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Materials needs for fusion, Generation IV fission reactors and spallation neutron sources – similarities and differences

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Abstract

Fusion reactors, advanced fission reactors and high power accelerator spallation targets subject materials to damaging particle irradiation. Although these technologies derive their utility from different nuclear reactions and divergent applications, they experience many common features. Further, the physical mechanisms of radiation response are crosscutting. For example, swelling, phase instability, hardening, flow localization, and embrittlement must be understood in order to estimate component lifetimes. Additional commonalities include reliance on the same classes of materials and sometimes on the identical alloy for critical components. In addition, databases supporting designs are mainly derived from the same relatively few irradiation facilities and from similar types of experiments. Opportunities are examined for coordinated efforts. Emphasis is placed on the development of fundamental knowledge to support alloy design strategies for resistance to irradiation and to form a scientific basis to develop better materials.
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1. Introduction

Fusion reactors [1], advanced fission reactors [2] and high power accelerator spallation targets [3] are presently under intensive research and development. Advanced high performance materials are essential for the realization of these technologies. Historically, research and development for fusion and fission reactor materials technologies, especially in the field of radiation effects on materials, have benefited mutually from the extensive base of knowledge developed in programs targeted to particular reactor concepts and in the pursuit of fundamental information on materials behavior under irradiation. More recently, materials R&D for high power accelerator targets has both benefited from this knowledge base and contributed to it. In the future the opportunities for mutual benefit are expected to be even greater with the recent launch of the international Generation IV fission reactor program. Concepts for water-cooled, gas-cooled and liquid metal-cooled devices are being considered for these reactors as well as for fusion reactors. Similarly, water-cooled and liquid metal-cooled systems are under development for high power accelerator targets. These environments impose aggressive conditions on materials. It is generally understood that the lifetimes and the performance of these systems to their intended service will be dictated largely by the operational limitations of advanced structural materials.

With this perspective, important requirements and parameters for structural materials will be explored in this paper for fusion reactors, Generation IV fission reactors and high power accelerator spallation targets. Special emphasis will be placed on identifying and analyzing common R&D needs and differences among these systems. By deliberately spotlighting these issues it is suggested that the materials community may better exploit pooling of knowledge and utilization of relatively sparse resources in the field of materials R&D for advanced nuclear technologies.

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2. Background

2.1. Common issues

In fusion, fission and spallation technologies, the performance of structural materials is limited in general by the degradation of physical and mechanical properties by long-term exposure to fluxes of protons and neutrons, or by chemical interactions with working fluids that transfer heat, breed isotopes or produce spallation neutrons [4,5]. Although each nuclear component operates under a unique set of conditions, materials behavior is linked by the same underlying physics and the consequent radiation effects on properties. Thus advances in the understanding of the mechanisms of radiation effects and the emergence of mitigation strategies based on compositional and microstructural manipulation will have benefits that cross-cut a range of nuclear environments.

Critically important data on radiation effects in highly irradiated materials must be obtained from a diminishing set of higher flux fission reactors such as the HFIR, ATR, HFR, JMTR, JOYO, BOR 60, and the spallation sources LANSCE and SINQ. Similarly, the number of lower flux materials testing reactors used for experiments on reactor vessel materials is also diminishing. Significant opportunities exist for the sharing of information on the technology of irradiation testing, specimen miniaturization, advanced methods of property measurement and development of materials property databases to ensure consistency and to facilitate the exchange of data among programs. Because of particle flux spectrum differences and other non-prototypical features, the experimental data obtained from these facilities can only partially reproduce conditions for structural materials in the advanced concepts under consideration. These concepts involve a range of structural materials exposed to a variety of neutron and proton spectra and to spatial and temporal gradients in particle flux, temperature and mechanical loading and exposure to chemically active fluid media. Of necessity, materials selection will have to be based on incomplete experimental data and there is consequently a strong and cross-cutting need for physically-based modeling and microstructural analysis to develop a foundation of knowledge and understanding for extrapolations beyond the experimental database.

