

Critical questions in materials science and engineering for successful development of fusion power

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Abstract

It is the general conclusion of all national programs that the development of high-performance reduced-activation structural materials is essential for the successful development of fusion power. In this paper, the experience gleaned from previous programs to develop materials for high temperature structural applications is used to identify and discuss some of the most critical issues that must be addressed in the development of candidate materials for fusion structural applications. Critical issues discussed include radiation-induced solute segregation and implications on phase stability in the development of high-performance alloys/ceramics; the effects of very large amounts of helium on mechanical properties and the implications for alloy design/development; development of high temperature design methodology and incorporation of radiation effects into this methodology; the effects of radiation damage on flow localization, and the implications and approach to control the phenomena; and considerations of mass transfer and corrosion in complex fusion systems. © 2007 Elsevier B.V. All rights reserved.

1. Introduction

Increased energy consumption [1] and the desire to decrease carbon-dioxide emissions have both spurred interest in alternative and cleaner energy sources. Alternative energy sources must be available in the latter half of this century to meet the increased demand and, at the same time, reduce greenhouse gas emissions. Fusion energy is an attractive option as it offers the potential of a sustainable and abundant power source with no green-

house gas emissions, no long-term radioactive waste, and no risk for a severe accident.

The current fusion strategy is designed to demonstrate the potential for fusion energy in the next 35 years, primarily through the International Thermonuclear Experimental Reactor (ITER) facility. Although ITER required development and validation of several new materials and technologies, many of the technologies needed to demonstrate the engineering feasibility of fusion energy will not be tested in ITER due to its limited planned duty cycle. Commercial fusion power reactors will be much more complex than ITER and will require materials with excellent physical and mechanical properties as well as the stability of these properties in the challenging fusion operating environment. In particular, the

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goals for fusion power systems place significant demands on the performance of structural materials, including safety, the minimization of radioactive waste, and increased economy (through high thermal efficiency, longer lifetimes and increased reliability). After more than two decades of research only three candidate materials systems appear to have the potential to meet the low-activation, high-performance goals: 8–9Cr ferritic/martensitic steels (including the reduced activation variants), SiC/SiC composites and V–Cr–Ti alloys [2,3].

While these materials are fairly well understood, considerable gaps in the databases still exist (most notably the need for experimental data in a close simulation of a fusion environment) [2]. As research moves forward and these materials enter new performance regimes (temperature, dose, He, etc.), new phenomena may be found which may slow the development of fusion energy. Indeed, unexpected material behavior can cause major disruptions in a development program. For example, the first open literature report [4] of void formation during neutron irradiation raised concerns about swelling which dramatically slowed the development of the liquid metal fast breeder reactor program (LMFBR). It required nearly a decade to fully understand the phenomenon of swelling and another decade to develop materials with satisfactory performance.

Obviously, it is imperative to avoid such delays in the development of fusion energy. It is also very informative to look back at previous large scale breeder reactor programs to identify lessons which may be applied to the development of materials for fusion power reactors. The objective of this paper is to examine several critical materials issues identified in previous fission breeder reactor programs which, looking forward to fusion, may be important to consider and examine. These issues include radiation-induced solute segregation and implications on phase stability, the effects of very large amounts of helium on mechanical properties, the effects of radiation damage on flow localization, mass transfer and corrosion in complex fusion systems, high temperature design methodology, and the incorporation of radiation effects into this methodology.

2. Radiation-induced segregation and phase stability

Radiation-induced solute segregation (RIS) refers to the redistribution of elements (solute or

interstitial impurities) under irradiation. While this form of microstructural change was one of the last major radiation-induced changes to be experimentally observed, it was predicted theoretically before actually measured. Anthony [5] in 1972 predicted non-equilibrium RIS based on the diffusion behavior of particular atoms in complex alloys by considering several possible interactions with the excess irradiation-induced vacancies or interstitials diffusing to defect sinks [6]. Any preferential association between an atom and one type of defect will result in enrichment or depletion of that atom at defect sinks such as grain boundaries.

