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MATERIALS FOR LIQUID METAL FAST BREEDER REACTORS
VESSELS, PIPING, INTERNALS AND CLADDING

by

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229

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ABSTRACT

Materials selection for liquid metal fast breeder reactor (LMFBR) primary systems including the vessel, piping, heat exchanger, steam generator, core internals, and fuel cladding is reviewed on the basis of a logic chain. Based on this approach the austenitic stainless steels appear to be the most acceptable candidates for first generation demonstration LMFBRs. However, several real or potential problems exist with these or other materials when exposed to the LMFBR environment including temperature, neutron fluence and sodium.

Specific problems discussed are creep - fatigue interactions resulting in reduced component life, sigma or chi phase embrittlement, thermal sensitization, possible stress corrosion, and void formation and swelling of core internals and fuel cladding when exposed to high neutron fluences at proposed reactor temperatures.

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generators are recognized as representing an unique problem in that they are the interface between a sodium and a water system with the probability of stress corrosion cracking if leaks occur between the primary and the secondary sides.

Selection of Materials by a Logic Chain Approach

A previous paper⁽¹⁾ introduced the concept of selecting materials by the use of a logic chain to characterize key parameters and selection factors for vessels and piping. A similar approach will be used in expanded form covering vessels, piping, core internals, and fuel element cladding. It is felt that these components cover the spectrum of problems anticipated in LMFBR systems. Table I summarizes key parameters and selection factors relevant to both demonstration and commercial LMFBRs. While no claim is made that every factor is covered in this logic chain approach, significant factors and parameters are believed to be covered.

Before considering the factors influencing the selection of materials for LMFBRs, it is appropriate to consider a typical plant. Figure 1 is representative of a concept currently pursued in the United States. The contenders for the demonstration LMFBRs favor the

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INTRODUCTION

A comprehensive review of materials problems for liquid metal fast breeder reactors (LMFBRs) is beyond the scope of this paper. In lieu of such a review attention will be devoted to specific problems or unknowns influencing the selection of materials for LMFBR primary and secondary systems.

A logic chain approach will be used to develop the rationale leading to the selection of specific materials for LMFBR components. It becomes obvious in pursuing such an approach that some materials are selected with a definite knowledge that insufficient data are available to permit an optimum design. In fact a very conservative approach must be taken in the case of creep-fatigue interactions.

A parametric approach is used to justify the current selection of materials for first generation LMFBRs. Specific attention will be given to the pressure vessel, piping, core internals, and fuel cladding. In essence problems affecting these components will probably affect pumps, valves, heat exchangers and steam generators. The steam

concept of a pipe system where the piping circulates the sodium. The "Pot" type LMFBR is being constructed in Europe.

Ferritic Alloys

An examination of Table I discloses that the ferritic alloys will probably be limited to use in the guard vessel surrounding the reactor pressure vessel, assuming such is required, or to steam piping. A major limitation to the use of ferritic alloys in the primary system is compatibility. Mass transport of alloy constituents between ferritic and austenitic systems is a recognized problem. For example, a ferritic-austenitic system operating at the proposed temperatures with sodium may undergo mass transport of carbon from the ferrite to the austenite, leading to a gross degradation of the properties of the austenitic components. While carbon stabilization by elements such as niobium is possible, or low carbon ferritic alloys might be used, the risk inherent in using such materials is greater than the gain. Furthermore there may be problems of long term embrittlement of the ferritic alloys or compatibility of other alloy constituents with the austenitic components. Certainly, we cannot consider ferritic pressure vessels at this time

due to the possibility of irradiation damage.

On the water side of the steam generators ferritic piping will undoubtedly be used. There is extensive experience with such piping in fossil fuel plants operating at comparable pressures and temperatures.

Austenitic Alloys

The austenitic alloys currently are the major contenders for most components of LMFBRs. It is anticipated that such alloys as AISI 304 and 316 will be used for the pressure vessels, core internals, piping, and fuel cladding. Analogous cast alloys may be used for the pressure boundaries of pumps and valves.