A final important thread that links these technologies is that several classes of structural alloys find applications in more than one system. Examples include (a) austenitic stainless steels for near term fusion applications, spallation neutron sources and several Generation IV fission reactor concepts, and (b) ferritic–martensitic steels for long-term fusion systems, for advanced fission reactors and possibly for long-term spallation neutron sources used for transmutation. For very high temperature applications, refractory metal alloys and structural composites such as SiC/SiC are being explored as potential materials for both advanced fission and fusion reactor concepts.

2.2. Fusion reactors

Since this paper is part of the proceedings of a conference on fusion reactors, the background in this area is provided by context within the meeting and the published proceedings. However, it should be mentioned that a very wide variety of fusion reactor concepts have been developed to some level. These range from the ITER, which is a low-dose low-temperature water-cooled blanket design [6], to high temperature helium-cooled blanket concepts and Li-cooled or Pb–Li-cooled blanket concepts where structural materials are projected to endure 150–200 dpa maximum dose. Some of the design conditions, materials selections and operating parameters for a wide spectrum of fusion reactor designs are reviewed in [4,5,7].

2.3. Generation IV fission reactors

Today fission reactors provide about 16% of the world's electricity supply and more than 20% of the US supply. The world-wide distribution of 438 nuclear reactors is aging and will need replacement and enhancement to both keep pace with and to take up a larger share of growing world-wide electricity demand. It is a widely held goal to augment the current fleet with significantly improved technology. A new generation of nuclear plant concepts has become the focus of international advanced reactor activity. It is termed Generation IV. The early prototype reactors built in the 1950s and 1960s, and the commercial power production reactors of the 1970s and 1980s constitute Generations I and II, respectively. Generation III denotes the advanced light water reactors whose designs were developed from the 1990s up to the present time, some of which have already been built outside the US. Generation IV embodies greater improvements and innovative advances in technology over earlier systems.

Ten countries have joined together to form the Generation IV International Forum (GIF): Argentina, Brazil, Canada, France, Japan, Korea, South Africa, Switzerland, the United Kingdom, and the United States, joined by the International Atomic Energy Agency, and the OECD Nuclear Energy Agency. Goals lie in four key areas: sustainability; economics; safety and reliability; and proliferation resistance and physical protection. These systems are intended for international deployment by 2030. Beginning in January 2000 the participants developed a technology roadmap. More than 100 concepts were evaluated; six were selected as most promising. Additional information is available in reports on the roadmap activity [2,8]. Below we distill key conditions to be experienced by the structural materials, in order to emphasize the similarities and differences with fusion reactors and spallation neutron sources.

The six concepts are categorized by neutron energy spectrum as thermal (two), fast (three) and one liquid-fueled epithermal/thermal system. The thermal concept closest to near-term deployment is the very high temperature reactor (VHTR), also termed the next generation nuclear plant (NGNP), with a gas outlet temperature in the vicinity of 1000 °C. The core structural material is primarily graphite utilizing prismatic block or pebble bed fuel configurations. Another concept utilizes a molten salt rather than gas as the coolant. Metallic components experience low displacement doses, significant thermal neutron fluxes and very high temperatures.

The other thermal reactor is the supercritical water cooled reactor (SCWR) operating at a pressure of 25 MPa, above the thermodynamic critical point of water, and with an outlet temperature of ~500 °C. The fast neutron concepts are the gas cooled fast reactor (GFR), the lead cooled fast reactor (LFR) and the sodium cooled fast reactor (SFR). In the liquid-fueled epithermal/thermal molten salt reactor (MSR) the molten salt fuel is a mixture of the fluorides of sodium, zirconium and uranium. The reference concept has an outlet temperature of 700 °C. For these reactors, structural materials are challenged by high levels of neutron displacement damage with temperatures ranging from 550 to 1000 °C. Operating conditions and materials R&D for VHTR and SCWR are described in [9,10], respectively.

2.4. Spallation neutron sources

Spallation neutron sources impinge a proton beam with energy of order GeV onto a high atomic number target to produce nuclear spallation reactions. Tens of neutrons are produced for each incident proton. These high energy neutrons are thermalized through nearby hydrogen rich moderators. The thermal neutrons may be applied in neutron scattering research, or to induce nuclear reactions such as for transmutation of radioactive wastes.