RIS has been examined and measured in detail for decades in austenitic stainless steels. In light water reactors (LWR), stainless steels experience Cr depletion at grain boundaries, which, in turn, can impact corrosion resistance and lead to irradiation-assisted stress corrosion cracking [7]. While water corrosion is not expected to be a problem in ITER due to the relatively low temperatures (<250 °C) and low cumulative damage levels (<3 dpa), RIS can lead to areas with local concentrations that are significantly different than the bulk material and phase stability can become a concern. In particular, the formation of non-equilibrium gamma, gamma prime and G phase have all been observed in 316 stainless steels. Phase stability is a key concern as changes in alloy microstructure can influence mechanical properties, deformation mode, and corrosion behavior.

Radiation-induced precipitation (RIP) is only one example of phase instability under neutron irradiation in complex alloys like austenitic stainless and martensitic/ferritic steels [8]. Other effects include radiation-modified, -enhanced, or -retarded thermal precipitation [9–12].

Prior reviews detail effects of both temperature [8] and dose [13] on microstructural evolution in 300 series austenitic stainless steels. While RIS and RIP are observed in the 250–300 °C range, they can be quite pronounced at 400–600 °C, particularly in ‘easy swelling’ materials like solution-annealed (SA) 316, 316 + Ti or the various advanced austenitics with 15Cr/15Ni + Ti or 14Cr/16Ni + Ti (a legacy alloy from the US Fusion Materials Program in the later 1970s, termed the prime candidate alloy).

Helium effects were also fairly straight forward in such alloys, with higher He/dpa ratios simply increasing void nucleation, and in many alloys, increasing Frank loop nucleation and accelerating the RIS/RIP associated with voids and loops,

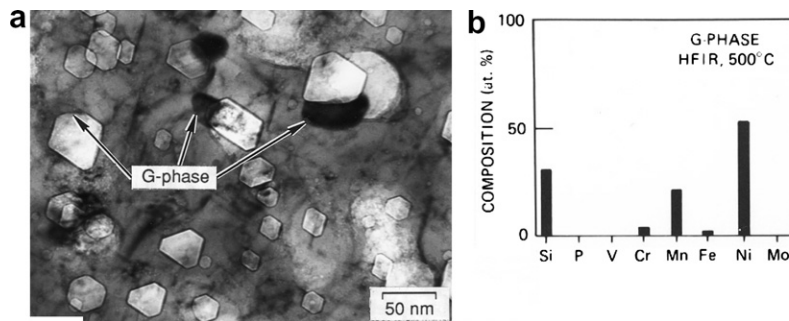


Fig. 1. SA PCA steel irradiated at 500 °C in ORR to 11 dpa (200 appm He), showing the association between (a) radiation-induced G-phase silicide ($\text{Mn}_6\text{Ni}_{16}\text{Si}_7$) particles and the largest voids and (b) XEDS phase composition from analysis of such particles on extraction replicas. Such co-development of void/RIP couples is a direct consequence of both RIS at the void sink and the collector effects that enhance void growth at precipitates [8].

particularly at 400–500 °C [8]. The coupled effects of RIS and RIP at vacancy-biased sinks like voids are shown for the RIP G-phase silicide in Fig. 1. In this case, G-phase develops in the RIS ‘micro-alloying’ region that surrounds the void, and then in turn enhances void growth as the particle forms and grows.

In addition to the G-phase silicide shown in Fig. 1, many other types of phases have been observed in irradiated 316 stainless steels, including carbides, laves, gamma, and various combinations of those samples. The extent of precipitation and type of precipitate are extremely sensitive to the exact operating conditions (temperature, dose, dose rate, and He/dpa ratio), but are also dependent on the details of the damage microstructure.

RIS and RIP may also play an important role in fusion reactor materials and conditions. Measurements of RIS in ferritic/martensitic alloys are very difficult due to the magnetic nature of the samples; however, segregation has been measured [14]. There are only limited known studies of segregation in irradiated V–Cr–Ti alloys or in SiC materials, but clearly this must be done for all candidate fusion materials. In V–Cr–Ti alloys, Ti-enrichment has been observed by Rice and Zinkle [15], which may be a precursor to Ti-rich precipitates [16]. In SiC, segregation raises concerns regarding phase stability associated with the departure from stoichiometric Si–C compositions which can also arise from the preferential transmutation of Si in a fusion neutron spectrum (and resulting segregation of transmutant solutes) [17].

