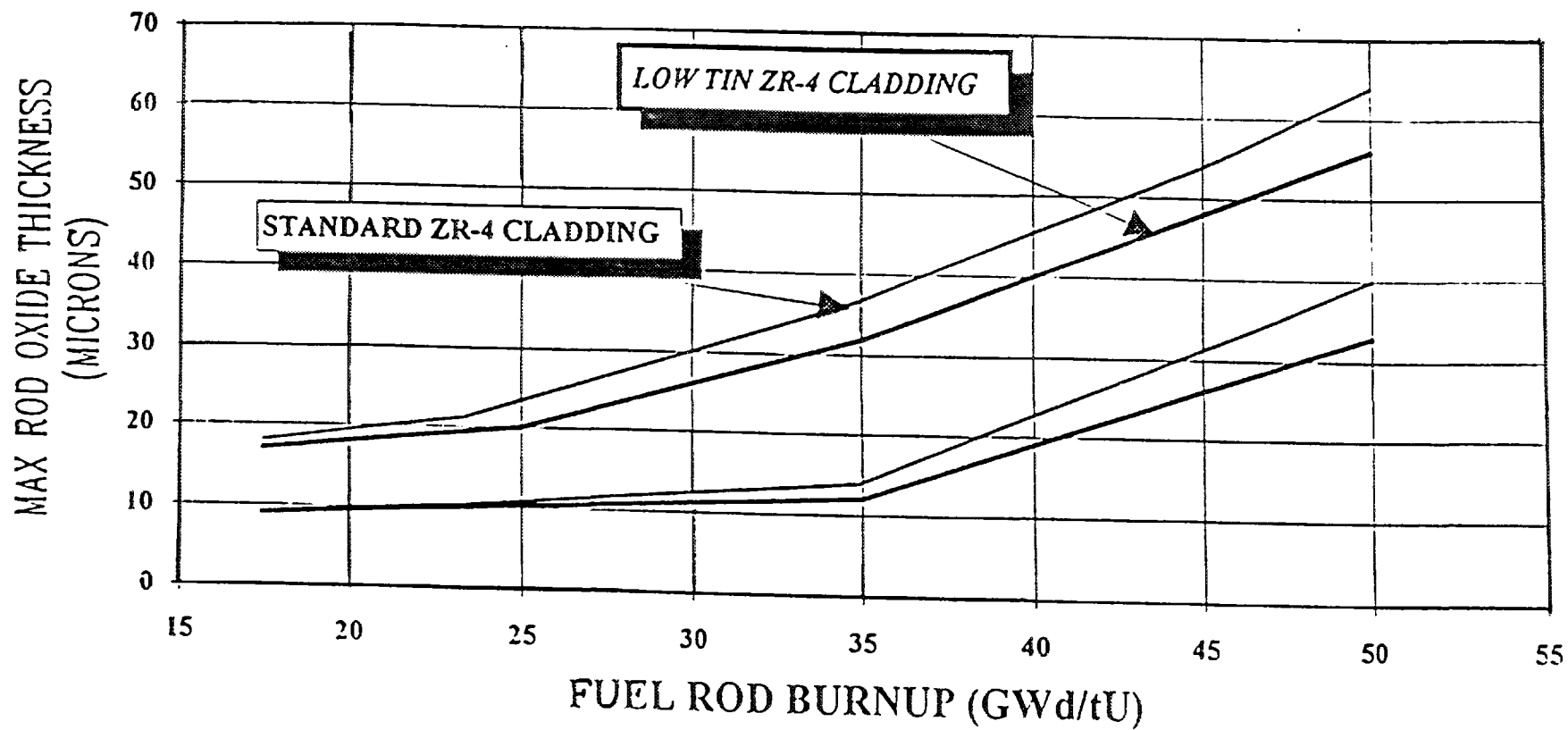


Figure 9 : Corrosion Experience at EDF - Effect of Clad's Tin Content (12).

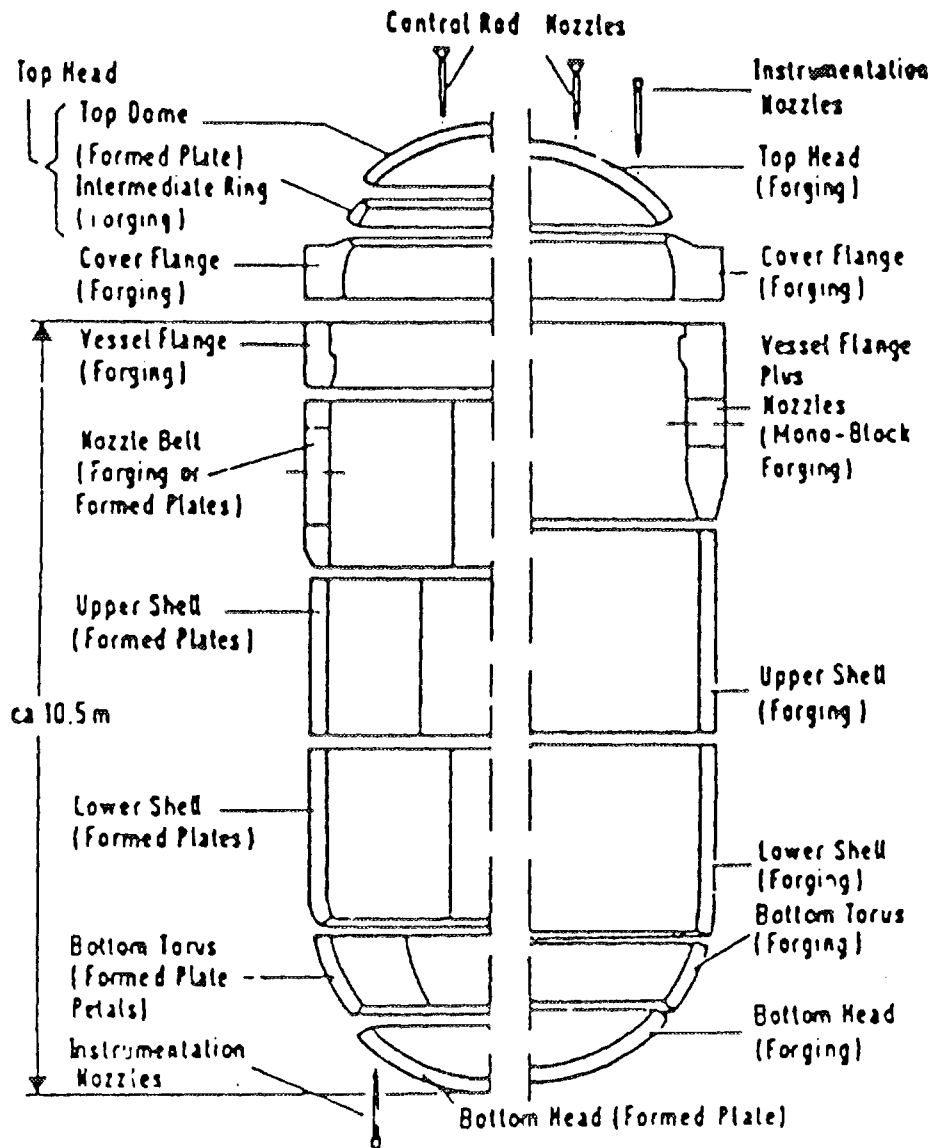


Figure 10 : Development of Material Layout in A PWRPV (16).