The largest spallation neutron sources for neutron scattering are the SINQ in Switzerland, the ISIS in England, and the LANSCE in the US. In the first of these the proton beam power is of order 1 MW, and in the latter two the beam power is of order 100–200 kW. The SINQ utilizes several types of targets ranging from a water-cooled solid rod target (zirconium alloy or lead) to a planned liquid Pb–Bi eutectic target. In LANSCE and ISIS the targets are water-cooled solid rod or plate tungsten or tantalum assemblies. To our knowledge there are no transmutation facilities operating.

Two advanced neutron scattering facilities are under construction, the SNS in the US and the JSNS in Japan, a part of the multipurpose accelerator facility JPARC. These facilities will become operational in 2006 and 2007, respectively. Both SNS and JSNS feature liquid mercury spallation targets contained in multiple walled vessels constructed of austenitic stainless steels. The beam power will be 2 MW produced by protons of 1 GeV and 2 mA in SNS, and 1 MW with 3 GeV protons and 0.33 mA in JSNS. The proton beams will be pulsed with pulse length <1 μs and repetition rate 60 Hz in the former and 25 Hz in the latter. Temperatures in the target during operation will be in the range 100–150 °C. In Europe, a conceptual design for ESS, a facility that may be constructed in the future, has been completed. Details of the materials research and development program for the SNS target are described in [11,12]. There has been extensive collaboration among international spallation materials R&D programs. The results of much of this work are documented in Proceedings of the International Workshops on Spallation Materials Technology [3,13].

2.5. Overlap in operating conditions

Several key parameters can more clearly show the overlap in these technologies. A most important consideration is temperature. Fig. 1 shows the respective operating regimes. The SCWR and VHTR cover the temperature extremes for the fission reactors, with the SCWR at the low end and the VHTR at the high end; dashed lines denote temperatures that could be reached in off-normal conditions.

For fusion, ITER is at the bottom of the range and A-SSTR2 at the top. Also shown for reference is DEMO, the fusion power reactor that will be constructed after ITER. Spallation source temperature ranges are shown for the neutron scattering facility SNS

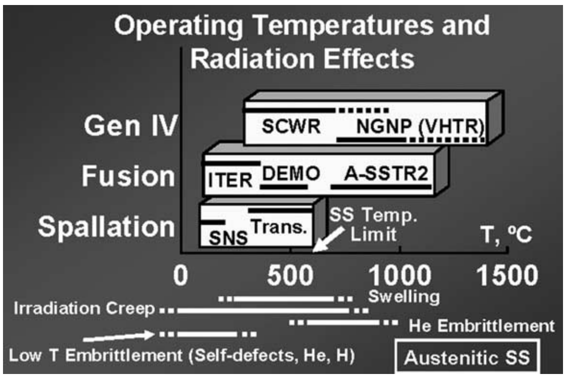

Fig. 1. Overlap in temperatures for fusion, fission and spallation facilities.

Table 1
Summary of operating parameters for fusion, fission and spallation facilities

Parameter	Technology		
	Fusion	Fission (Generation IV)	Spallation
Working fluid	H_2O, He, Li, PbLi, FLiBe	H_2O (SC), He, Na, Pb, PbBi	Hg, PbBi, H_2O
Energy	<14.1 MeV	<1–2 MeV (most n's)	$\leqslant$ 1 GeV (p and n)
He/dpa	10	0.1–50	50–100
Stresses	Moderate, slowly varying[a]	Moderate, slowly varying	High, pulsed

[a] For normal operation. In a plasma disruption the stresses to the structure are to 'high, transient'. High, for both fusion and spallation, indicates stress levels that are near the allowable maximum fraction of yield stress under applicable engineering design rules.

and for proposed transmutation facilities. In general there is more overlap in temperature between fusion and fission reactors than between fusion and spallation. For SNS and JSNS, there is overlap only with ITER.

Also shown for reference are ranges for certain radiation-induced degradation phenomena in austenitic stainless steels like type 316 LN, planned for the SNS target, ITER first wall and possibly SCWR core components. The temperature bars and corresponding labels indicate where phenomena of swelling, irradiation creep, low temperature embrittlement and helium embrittlement occur. Also shown is the upper temperature limit for application of this material in a radiation environment. Beyond this temperature the alloy strength would not be sufficient. It is also implied in this figure that within the possible range of application, variants tailored specifically for resistance to swelling and embrittlement must be used.