If fusion energy is to continue to develop, predictive models of segregation in candidate materials must be developed. These models must eventually be incorporated into comprehensive models for irra-

diation-induced structural evolution. This involves using existing RIS models (e.g. modified-inverse Kirkendall model developed by Allen [18] for austenitic stainless steels) which must be modified to account for solubility limits and precipitation. However, the development of accurate models for RIS in other alloy systems and/or crystal structures relies on knowing the diffusivities for all elements, point-defect solute interactions and solubility limits, which must be determined either experimentally or from first principles.

3. Helium effects/embrittlement

A major focus of fusion materials research and development must be the effect of He on the microstructural stability and properties of materials used in the first wall blanket and diverter regions. Helium concentrations for demonstration plants will reach levels that are orders of magnitude above those encountered in other nuclear power systems [2]. Indeed, as Bloom et al. also note, void swelling may reach a maximum near fusion-relevant operating conditions [2]. However, helium embrittlement may also be a key form of degradation.

High temperature helium embrittlement has been known and examined for some time [19–21]. The conventional view is that He-embrittlement is a high temperature phenomenon for temperatures over half the melting point. This mode of failure is also typically associated with slow strain rates and is caused by the stress-induced growth of He bubbles along grain boundaries. Indeed, the degradation of creep-rupture properties and ductility can be severe and has been observed in numerous pure metals as well as austenitic stainless steels and vanadium alloys. As noted by Bloom et al. [2], He transmutation

products can be effectively managed (by trapping with engineered nanoparticles or other sinks), but this also presents other challenges as the processes and microstructural stability are strongly affected by the He/dpa ratio.

There is, however, additional evidence that He embrittlement can occur at lower temperatures (i.e. $<T_m/2$). Intergranular fracture has been observed in 304 stainless steel irradiated at 370 °C to 1×10^{23} n/cm² and then tested in vacuum at 550 °C [22]. More relevant to fusion, intergranular fractures have also been recently observed in T91 steels implanted with high-energy He ions at low temperature [23]. Although He is affecting the fracture processes, it is most likely not associated with stress-induced bubble growth at such low temperatures.

Another example of He embrittlement and the effect of irradiation on ductility is shown in Fig. 2. Here, the total elongation during creep-rupture testing of annealed 316 SS is plotted as a function of test temperature [24] and irradiation (the upper band indicates the unirradiated material trends). The data show very low ductility at 550 °C which rises to a maximum at 650 °C before falling again at 750 °C. This behavior suggests that there could be a synergistic effect of RIS and He acting together at grain boundaries. At lower temperatures, RIS is at a maximum, drawing more He to the boundaries. As temperatures increases, the degree of RIS decreases along with the amount of grain boundary

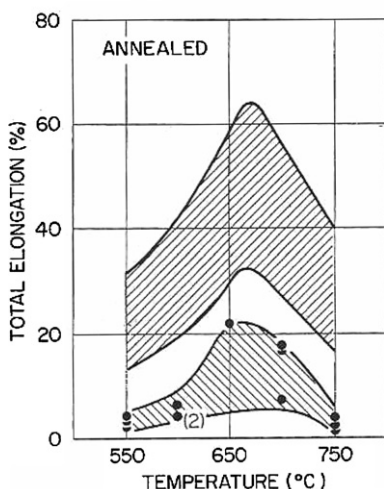


Fig. 2. Effect of irradiation on the creep-rupture ductility of Type 316 stainless steel [24]. The upper band represents the data trend for the unirradiated material while the lower band and data points are for irradiated specimens.

He. However, at even higher temperatures, traditional He embrittlement is again dominant. While plausible, this possible connection between RIS and He must be further evaluated.

Although He embrittlement is well known from breeder reactor programs, this form of failure is still a key concern for all fusion materials and must be better understood in order to develop viable mitigation strategies. This can be achieved only through coordinated experiments and through theory and modeling over the entire fusion-relevant range of interest for temperature and He contents.