Left - Conventional Design
Right - Advanced Design

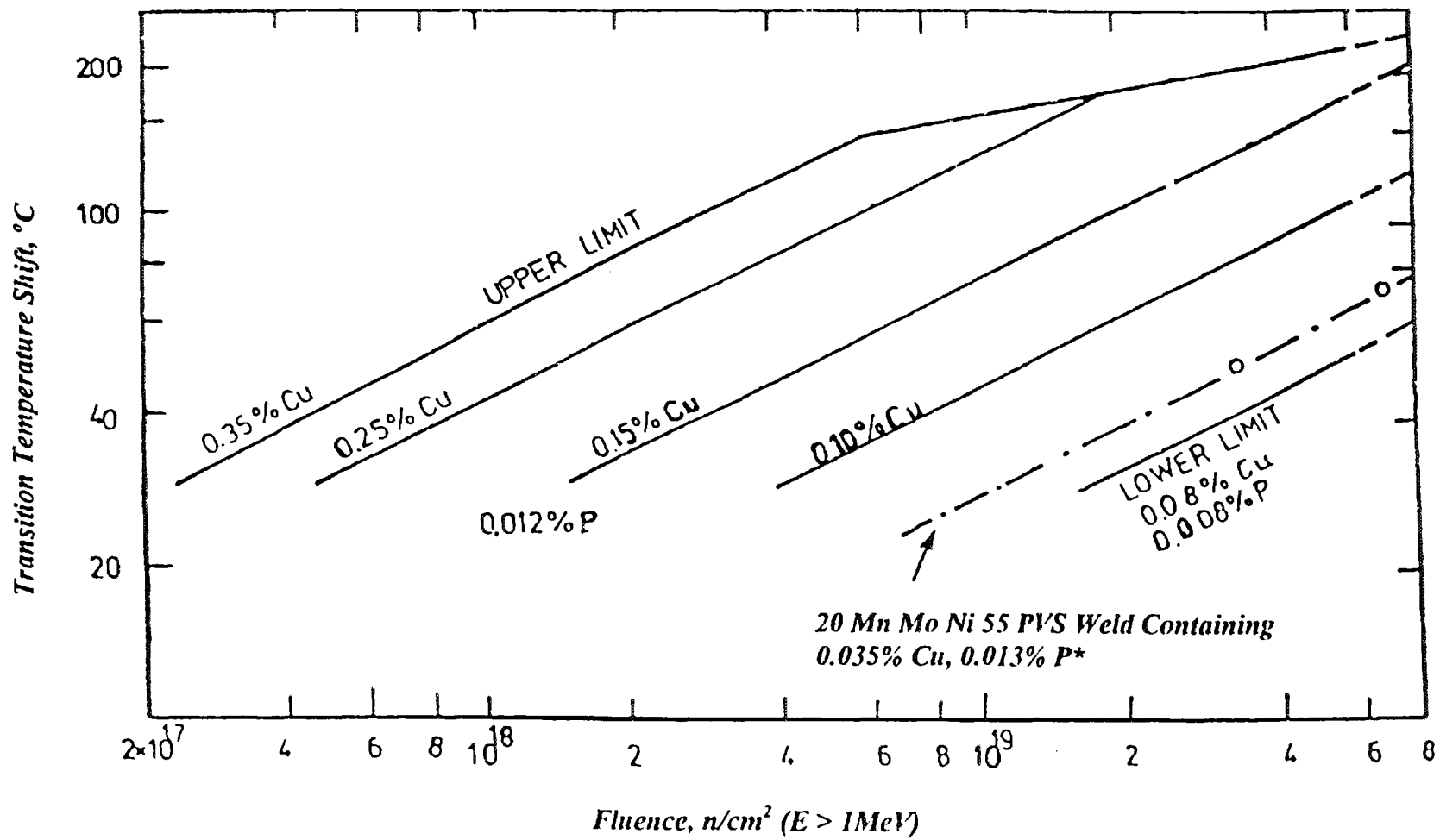


Figure 11 : Effect of Cu and P Content on the Transition Temperature of PVS (18,19).

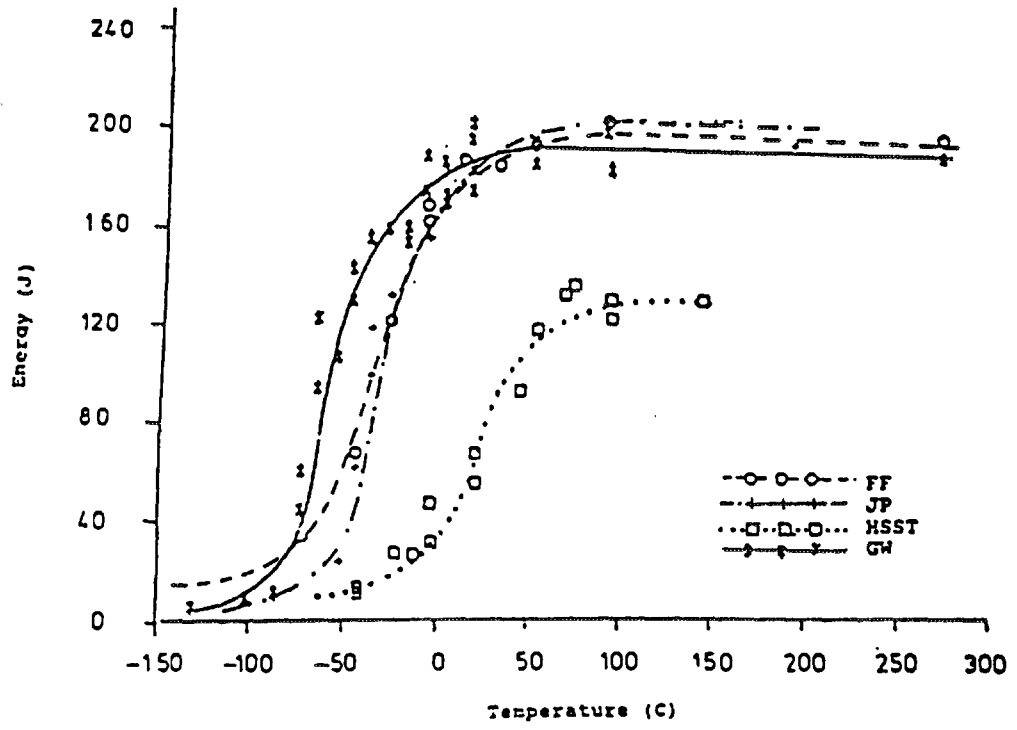


Figure 12 : Impact Transition Curves of Tested Steels (20).

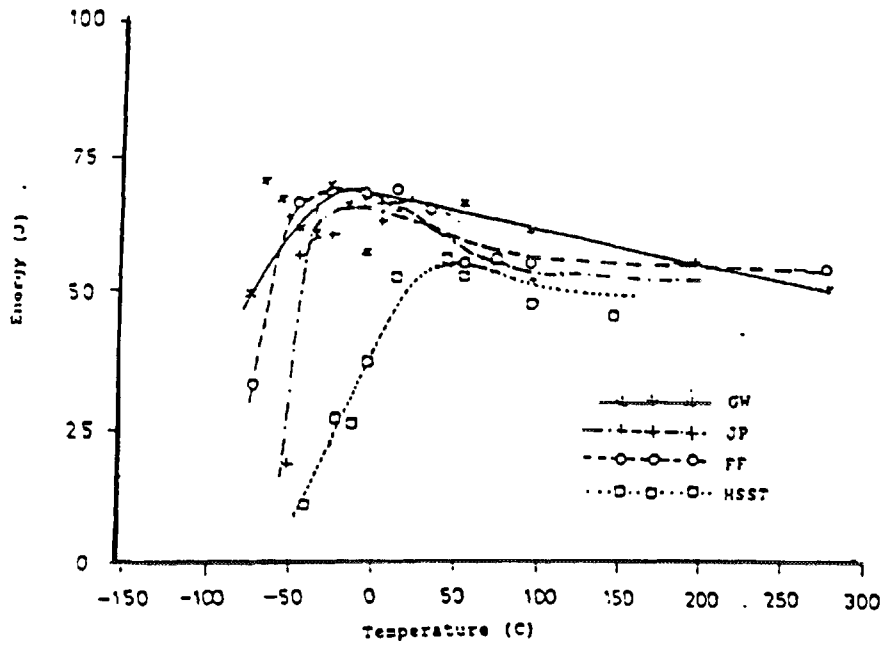


Figure 13 : Effect of Test Temperature on Initiation Energy (20).

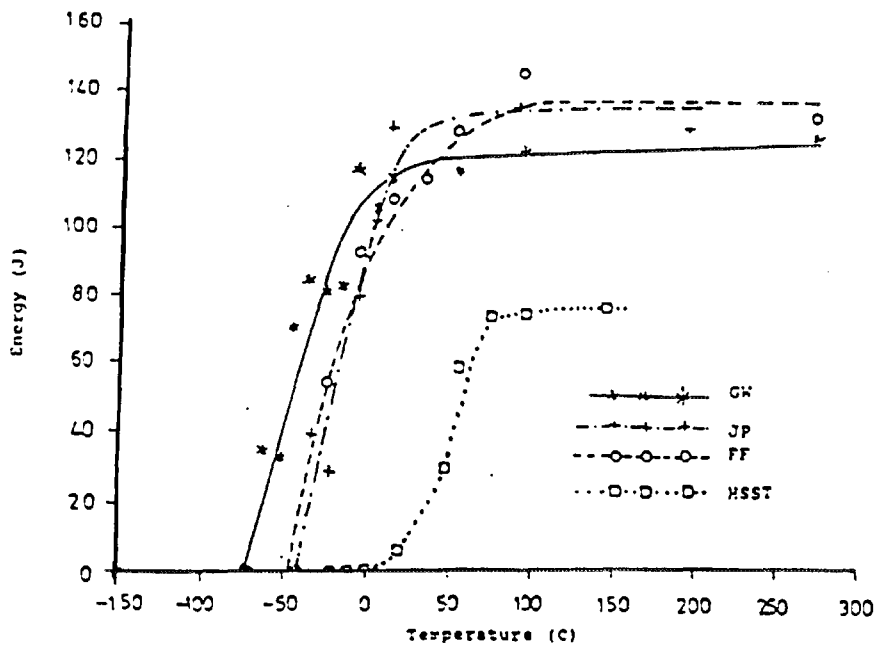


Figure 14 : Effect of Test Temperature on Propagation Energy (20).