An analysis of Table I, the logic chain, reveals some real or potential problems. For example, the proposed operating temperatures inevitably, will sensitize the stainless steel. Whether sensitization will be a problem will depend on control of operating conditions in the reactor. Another condition that is an inevitable consequence of reactor operation is the formation of sigma and chi phases at operating temperature. These phases definitely embrittle the alloys; however, most of the loss in notch ductility occurs near room temperature. Since the system should remain sub-

stantially above ambient temperature even under standby conditions, neither sigma nor chi phase formation should prove to be a problem.

The next problem, reduced life due to creep-fatigue interactions, is a very real one; however, the long term consequences of creep-fatigue is not well known. Certainly, there is a definite and deleterious effect on anticipated life, influenced by a complex interaction among such variables as alloy composition, temperature, environment, strain amplitude, and hold-time load. This problem will be discussed separately in a later section of the report.

Another very real problem affecting both cladding and core internals is void formation and swelling that occurs at reactor operating temperatures at fast neutron fluences of 10^{22} to 10^{23} neutron per cm^2 . The mechanisms of swelling and void formation are quite interesting, as are their possible control mechanisms. These will be discussed later.

High Nickel Alloys

The high nickel alloys, particularly nickel-chromium-iron alloys, have been considered for pressure vessels, piping, and cladding in LMFBRs. However, there is no strong expression of interest in their use in the

proposed demonstration LMFBRs.

Some obvious problems with these alloys are the increased cost and greater difficulty to inspect nondestructively by some techniques, compared to the lower nickel austenitic alloys.

Another possible problem with the high nickel alloys is the solubility of nickel in sodium. With more nickel present more is in solution, and the neutron activated nickel increases the level of background radioactivity in the system.

There are some data available that the high nickel alloys are less susceptible to void formation and swelling at high neutron fluences. This will be examined in the context of the general problem of swelling and void formation.

Other Materials

Several alloys are used in minor amounts in the LMFBRs. None appears to represent a significant problem.

One interesting program was the evaluation of vanadium alloys for fuel cladding. It was assumed initially that the body-centered-cubic alloys would be less subject to void formation and swelling. This did not prove to be the case. Indeed most metals, with the

possible exception of titanium and zirconium, or believed to undergo void formation and swelling to some degree at appropriate fluences and of temperatures near $0.5 T_m$ where T_m is the melting temperature in degrees absolute.

Non-Parametric Factors

In selecting materials for LMFBRs such factors as fabricability, inspectability, and cost must be considered. For example, large, relatively thin-walled vessels fabricated of stainless steel or high nickel alloys are not a common item. Their fabrication represents substantial problems, and the ultimate proof of success will be the successful fabrication of several vessels.

A specific example of an unanticipated problem was described by Slaughter and Goodwin⁽²⁾. The authors reported a surprising influence of electrode coating upon creep-rupture properties. For example, welds made with lime-coated electrodes had substantially shorter stress-rupture lives than did welds made with either titania-coated or lime-titania-coated electrodes. This behavior was more apparent in submerged-arc welds than shielded metal-arc welds.

Problems and Unknowns

Some idea of the potential problems influencing materials behavior in an operating LMFBR can be obtained by considering a specific material. Since the 304 or 316 stainless steels represent current choices for essentially all of the components in the primary system of LMFBRs proposed as demonstration plants in the United States, they will be used as examples in examining these problems or unknowns.

It is possible to subdivide problems into three categories: (1) The first will pertain to the pressure boundary where neutron fluxes are low or non-existent, but a variety of dynamic and static loads may occur; (2) A second category includes components such as core internals and fuel cladding that are expected to experience high neutron fluxes and fluences; (3) All components may experience a number of problems in common such as mass transport, thermal embrittlement, sensitization, and possible stress corrosion. These three areas will be reviewed separately.

The Pressure Vessel, Piping, and Heat Exchangers

While the creep-fatigue problem is not limited to materials in the pressure boundary, it probably is most significant there from the design point-of-view.