4. Localized deformation

The evolution of radiation-induced microstructure can have a profound time- or damage- level dependent influence on deformation mode. The interaction between small loops and moving dislocations may result in very inhomogeneous deformation, through either twinning or dislocation channeling. In the channeling process (typically for irradiation temperatures less than $0.3T_m$), the initial, moving dislocations encounter a dislocation loop (or void or other defect cluster), which acts as a barrier to slip. To overcome this barrier, dislocations annihilate or combine with the defects on the slip plane and continue to glide. Subsequent dislocations will tend to glide along this same path, clearing out additional defects resulting in a channel that is free of defects. The areas in between the channels usually remain untouched, so all of the deformation in the material is concentrated in these channels, resulting in highly localized deformation. These channels have been observed in a variety of pure metals and engineered alloys following both neutron and ion irradiation [25–29]. An example of a channel in neutron-irradiated vanadium is shown in Fig. 3 along with a schematic deformation map indicating the regions in strain-rate-temperature space where localized deformation is likely to occur.

Ultimately, channeling will result in the highly localized pile-up of dislocations at distinct locations along a grain boundary. While the total strain for a sample may be relatively low, virtually all of that strain is accommodated within the channels resulting in a very low uniform elongation. This localized deformation may be important in fracture as the intersection of channels and grain boundaries may lead to cracks, as modeled and discussed by Das and Marcinkowski [30]. The localized deformation

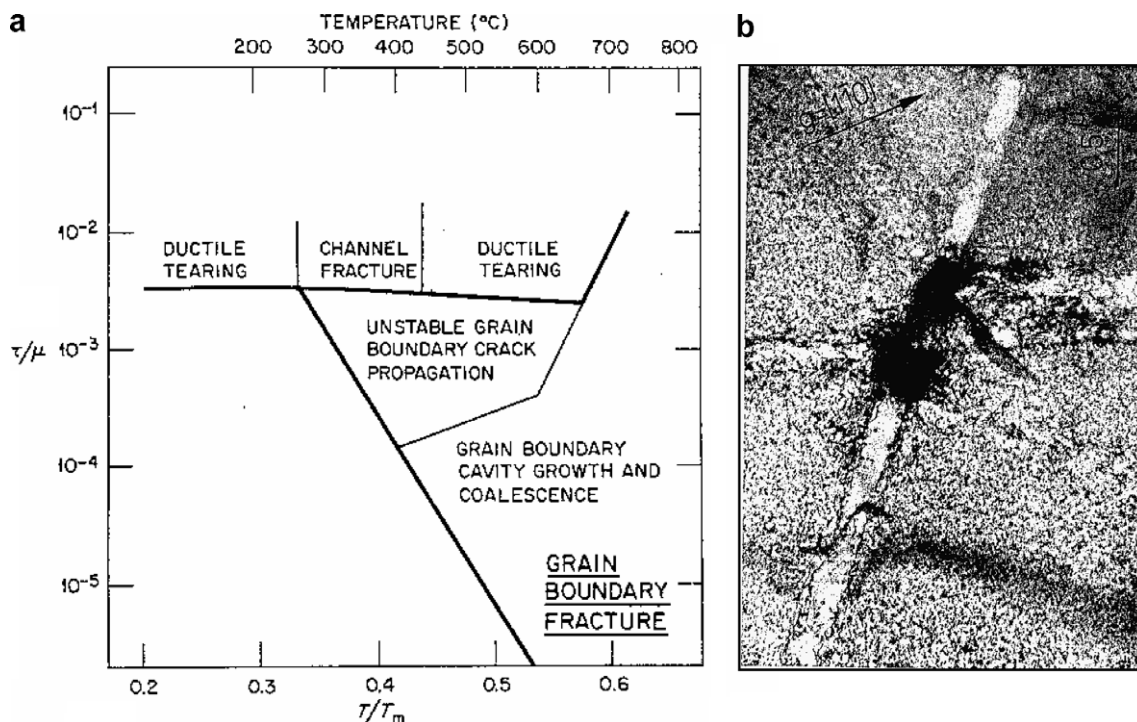


Fig. 3. (a) Schematic representation of the stress and temperature dependence of the fracture behavior of Type 304SS after irradiation to high fast neutron fluences. Ordinate scale is the ratio of shear stress to shear modulus [33]. (b) Dislocation channels in vanadium irradiated to 8×10^{19} n/cm² at 380 °C and deformed in tension. Reproduced from [25].

leads to a mismatch at the grain boundary; called a 'deformation ledge'. In the case of localized deformation and channeling, dislocations pile-up at the grain boundary at only a small number of locations. This localized strain at the grain boundary must be accommodated or grain boundary failure may occur [31,32].