Steam Generator

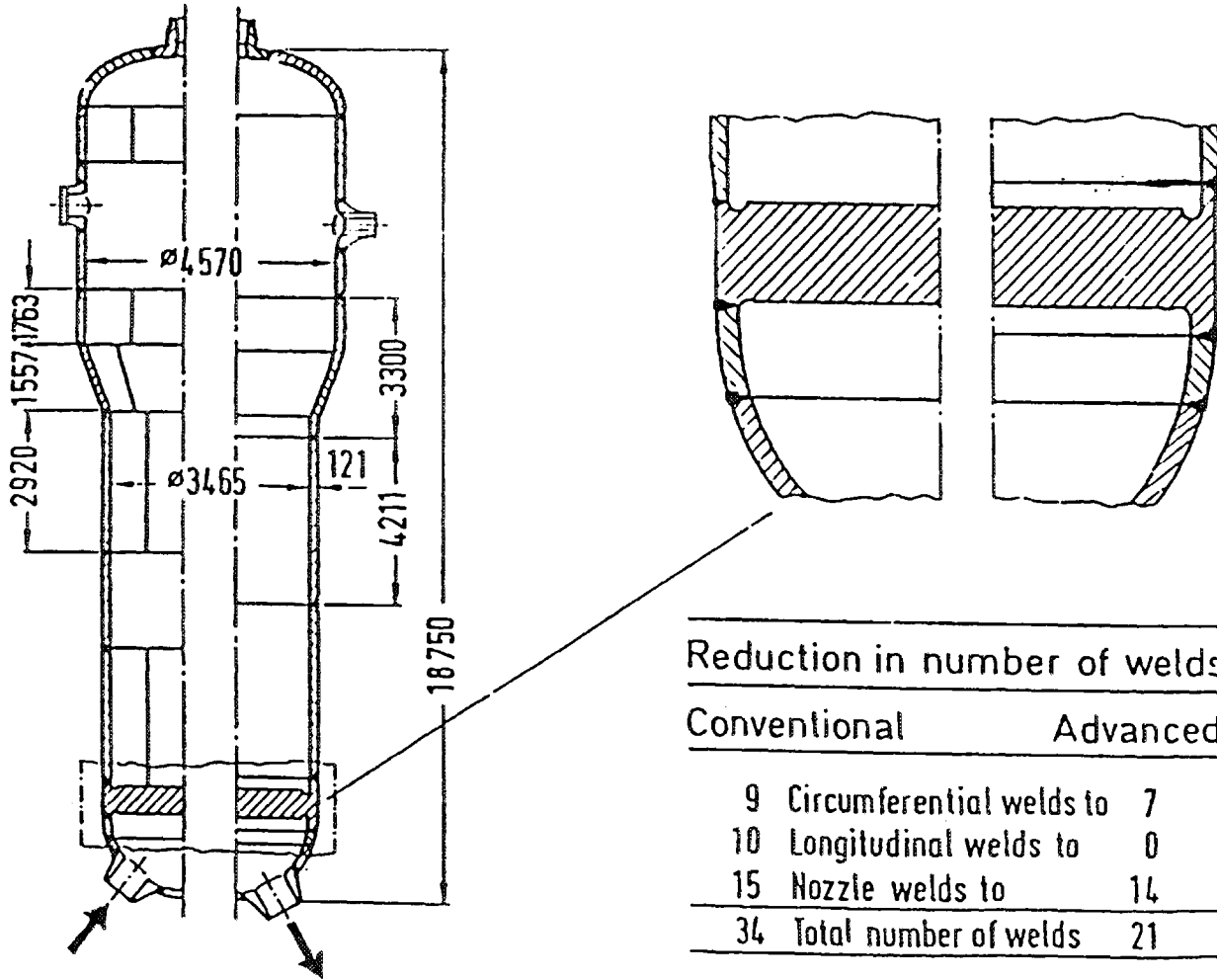


Figure 15 : Example of Conventional and Advanced Design for Steam Generators (21).

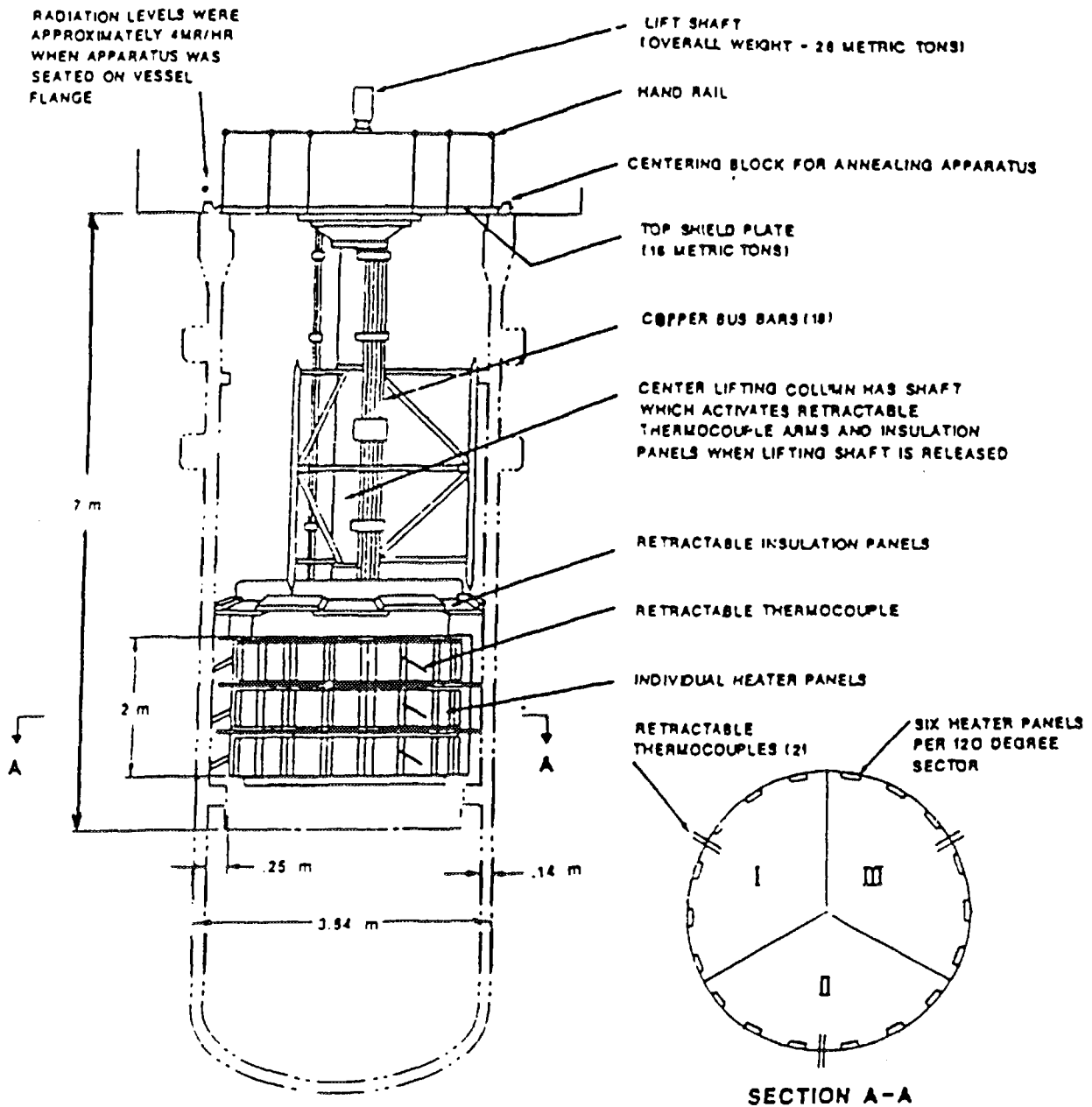


Figure 16 : Soviet Thermal annealing Apparatus Showing the Annealing Zone of VVER-440- Type RPV(22).