Essentially the problem is one of uniform loads that cause creep combined with intermittent cyclic loads that cause fatigue. The combined creep-fatigue life is much less than would be predicted from an arithmetic combination of the life under pure creep or pure fatigue conditions.

While there are several programs underway to better elucidate this phenomenon, the data available at this time are relatively limited. A great deal of work will be required before sufficient information is available to permit the designer to predict behavior with some degree of confidence.

The early work of Berling and Conway⁽³⁾ revealed a substantial decrease in the cycles to failure as the tensile hold times increased from 0 to 60 minutes. Figure 2 indicates the cycles to failure may decrease from more than 10,000 to about 600 as hold times increase from 0 to 60 minutes.

Coffin⁽⁴⁾ investigated the effect of frequency on high-temperature, low-cycle fatigue and developed an equation-of-state to predict the life as a function of the nature of the cycles. Figure 3 represents the strain programs used in developing the equation-of-state. In Figure 3a the wave shape can be defined as

$$v = \frac{1}{\gamma_c}$$

where γ is frequency in cycles per unit time and γ_c is the period of the triangular wave shape. In Figure 3b the wave shape is defined as

$$v = \frac{1}{\gamma_c^{-1} + \gamma_H}$$

where γ_H is the time a tensile strain is held and $\gamma_c^{-1} + \gamma_H$ is the total period.

Using this approach Coffin compared actual and predicted lives with hold times for AISI 304 SS as shown in Figure 4.

The work of Achter⁽⁵⁾ and of Coffin⁽⁶⁾ indicates that environment plays a critical role in creep-fatigue life. Tests in air, the conditions of the tests by Berling and Conway⁽³⁾ result in a marked reduction in life. Similar tests in vacuum reveal a markedly different behavior. For example, Achter⁽⁵⁾ cites fatigue tests with type 316 stainless steel at 1500°F (720°C) in air and

vacuum, Figure 5. The effect is even more pronounced in Figure 6, where cycles to failure of Type 316 SS were examined as a function of oxygen pressure over a range of 10^{-7} to 10^5 Torr⁽⁷⁾.

Coffin⁽⁶⁾ investigated A-286, a precipitation hardening austenitic alloy, to determine the plastic strain range versus fatigue life in air and vacuum at 1100°F (590°C). Figure 7 presents the data as functions of frequency. The regression analyses for frequencies of 10, 1, 0.1 and 0.01 cycles per minute reveal definite sensitivity to frequency in air while no change is observed over a range of cycles when tests were conducted in vacuum.

An attempt has been made by the American Society of Mechanical Engineers (ASME) in Code Case 1331-6⁽⁸⁾ to develop a conservative rule for handling combined creep - fatigue interactions. This is shown in Figure 8; it is apparent that there is a drastic reduction in design life under the worst set of creep and fatigue conditions.

While the creep-fatigue problem is a potentially serious one, we know very little about the behavior of

stainless steel in inert environments. Tests must be run in sodium and nitrogen to determine if the effects in these environments are similar to, better than or worse than that observed in air.

A number of programs are underway in the United States and other countries to assess the significance of this problem and to develop necessary design data.

Reactor Core Internals and Fuel Cladding

In essence reactor core internals, including fuel cladding, experience the same environment as other portions of the primary system with one notable exception; the neutron fluxes are much higher than seen by the vessel, and fluences approaching 10^{23} neutrons per cm^2 are necessary in the fuel if the reactor is to be economically viable. An anticipated consequence of high neutron fluences at operating temperatures will be a gross loss in ductility, both in uniform and in total elongation. Values may drop to 0-2 per cent so that combined thermal and bending loads might fail internal components unexpectedly. An example of neutron induced loss of ductility is given in Figure 9⁽⁹⁾.

A more serious problem is the void formation and swelling that occurs in stainless steels and other metals, at neutron fluences of 10^{22} - 10^{23} neutrons per cm^2 . An

example of voids is given in Figure 10⁽¹⁰⁾.