This mode of failure was observed early in the breeder reactor programs. As shown in the fracture map in Fig. 3(a), this mode of failure is highly dependent on the irradiation and test temperature, the stress-state and the irradiation dose [33]. Experimental evidence supporting this mechanism has been observed in different irradiated alloys. Steps associated with channel deformation were observed on grain boundary fracture surfaces in irradiated type 304 by Clausung and Bloom [34]. Onchi et al. [35] in a recent study also found distinct ledges on IG facets and linked deformation banding for stainless steel irradiated under light water reactor conditions.

A key step forward is characterizing and understanding the extent and development of localized deformation. This involves accurate measurement of the extent of localized deformation (slip step

height and spacing, channel width, or more importantly, strain within the channel). The characterization of localized deformation in the key alloys and components over a fusion-relevant range of temperatures and doses is especially critical. This detailed characterization will also greatly enhance the modeling and prediction of the development of localized deformation under service conditions.

5. Corrosion in liquid metal coolants

A number of different coolants have been proposed for fusion reactors, including water, PbLi, Li, and He. Because of their excellent heat transfer properties, liquid metals have long been considered as coolants for high power-density reactor systems and research in support of reactor technology has greatly influenced the understanding of the corrosion of structural materials by liquid metals. In particular, the worldwide efforts conducted during the development of LMFBRs (with a molten sodium coolant) have greatly contributed to the knowledge base related to liquid metal corrosion and mass transfer from both scientific and technological perspectives [36–38]. As such, reactions of

materials exposed to molten sodium have been studied in greater detail than any other liquid metal, particularly under conditions representative of heat transfer (flow velocity, temperature gradients, heat flux) and, in many ways, the LMFBR work served as a basis of reference for later studies of the effects of lithium on ferrous alloys conducted in support of fusion technology in the later 1970s and 1980s. (Although lithium corrosion studies date to the 1950s or earlier, most of this work was devoted to refractory metals or other high-temperature materials at much higher temperatures than have been considered in fusion technology over the past 25–30 years, see, for example, [39].)

In the broadest sense, corrosion-related reactions between solids and liquid metals can be categorized as either dissolution, impurity and interstitial transfer, alloying between liquid metal coolant, or solid or compound reduction [40] (although each type is not necessarily independent of the others). Under non-isothermal conditions (e.g. in heat transfer systems), dissolution of material in one region can lead to mass transfer and deposition of solute in other parts of the system (typically at lower temperature) [41]. Studies with sodium in support of LMFBR concepts led to a more thorough characterization of the factors affecting dissolution [42–44] and modeling of mass transfer [36,37,45].

While the scientific and technology knowledge bases for liquid metal corrosion were greatly expanded by the development of LMBFR, fusion reactors will have their own particular compatibility issues. Specifically, fusion reactors will use lithium or lead–lithium as a coolant rather than sodium (as lithium will also breed tritium under irradiation). While the general mechanisms remain the same, the specific reactions that control dissolution, deposition, and impurity reactions in lithium (and lead–lithium) are quite different than in sodium, as first addressed in some detail by DeVan and Bagnall [46]. For example, lithium and sodium have important differences with respect to the effects of impurities such as nitrogen, oxygen, and carbon on corrosion and mass transfer behavior. Such differences between sodium and lithium have become clearer in the years since the DeVan and Bagnall review as work in the 1980s progressively revealed the details of the roles of nitrogen and carbon on controlling corrosion and mass transfer in lithium–steel systems [47–49], whereas oxygen is the most important impurity element in sodium [42,44,46, 50–52].