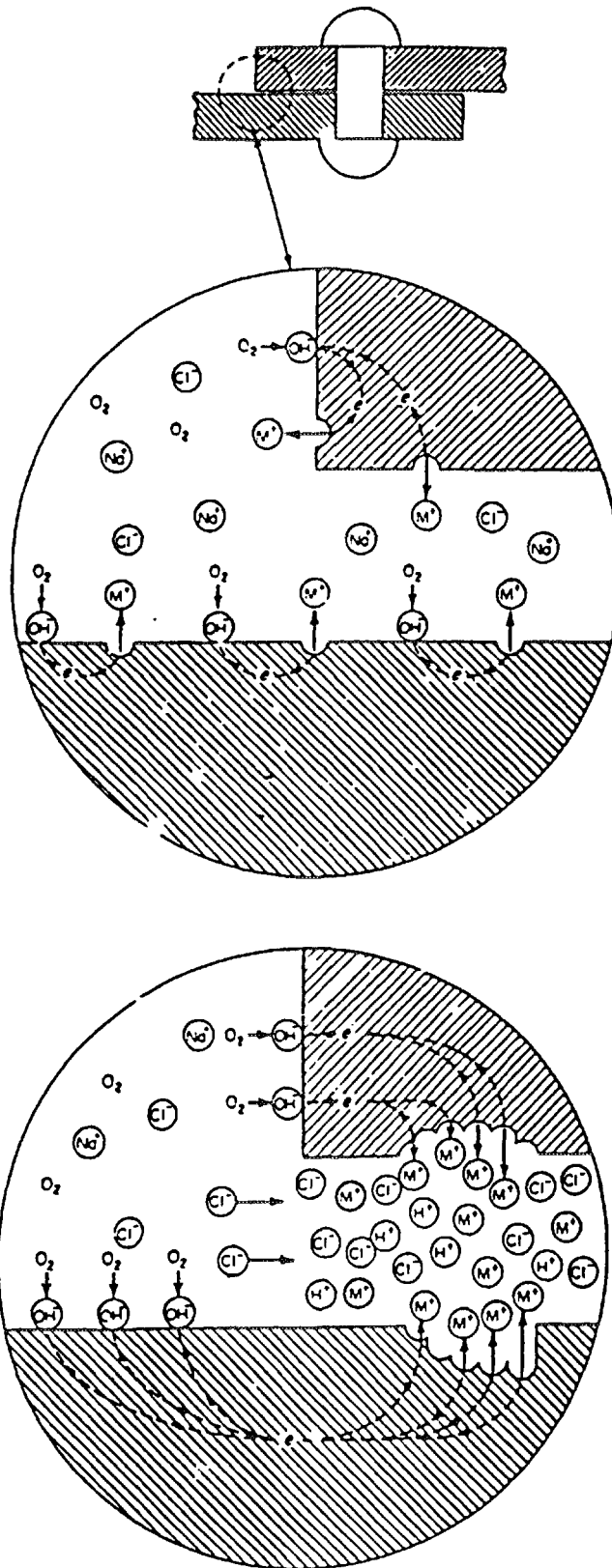
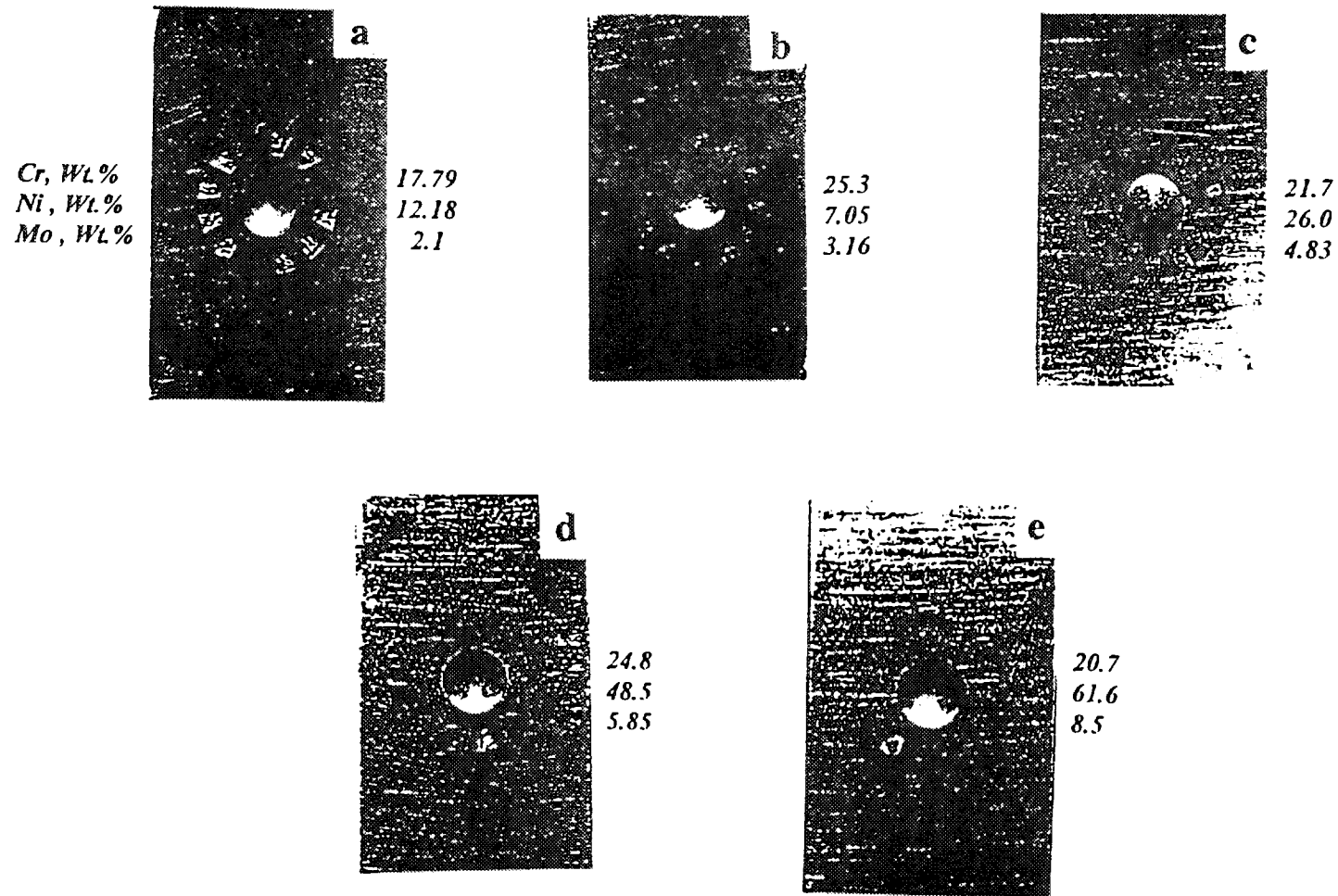
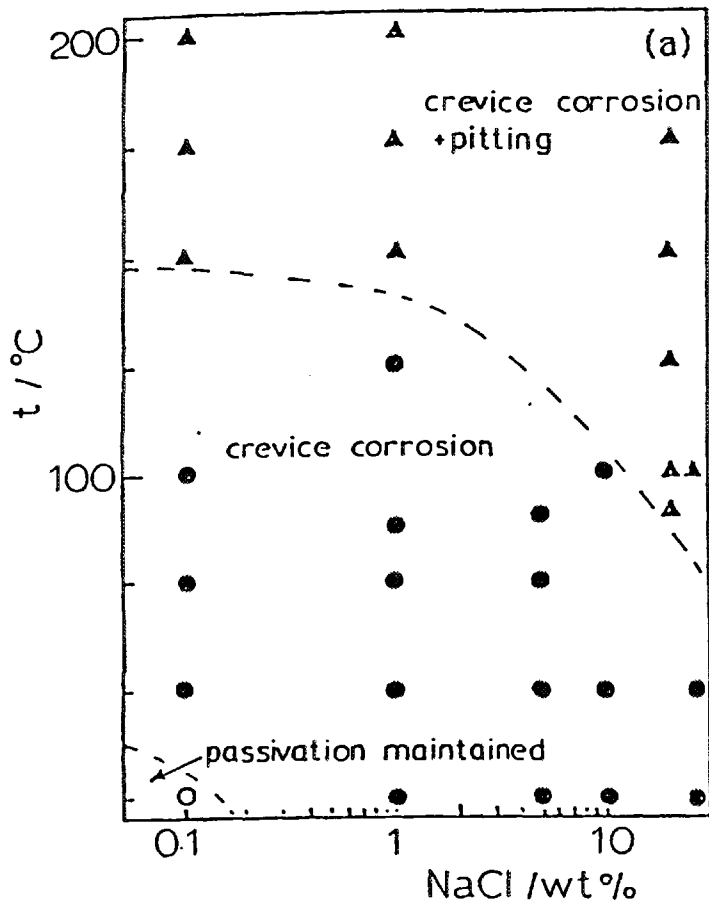


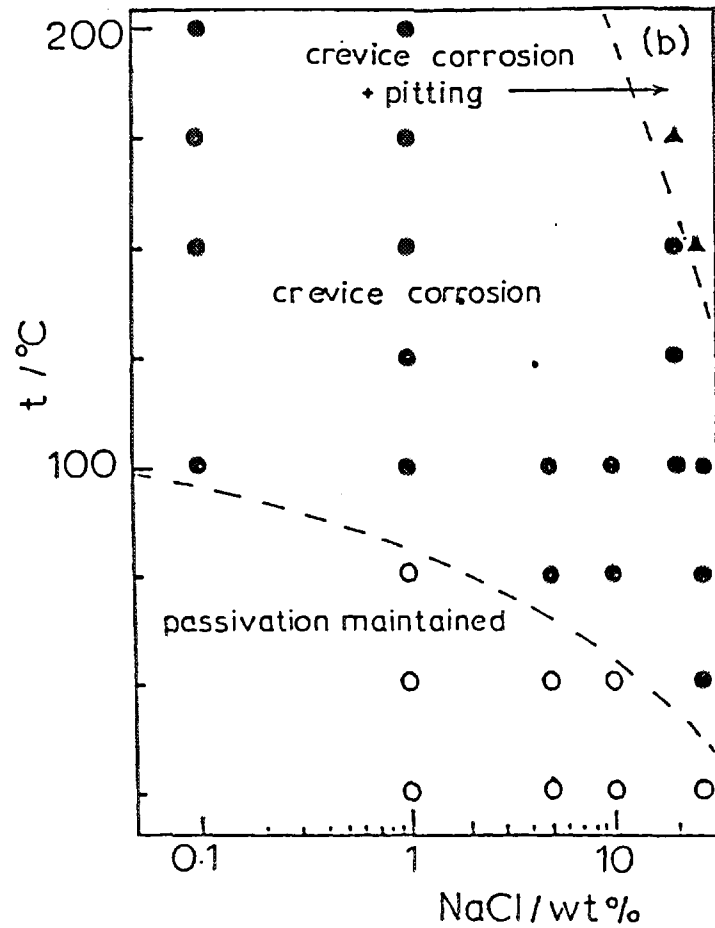
Figure 17 : Crevice Corrosion Mechanism (23).