The degree of void formation and swelling is believed to be a complex function of temperature, fluence, alloy composition, and prior fabrication history. Shewmon⁽¹¹⁾ elegantly summarized the current void formation model; briefly, voids are attributed to the agglomeration of vacancies with helium formed by (n,α) reactions stabilizing the voids. Temperature is a critical parameter. Results indicate swelling is limited to a range of 0.3 to 0.5 T_m where T_m is the melting point in degrees Kelvin or Rankine. This is illustrated in Figure 11.

It is apparent that the number of sinks per unit volume is a significant factor in the rate of swelling. These sinks affect the variables in Shewmon's⁽¹¹⁾ equation

$$\alpha \phi = \beta D_i N_v + \gamma_v P_s N_v D_v$$

ϕ = neutron flux

$\alpha\phi$ = production rate of vacancy interstitial pairs

β, γ = constants of proportionality

D_v and D_i = diffusion coefficients for vacancies & interstitials

N_v and N_i = average vacancy or interstitial concentration

P_s = concentration of sinks

Below $0.3 T_m$ recombination is the limiting factor; above $0.5 T_m$, the vacancy supersaturation is inadequate to sustain voids.

From an engineering point-of-view we must accept the fact that LMFBR operating temperatures fall between $0.3-0.5 T_m$. Therefore, it is necessary to see what can be done to minimize the amount of swelling at anticipated maximum fluences. The magnitude of the swelling problem in annealed stainless steel is apparent in Figure 12.

Swelling of internals must be minimized due to the close dimensional tolerances necessary in the core of an LMFBR. The problem may be accentuated by variations in flux leading to differences in swelling rate and bowing of components. It becomes an engineering and economics problem to decrease the swelling at the higher fluences. For example, a factor of two decrease in swelling is worth about half a million U.S. dollars per year for a 1000 MW(e) LMFBR.

The logical point of attack is in the concentration of sinks, in the metal. We can influence sink concentration by cold-work, special heat treatments, or by minor changes in composition. All of these approaches have been investi-

gated with some success. For example commercially pure nickel will sustain ten times more fluence in reaching the same per cent swelling as high-purity nickel, indicating the role of trace elements. Another example of trace element effects is in 304 SS plus 0.2% titanium. In solution the titanium reduces swelling while there is little or no effect if the titanium is precipitated prior to irradiation.

Cold work and heat treatment will decrease swelling substantially. For example 10-20% cold work in a stainless steel markedly reduces the swelling. An example of design curves is given in Figure 13, illustrating the gain that can be achieved by appropriate treatment.

While steps have been taken to control swelling, we still lack the fundamental information necessary to permit quantitative prediction of the dependence of swelling on fluence, stress, temperature, and prior and existing metallurgical condition. We know that substitutional and interstitial elements both affect swelling as does gamma-prime phase in nickel-base alloys. Cold work helps, but its effect varies with alloy chemistry and irradiation conditions. The effects of imposed stress or varying stress is unclear at this time.

While we lack the information necessary to quantify the fundamental mechanism, it is still possible to establish conservative engineering design limits, based on available data; however, more data will be required before these limits can be expanded to include higher fluences, particularly due to the possibility of subtle differences in mechanical working heat treatment, and composition.

Problems Common to All Components

Some problems such as mass transport of elements, thermal embrittlement, sigma and chi phase formation, sensitization, and possible enhanced local fatigue-corrosion or stress-corrosion may prove to be relatively insignificant or may be critical. In most instances the potential for damage exists; however, the environmental conditions may not favor failure of the component or components. A few examples will be given of such cases.

It is virtually certain that the stainless steel components will be severely sensitized shortly after reactor startup since the proposed operating temperatures ($\sim 1100^{\circ}\text{F}$ or 600°C) maximize thermal sensitization. Whether this sensitization will be a problem will depend on control of the gaseous environment, particularly moisture content; however, moisture is not the only concern. It is possible

that sufficient oxygen will exist as an impurity in the nitrogen atmosphere to accelerate intergranular cracking.

Certainly the external environment surrounding the primary and secondary systems must be carefully monitored so that stress corrosion does not occur.