Just as lithium and sodium differ in the specifics of the reactions that control corrosion and mass transfer and determine the effects of temperature, flow velocity, microstructure, etc. on such, lead–lithium and lithium often present differing critical issues depending on the specific blanket concept and materials. The low melting point (235 °C) eutectic composition of Pb–17 at.%Li is sufficiently dilute in lithium that the molten alloy acts more like pure lead. Corrosion and mass transfer in lead has been studied in support of reactor coolant technologies [52–55]. In general, for the principal elements of steels, dissolution and mass transfer is greater in lead than in lithium [56]. While high levels of oxygen in the lead–lithium can inhibit dissolution (at least initially) by formation of protective oxide surface layers [56], lithium is highly reducing such that most oxides are unstable in contact with it. This not only precludes the use of oxide layers for corrosion protection, but makes it very difficult to develop stable electrically insulating layers on alloys to reduce the magnetohydrodynamic (MHD) force exerted by lithium flowing through a magnetic field [57,58]. If the high temperature capabilities of V–Cr–Ti alloys (~700 °C) [59], including its good resistance to corrosion and mass transfer, are to be utilized in a reactor design, then a corresponding method for reducing the MHD pressure drop must be developed for this system. However, there are relatively few potential insulating material compositions that could serve as an MHD insulating layer and be stable in the lithium environment [57]. The most promising candidates, Y_2O_3 and Er_2O_3 , both show degradation in static lithium tests at 600–800 °C [60,61]. Thus, the most likely alternative either employs a flow channel insert, made up of V foils with an oxide inner layer [62] or a multi-layer coating with alternating layers of vanadium and oxide.

Assessing this flow insert concept remains one of the critical steps forward for the use of Li or Pb–Li in fusion reactors. However, other questions must also be addressed. A more fundamental understanding of the controlling reactions of SiC and SiC/SiC composites in Pb–Li must be developed (including non-stoichiometric effects), in addition to a better understanding of liquid metal chemistry and the stability of corrosion resistant coatings on ferritic alloys. Mass transfer tendencies for SiC and coated ferritic steels in flowing Pb–Li must also be examined. Finally, with this knowledge, well-established models of mass transfer in liquid metals (and flowing He) must be applied to candidate blanket

designs using finite element modeling to determine temperature limits, burdens, and system lifetimes.

6. High temperature design methodology

While each of the issues discussed above presents a challenge for fusion materials, they also pose a significant challenge for designers when combined. A key requirement in the design process of any power system is the use of proven, conservative design criteria. ITER will operate at temperatures much lower than those required in future fusion power reactors ($< \sim 300$ °C for ITER versus ~ 550 °C for fusion power reactors). The current design rules for ITER (ITER Structural Design Criteria (ISDC) [63]) use similar low-temperature design codes of the member nations, including Europe [64], Japan [65], Russia [66], and the US [67]. Rules in the established criteria are based on a limit analysis of plasticity, which assumes that the structural materials have unlimited ductility relative to the strain limits [68]. The balance of plant (pumps, motors, etc.) will be regulated by non-nuclear design procedures and criteria, but these should also be evaluated for fusion reactor applications.

Design procedures often include ‘simplified design methods’ for use in nearly all stages of design, but are far from simple as they are conservative criteria derived from and bounded by more complex and rigid solutions. As a result, these ‘simplified design methods’ are often used as they are more practical, especially at the conceptual design level. The use of analytical solutions are also invaluable as they provide information on trends, permitting the designer to more effectively assess the effects of material, geometry, and loading changes prior to final design analysis. This is particularly true when designing structures at elevated temperatures where creep and irradiation are significant. Integration of design procedures and material behavioral changes due to irradiation and environment is critical in arriving at effective design tools.

The impact of irradiation will clearly cause changes in mechanical properties over the lifetime of the fusion reactor. However, there are no established high temperature design codes which fully incorporate irradiation effects for fusion environments. The current ISDC [68] does attempt to account for irradiation embrittlement by implementing two additional design rules to protect against failure by plastic flow localization and local exhaustion of ductility. The ISDC has also estab-

lished acceptable deformation limits resulting from thermal creep, irradiation-induced creep, or irradiation-induced swelling. However, each of these factors must be fully characterized in fusion-relevant environments to properly incorporate their effects into final design criteria. Some guidance on high temperature design methodology can also be taken from the Prototype Large Breeder Reactor (PLBR) [69] where some basic design limits were established for fuel assemblies, although many of these criteria for fuels are not directly applicable to a fusion reactor.