**Figure 18 : External View of Crevice Corrosion for Various Test Alloys
Exposed in Synthetic Waste Solution (pH=5) for 500 hour (24).
a type 316L, b DP-3, c NAR-A, d NAR-B, e alloy 625.**



(a) Alloy 625



(b) Alloy C-276

Figure 19 : Susceptibility Diagrams for Various Corrosion Processes Determined from Cyclic Polarization Experiments (27).

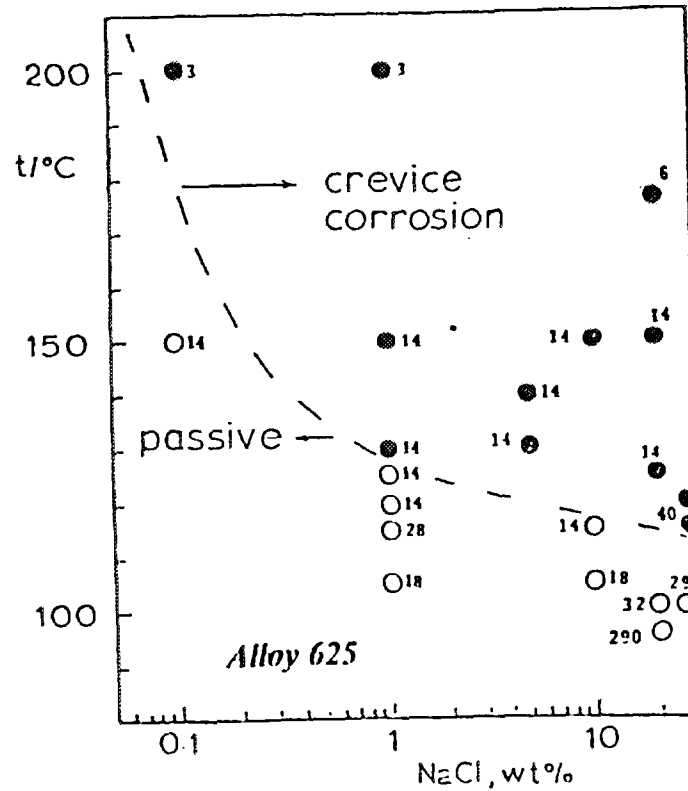
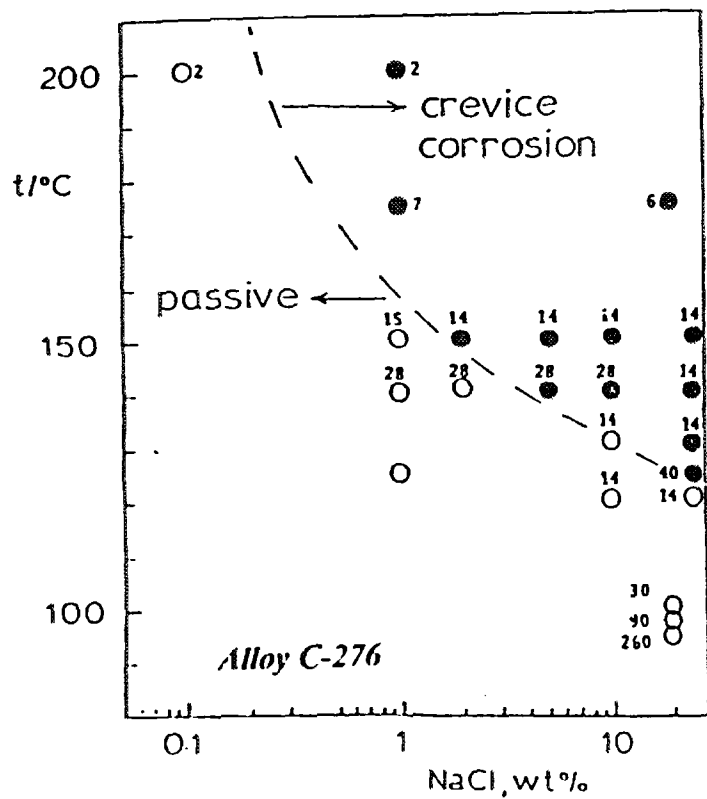


Figure 20 : Crevice Corrosion Susceptibility Diagrams for Hastelloy C-276 and Inconel - 625 in Aerated Solutions, the Number Beside Each Data Point is the Exposure Time in Days (27).

- = crevice corrosion with visible metal loss;
- o = no visible corrosion;
- = tentative crevice temperature envelope

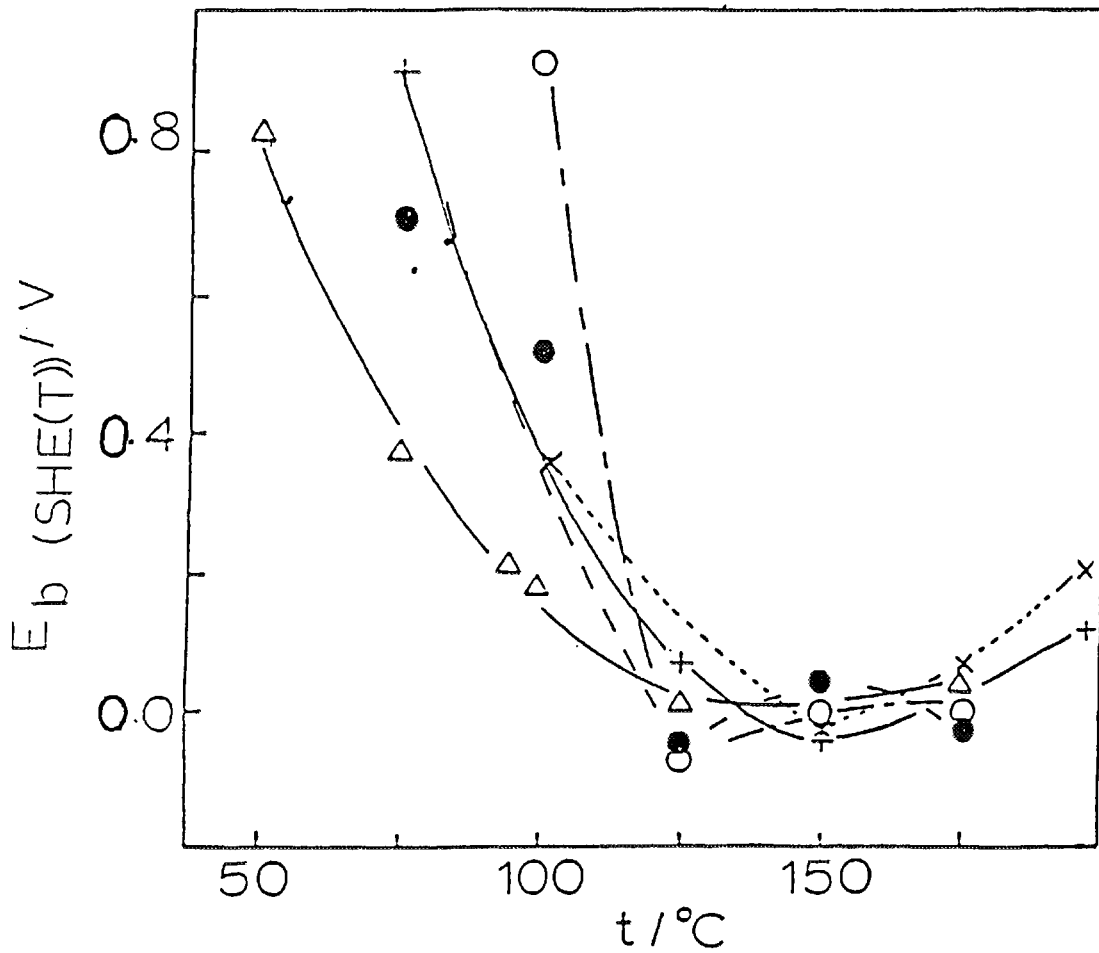


Figure 21 : Passivation Breakdown Potentials from Cyclic Polarization Experiments as a Function of Temperature (27).