In the steam generator the situation is even more critical. Any leakage will probably be from the steam side into the sodium, forming concentrated sodium hydroxide. This concentrated hydroxide will stress corrosion crack most of our steam generator materials, including low alloy steels, austenitic stainless steels, and the high nickel-chromium-iron alloys. This condition occurred in the Nuclear Submarine Sea Wolf superheaters; it also has occurred in a steam generator containing duplex components - stainless steel on the sodium side and Inconel-600 on the steam side. In-leakage of steam caused an environment that led to severe cracking of stainless steel tube sheets and tubes.

Since an LMFBR is expected to operate at 1000-1200°F (540 - 650 C), the formation of sigma and chi phase virtually certain. These phases severely embrittle stainless steels at or near room temperature. Since these units should never get within about 200°F of room

temperature, this sigma phase formation should be no problem. One cautionary note is necessary. If the system were to be drained for some reason and component temperatures approached room temperature, definite precautions will be required in the handling of components.

The third problem, mass transport, should be considered carefully. Several factors may play a role in mass transport including impurity level and impurity type in the sodium, chemical composition of the various components in the primary system, possibility that components are markedly dissimilar in chemical composition such as 304 SS and a low alloy ferritic steel, and the normal thermal gradient that exists in a system. An element whose mass transport is of particular concern is carbon. In one instance where stainless steel was present in the same loop with low alloy steel, mass transport of carbon from the low alloy steel to the stainless steel increased the carbon in the stainless steel to several per cent, sufficient to form a brittle two phase alloy.

While mass transport is not expected to be a major problem in systems constructed of the same alloy, we still do not know enough to predict confidently that mass transport will not be a problem. More test results

coupled with operation of nuclear LMFBRs are necessary before this bogey can be laid to rest.

SUMMARY

The austenitic stainless steels appear to be the most satisfactory materials for the current generation of LMFBRs. Even so we lack sufficient data on creep-fatigue interactions, void formation and swelling, and mass transport to be able to unequivocally predict behavior throughout design life.

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TABLE I

LOGIC CHAIN - SELECTION OF MATERIALS FOR LMFBR VESSELS AND PIPING (PRIMARY ENVELOPE)

A. KEY PARAMETERS

(T) TEMPERATURES (750-1150F) (400-620C)

(t) TIMES 20-40 YEARS

(L) LOADS - THERMAL, STATIC, DYNAMIC

(ϕ) VESSEL NEUTRON FLUENCE $> 10^{20}$ nvt

(E) ENVIRONMENT - SODIUM, HELIUM, NITROGEN

(C) SYSTEM COMPATIBILITY - CHEMICAL, DIMENSIONAL

B. SELECTION FACTORS

FROM PARAMETERSCREEP (T,t,L, ϕ ,E); FATIGUE (T,t,L,E,C)

CREEP-FATIGUE INTERACTIONS

EMBRITTEMENT (T,t, ϕ ,E

MASS TRANSPORT (T,t,E,C)

DIMENSIONAL CHANGES (ϕ ,T,t)NON-PARAMETRIC

FABRICABILITY

INSPECTABILITY (INITIAL AND CONTINUING)

COST

) LOW CYCLE(THERMAL)
) HIGH CYCLE(VIBRATIONAL)

)(IRRAD.,THERMAL)

FUNCTION OF MATERIALS

FUNCTION OF MATERIALS

C. POTENTIAL MATERIALS

LOW ALLOY FERRITIC MATERIALS (2-1/4 Cr-1 Mo, etc.)

AUSTENITIC ALLOYS (304, 316, 316 L)

HIGH NICKEL ALLOYS (INCONEL-600, etc.)