Clearly, irradiation, creep-fatigue, and environmental effects must be taken into account in high temperature design. However, the current design criteria for creep-fatigue are not very well founded, with the linear damage rule or a bilinear damage rule being employed due to a lack of more suitable physics-based criteria (see Fig. 4). Creep-fatigue damage is also material-specific, and may be influenced by the environment, hold time, and loading rate. Studying and documenting the effects of various loading conditions (static and cyclic) during exposure to neutron irradiation is critical; design procedures must address these issues properly. A limited number of studies have shown a disagreement between deformations that occur within the reactor and tests conducted on post-irradiated materials. For example, some in-reactor tensile tests have demonstrated extended rupture times compared to post-irradiation tests [70,22]. Fig. 5 illustrates the extended rupture life of biaxial creep samples during exposure to irradiation as compared to unirradiated and post-irradiated samples [71]. The dramatic increase in rupture life is attributed to short-lived point defects generated during

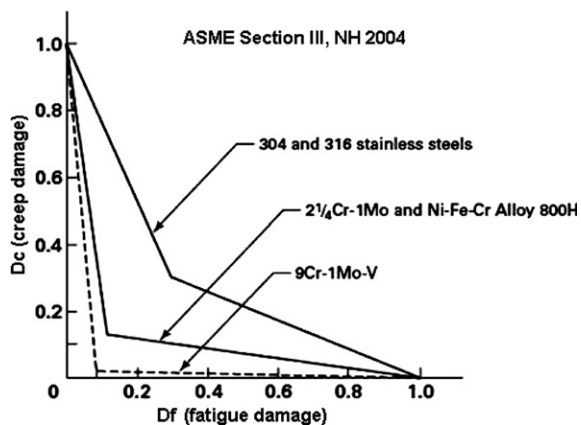


Fig. 4. Bilinear creep-fatigue damage for various materials.

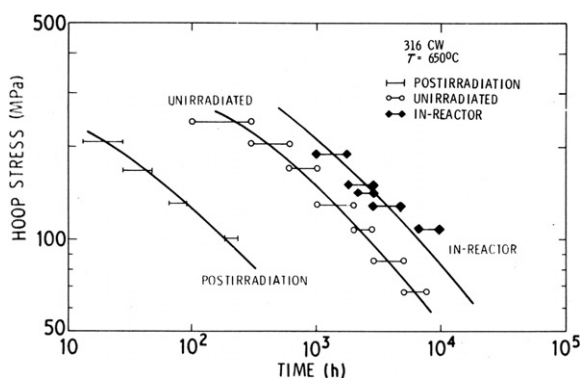


Fig. 5. Rupture behavior of post-irradiation, unirradiated, and in-reactor specimens [71].

irradiation in a process termed ‘dynamic hardening’. These defects are likely annealed out during elevated temperature post-irradiation testing, but constantly replenished during in-reactor testing. Therefore, the development of appropriate in situ testing techniques will be critical to accurately determining the complex interactions between irradiation, loading conditions, and environment.

Other key factors in high temperature design must also be addressed for the advancement of fusion power. Better understanding of the active damage mechanisms during combined creep and fatigue deformation is imperative and the underlying reasons for the large variations observed in creep-fatigue damage between alloys must be understood. Both of these factors may allow for the development of alloys with improved creep-fatigue performance. However, an even larger task will be the incorporation of irradiation damage into the high temperature design methodology and code. This requires a more complete database on irradiation effects in 9–12Cr steels, SiC materials and V–Cr–Ti alloys, which is a large undertaking in itself.

7. Summary and conclusions

Fusion energy has a very high potential as a clean and abundant energy source. However, the very challenging service requirements of a fusion reactor system place high demands on structural materials. Years of research have identified three high-performance, reduced activation materials as leading candidates (8–9Cr ferritic/martensitic steels, SiC/SiC composites and V–Cr–Ti alloys), although there is still much to learn about the performance of these materials in fusion-relevant environments.

Fortunately, some lessons can be taken directly from previous large scale fast breeder reactor programs to identify areas which may be of concern looking forward to fusion power reactors. This paper has examined several such issues: radiation-induced solute segregation and phase stability; helium embrittlement; localized deformation; mass transfer and corrosion in liquid metal coolants, and high temperature design methodology. Previous research into each of these areas is an invaluable resource for identifying potential issues for each of the candidate materials for fusion reactors. While difficult, identifying and addressing these issues in advance are key steps toward sound materials science and fusion reactor design.

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