+	= Alloy 625,	0.1 wt % NaCl
Δ	= Alloy 625,	20.0 wt % NaCl
•	= Alloy C-276,	20.0 wt % NaCl
o	= Alloy C-276,	1.0 wt % NaCl
X	= Alloy C-276,	0.1 wt % NaCl

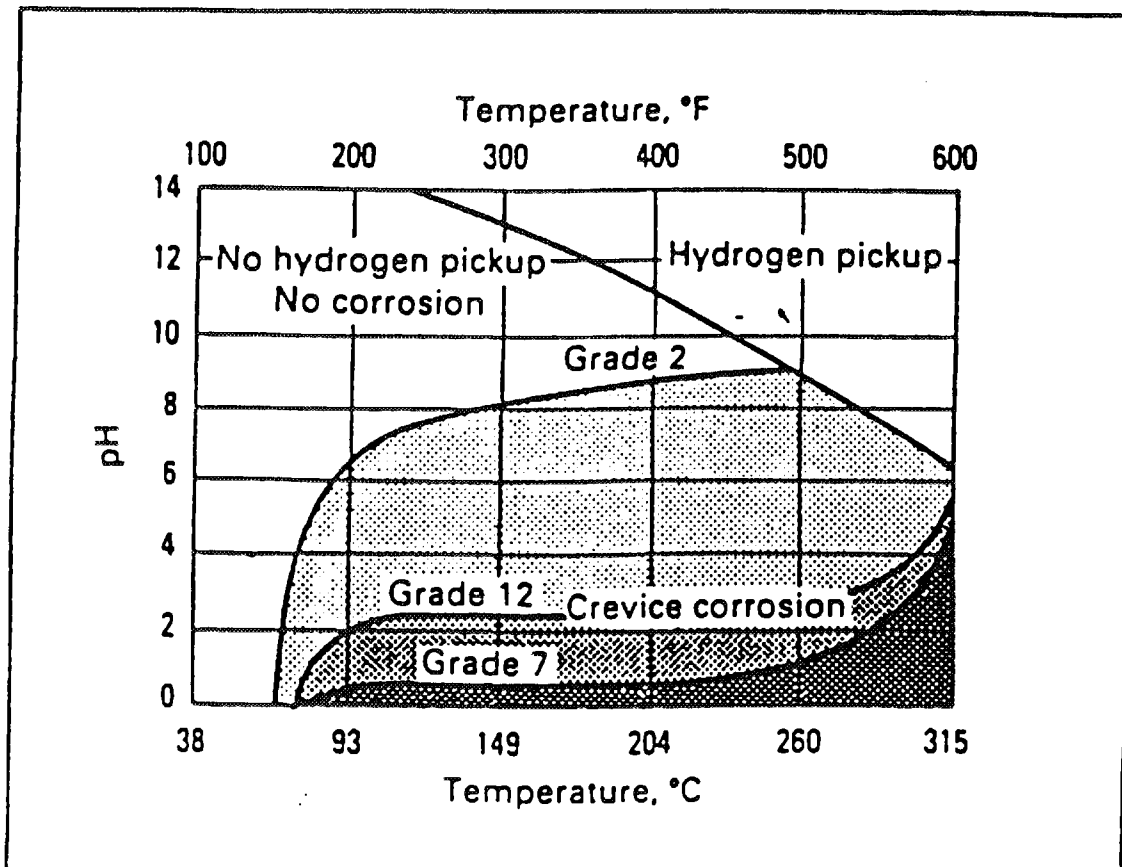


Figure 22 : Temperature - pH Limits for Titanium Alloys in NaCl Brines Based on Laboratory and Field Experience (30).

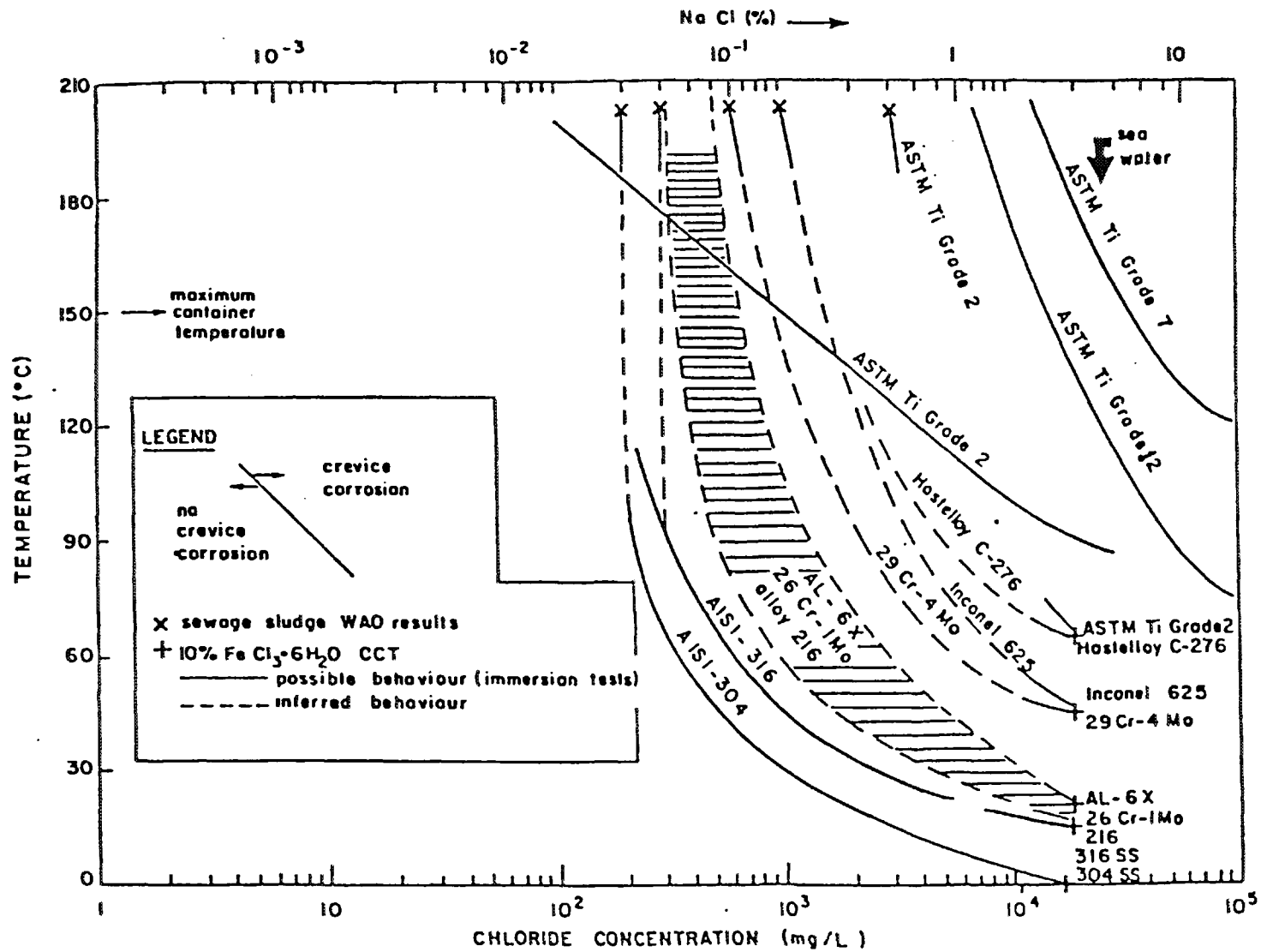


Figure 23 : Susceptibility Diagrams for the Crevice Corrosion of Various Alloys in Aqueous Chloride Solutions (27).