D. ADVANTAGES AND DISADVANTAGES
(MAJOR PROBLEMS OR MAJOR UNKNOWN)LIMITING FACTORS

1) 2-1/4 Cr - 1 Mo (Stabilized or Low Carbon

PRO	PIPING	CON
COST	OR	
AVAILABILITY	GUARD	MASS TRANSPORT A PROBLEM
FABRICABILITY	VESSEL ONLY	
BACKGROUND INFO.		LONG TERM COMPATIBILITY
HI TEMP. MECH. PROPERTIES		

2) 304 or 316

PRO	VESSEL	CON
AVAILABILITY	&	
FABRICABILITY	PIPING	LONG TERM EMBRITTEMENT SIGMA OR CHI PHASE
BACKGROUND INFO.		
HI-TEMP. MECH. PROPERTIES		
Na-COMPATIBILITY		
RELATIVE RESISTANCE TO IRRADIATION EMBRITTEMENT		SENSITIZATION THERMAL FATIGUE IRRAD. SWELLING

3) INCONEL-600

PRO	VESSEL	CON
HI TEMP. PROPERTIES	&	
	PIPING	COST
		INSPECTABILITY

FIGURE 1. GENERAL ARRANGEMENT OF A LIQUID METAL FAST BREEDER REACTOR - THE FAST FLUX TEST REACTOR

FIGURE 2. EFFECT OF HOLD TIMES ON LIFE. AISI 304 STAINLESS STEEL - 650°C AFTER BERLING AND CONWAY

(b)

(a)

FIGURE 3. STRAIN PROGRAM (a) TRIANGULAR WAVE FORM
(b) TENSILE HOLD

FIGURE 4. COMPARISON OF ACTUAL AND PREDICTED LIFE
WITH HOLD TIMES - AISI 304 STAINLESS STEEL 650°C

FIGURE 5. FATIGUE OF TYPE 316 STAINLESS STEEL AT 1500 °F (815 °C) IN AIR AND IN VACUUM TESTED IN REVERSED BENDING (5 CPS)

FIGURE 6. EFFECT OF OXYGEN PRESSURE ON FATIGUE LIFE OF 316 STAINLESS STEEL AT 500°C AND 800°C

FIGURE 7. PLASTIC STRAIN RANGE VS FATIGUE LIFE FOR A286 IN AIR AND VACUUM AT 1100°F. NUMBERS ADJACENT TO TEST POINTS INDICATE FREQUENCY IN CPM. SOLID LINES ARE REGRESSION ANALYSIS OF EQUATION:

$$\sum_i \left(\alpha \frac{n}{N_d} \right)_i + \sum_j \left(\beta \frac{t}{t_r} \right)_j \leq K_e$$

$$\alpha \approx 2.00$$

$$\beta \approx 3.00 \text{ (STRESS LIMITED)}$$

$$\beta \approx 1.50 \text{ (STRESS LIMITED)}$$

FIGURE 8. LIMITING VALUES OF CREEP/FATIGUE DAMAGE

FIGURE 9. LOSS IN TOTAL ELONGATION OF AUSTENITIC STAINLESS STEELS AS A FUNCTION OF TEMPERATURE AND LEVEL OF IRRADIATION AND OF TENSILE TESTING TEMPERATURE. TESTS ON UNIRRADIATED SPECIMENS AT SAME TEMPERATURE CITED FOR IRRADIATED. ELONGATION ON UNIRRADIATED MATERIAL 5-10% ($> 600^\circ\text{C}$), 40-50% ($< 300^\circ\text{C}$). FRACTURES - IRRADIATED-INTERGRANULAR; UNIRRADIATED-TRANSGRANULAR.

FIGURE 10. VOID FORMATION IN 316 SS IRRADIATED AT 400°C
TO 3×10^{22} n/cm² (E 0.1 MeV). SWELLING
ABOUT 0.8 PERCENT. MAGNIFICATION IN NANOMETERS (nm)

FIGURE 11. SWELLING AS A FUNCTION OF TEMPERATURE
IN ANNEALED TYPE 304 STAINLESS STEEL

FIGURE 12. SWELLING AS A FUNCTION OF FLUENCE IN ANNEALED TYPE 304 STAINLESS STEEL IN THE 370-550°C TEMPERATURE RANGE

FIGURE 13. IRRADIATION INDUCED SWELLING IN AUSTENITIC STAINLESS STEEL