- Uranium Once - through Option

Route 2 → 7 → 10 → 32

- Uranium Plutonium Fuel Cycle

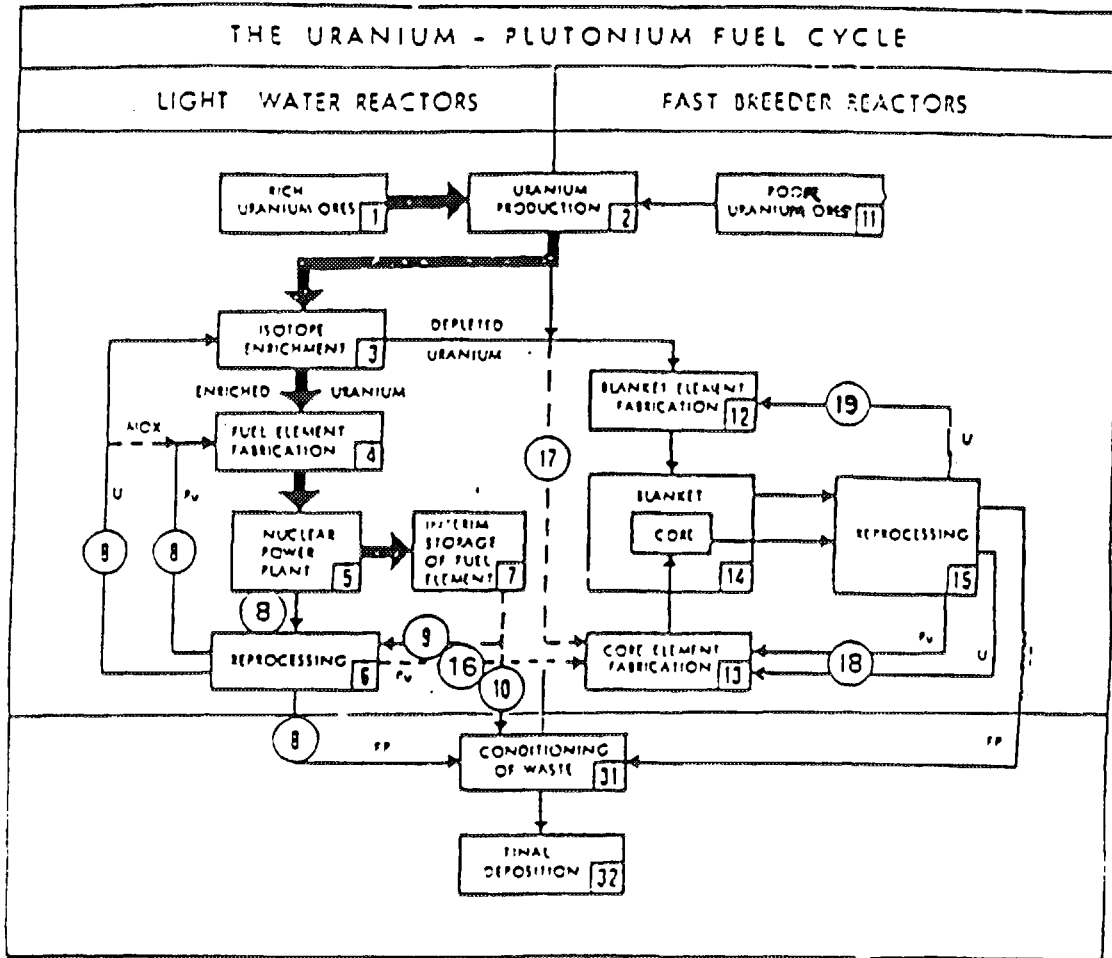


Figure 24 : The Uranium - Plutonium Fuel Cycle

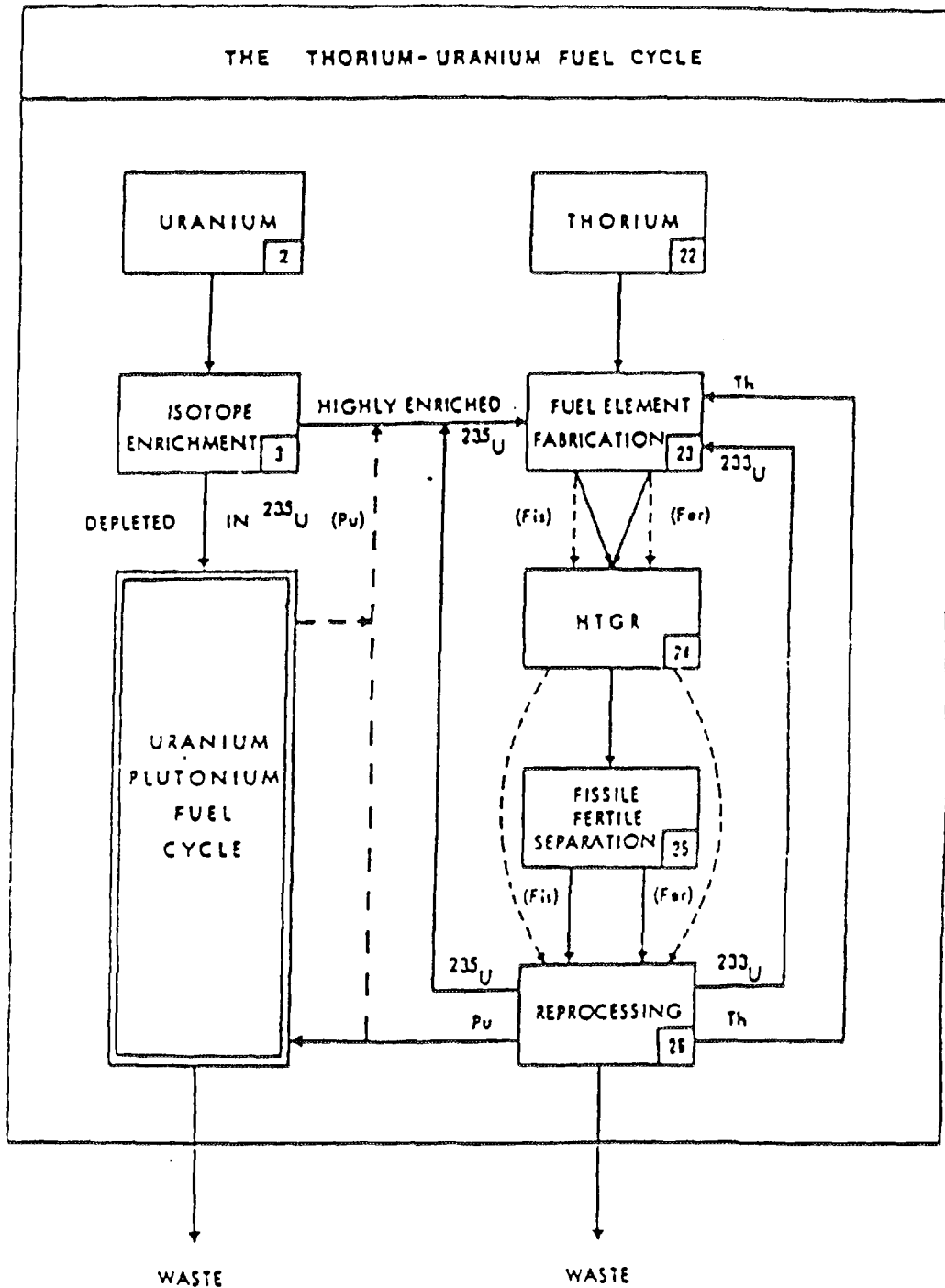
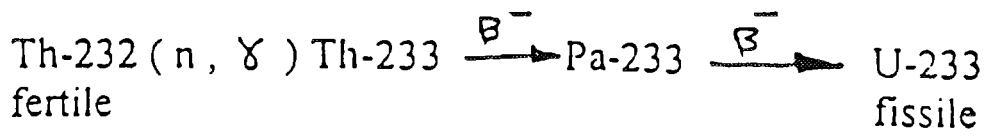
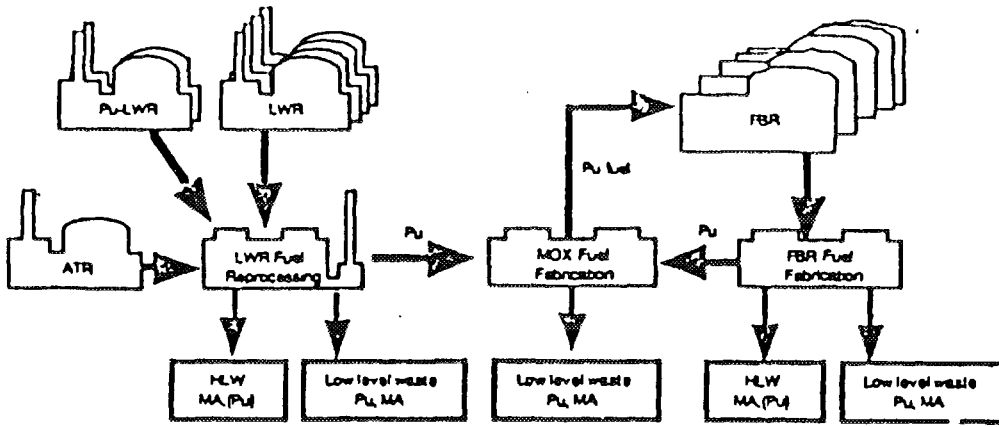
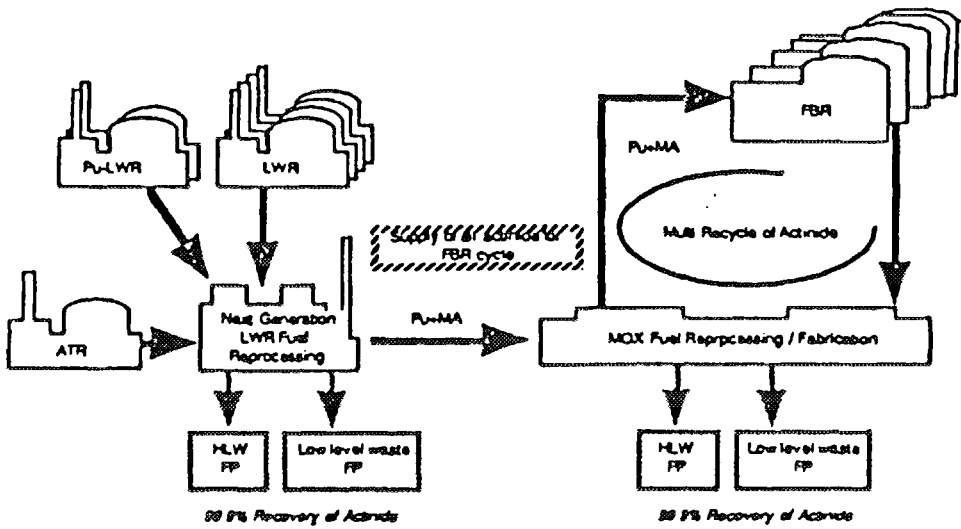


Figure 25 : The Thorium - Uranium Fuel Cycle



Concept of Existing Fuel Cycle



Concept of Actinide Recycle

HLW:	High Level Waste
FP:	Fission Products
MA:	Minor Actinide

Figure 26 : Actinide Recycle Concept (31)

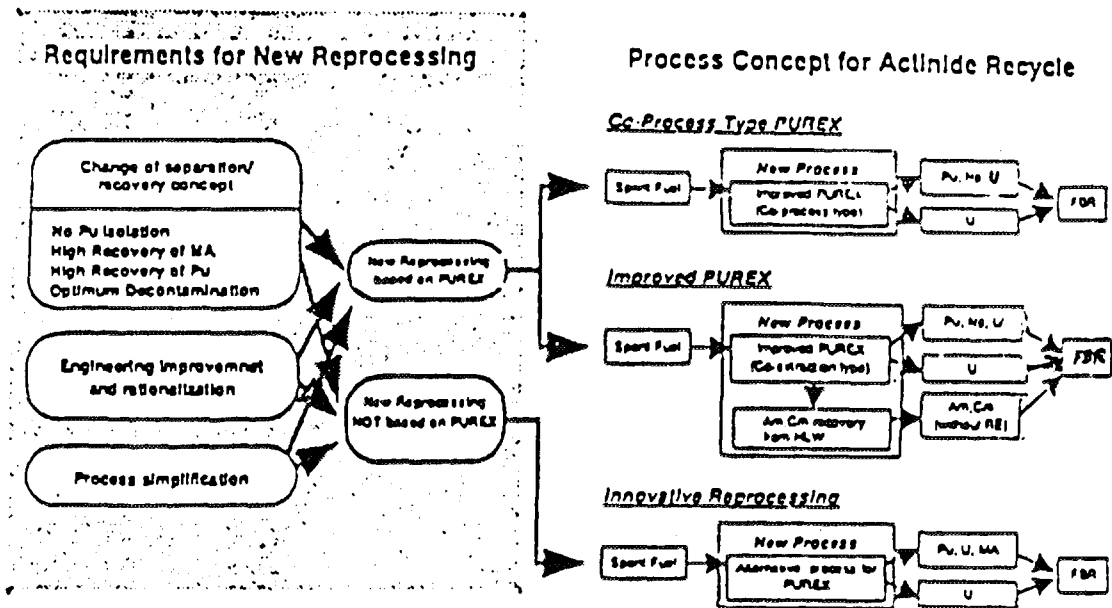


Figure 27 : Concept of Reprocessing for Actinide Recycle (31).

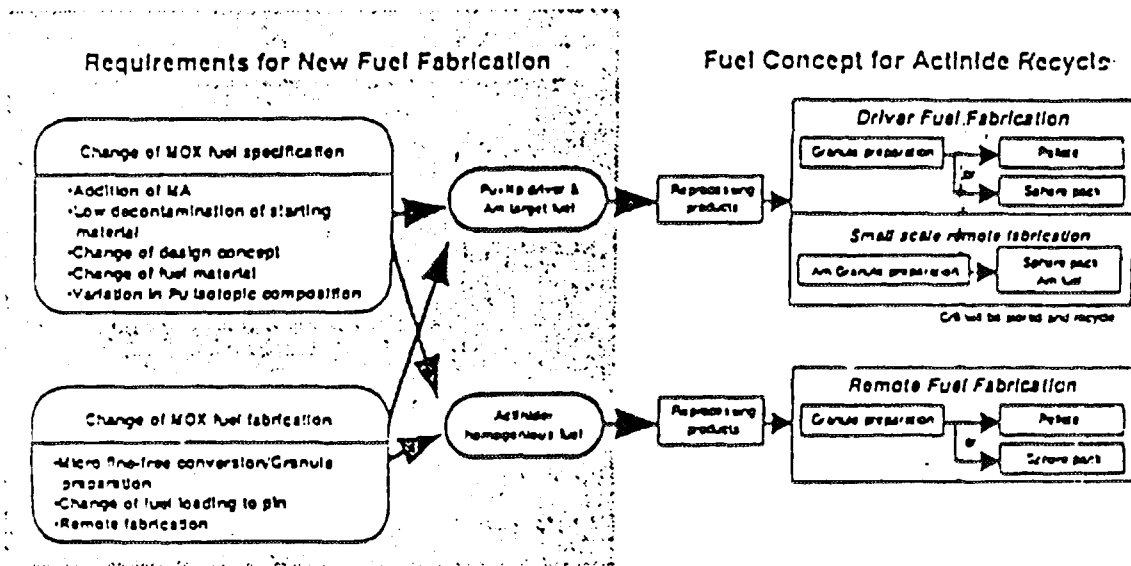


Figure 28 : Concept of Fuel Technology for Actinide Recycle (31).

Table 1 : Zirconium Characteristics Under Irradiation (11).

<u>Alloy</u>	<u>Oxidation Rate</u>	<u>Hydriding Rate</u>
1. Zircaloy-2	Intermediate rates	Intermediate rates
2. Low Ni Zircaloy-2	Similar to other Zircalloys	Like Zircaloy-4
3. Zircaloy-4	Like Zircaloy-2 in PWRs, but higher oxidation in BWRs	Generally less than Zircaloy-2
4. Zr-1.2 Cr-0.08 Fe	Like Zircalloys but susceptible to alloy segregation	Similar to Zircaloy-2 except when alloy segregation evident
5. Zr-1.2 Cu-0.28 Fe	Much higher than Zircalloys	Much higher than Zircalloys
6. Zr-3 Nb-1 Sn	Mildly lower than Zircalloys	Mildly lower than Zircaloy-4
7. Ozhennite-0.5	Similar to Zircalloys ^(a)	Higher than Zircaloy-2 ^(a)
8. Zr-2.5 Nb	Generally Lower than Zircalloys	Much lower than Zircalloys
9. Crystal bar zirconium	Higher than Zircalloys	Much higher than Zircalloys
10. Zr-1 Nb	(b)	(b)
11. Zr-3.5 Sn-0.8 Mo-0.8 Nb	Higher than Zircalloys	--
12. Scanuk Alloys	--	--
13. Zr-1.0 Nb-1.0 Sn-0.1 Fe	Lower than Zircaloy	Lower than Zircaloy-4 due to lower oxidation

(a) Loop Study Reported in Reviews on Coating and Corrosion, vol. I, No.4 (1975) pp. 299-366.

(b) Few Direct Comparisons to Zircalloys are Available. Some References Cite Relatively Higher Corrosion Rates Than Zircaloy - 4. Russian Experience Shows.(WVER - 440) Low Corrosion and Hydriding Rates.

Table 2 : Chemical Composition of some advanced PVSs, wt. % (20).

	GW	FF	JP	HSST
C	0.08	0.15	0.18	0.25
S	0.17	0.28	0.22	0.25
Mn	1.45	1.3	1.4	1.33
Mo	0.61	0.5	0.58	0.51
Ni	0.93	0.7	0.66	0.65
Cr	0.02	0.24	0.2	0.1
P	0.011	0.009	0.007	0.011
Cu	0.035	0.07	0.015	0.13
S	0.006	0.007	0.004	0.018

Table 3 : Crevice - Corrosion Temperatures (°C) in 10 wt% FeCl₃ (27).

<i>Reference</i>	<i>Inconel - 625</i>	<i>Hastelloy C-276</i>
<i>Streicher^a</i>	<i>RT-50*</i>	<i>65-75*</i>
<i>Steigerwald^b</i>	<i>RT-50*</i>	<i>>50</i>
<i>Tapping^c</i>	<i>45</i>	

- a) *M.A. Streicher, " Development of Pitting Resistant Fe- Cr- Mo Alloy", Corrosion 30, 77 (1974)*
- b) *F. R. Sreigerwald, " New Molybdenum Stainless Steel and Alloys for Corrosion Resistance", Paper presented at NACE, Corrosion/74, Chicago, 111., 1974, paper No. 44.*
- c) *R. L. Tapping, Chalk River Nuclear Laboratories, unpublished work (1981).*

**T₁-T₂= resistant at T₁ but not at T₂*

RT room temperature.

Table 4 : Pitting and Crevice - Corrosion Temperatures in Oxidizing NaCl-HCl solutions* (27).

	<i>Pitting temperature (°C)</i>	<i>crevice- corrosion temperature (°C)</i>
<i>Hastelloy C-276</i>	<i>150</i>	<i>80</i>
<i>Inconel-625</i>	<i>101</i>	<i>25</i>

** 4 wt % NaCl + 0.1 wt % Fe₂(SO₄)₃ + 0.01 mol/L HCL; pH = 2.0*

Table 5 : Industrial Titanium Alloys Used in Chemical Process Industries (30).

UNS NO.	ASTM GRADE/ALLOY DESIGNATION	COMPOSITION	CHARACTERISTICS
R50250	Grade 1 (C.P.)	Unalloyed Ti	high formability/ductility, lower strength
R50400	Grade 2 (C.P.)	Unalloyed Ti	good balance of moderate strength and ductility (common/workhorse alloy)
R50550	Grade 3 (C.P.)	Unalloyed Ti	moderate strength
R52400/R52250	Grade 7 and 11 (Ti-Pd)	Ti-0.15% Pd	improved resistance to reducing acids and superior crevice corrosion resistance
R53400	Grade 12	Ti-0.3%Mo-0.8%Ni	reasonable strength and improved crevice corrosion resistance
R56320	Grade 9	Ti-3%Al-2.5%V	medium strength and superior pressure code design allowables
R56400	Grade 5	Ti-6%Al-4%V	high strength and toughness (common/workhorse alloy)
R58640	Beta-C™	Ti-3Al-8V-6Cr-4Zr-4Mo	elevated strength and improved crevice corrosion resistance

Table 6 : Resistance of Titanium Alloys to Crevice Corrosion in Boiling Salt Solutions, Tight metal -to- teflon crevice were tested (30).

SOLUTION	pH	GRADE 2	GRADE 12	GRADE 7
Saturated ZnCl ₂	3.0	F	R	R
10% MgCl ₂	4.2	F	R	R
10% CaCl ₂	3.0	F	R	R
10% KCl	3.0	F	R	R
Saturated NaCl	3.0	F	R	R
Saturated NaCl + Cl ₂	1-2	F	F	R
10% NH ₄ Cl	4.1	F	R	R
10% FeCl ₃	0.6	F	F	R
10% Na ₂ SO ₄	2.0	F	R	R

F = failed; R = resistant