Table 1 summarizes other important operating parameters. There are large differences in particle energy and in resulting transmutation products. In spallation neutron sources the proton energies are of order GeV. As a result the spallation neutron spectrum peaks in the MeV range but includes a low population tail that extends up to the proton energy. The high energy protons give rise to high transmutation rates of helium per unit displacement damage, in the range 50–100 appm/dpa. In a fast reactor the corresponding rate is ~0.1 appm He/dpa. In a reactor with a significant fraction of thermal neutrons, this ratio can be nearly as high as in a spallation neutron source if there is significant nickel in the alloy, as there is in austenitic stainless steels, or if there is high boron content. In these cases helium arises from thermal neutron reactions with specific isotopes, Ni^{58} or B^{10}, respectively. For fusion, helium generation rates range from ~10–15 appm/dpa in structural alloys, to ~150 appm/dpa for SiC-based materials.

3. Materials

Initial designs for future fusion, fission and spallation facilities are based on the application of engineering materials for which a substantial body of information exists on physical and mechanical properties, large scale fabrication and joining, and operation in nuclear or non-nuclear environments under the appropriate corrosion/compatibility conditions. In order to meet potential licensing requirements, alloy selection is frequently confined to the restricted number of materials documented in the major design codes such as ASME and RCC-MR. However, only a very limited number of materials are presently incorporated into Section ASME III, Subsection NH, the relevant subsection for nuclear operation at high temperature (316, 304, Alloy 800H, 2.5Cr–1Mo, with 9Cr–1MoV in preparation). For many components the operating conditions exceed those allowed for these materials, necessitating a search for materials with superior properties and the development of appropriate code cases where dictated by licensing requirements for fission reactors; currently there are no spallation- or fusion-specific design codes or licensing processes.

Design codes do not provide guidelines for the treatment of environmental effects, i.e., chemical interactions between structural materials and coolants and effects of proton or neutron displacement damage and generation of transmutant gases. Within these environmental effects there exist a variety of time-dependent damaging mechanisms that frequently severely impact component lifetimes. In addition to temperature and dose, the radiation-induced phenomena are sensitive to dose rate, transmutant gas generation rate and operating temperature history. In some instances there is a strong interaction between corrosion phenomena and radiation effects. Meeting the performance goals of the concepts for fusion, spallation and fission systems will require long-term efforts to expand understanding of these phenomena and to develop sound scientific bases for (a) development of improved properties in existing alloys through compositional and microstructural modification, and (b) development of entirely new alloys specifically designed to resist environmentally-related property degradation in its many forms.

3.1. Austenitic and ferritic/martensitic steels

Although there are very significant differences in the fusion, fission and spallation environments, there is a range of performance-limiting phenomena arising from radiation effects that are common to all nuclear environments. It is beyond the scope of this paper to examine in detail the full range of materials currently being considered for Gen. IV systems [2,9,10]. However, to illustrate the similarities in challenges, the operating requirements for two of the principal structural materials used in component design, austenitic stainless steels and ferritic/martensitic stainless steels, are summarized in Tables 2 and 3.

The International Thermonuclear Experimental Reactor (ITER) is the first large-scale fusion device in which structural materials will be subjected to significant fluxes of fusion neutrons. Since it is the primary containment, the vacuum vessel will be constructed and operated within the design rules and requirements of a design code such as ASME or RCC-MR, although the possibility of developing a fusion-specific design code is under consideration. For the vacuum vessel material 316LN, the materials data base, design curves, specifications for fabrication and testing, the rules and constraints for design and service conditions are fully covered in the RCC-MR code. The in-vessel components are not restricted to code qualified materials and a wide range of existing engineering materials have been identified and design rules developed for their intended applications, e.g., Cu–Cr–Zr, Cu Al_2O_3, W and W alloys, C/C composites, Be, Ti–Al–V, and Ni-based alloys [14].

The austenitic stainless steels are important candidates for the SCWR fuel assemblies and core internals and there is some overlap with the ITER operating temperature regime for 316LN at 280–300 °C. In this regime, austenitic stainless steels experience strong radiation hardening coupled with reductions in uniform strain, flow localization and reductions in fracture toughness. Irradiation-assisted stress corrosion cracking, closely related to grain boundary segregation, is of significant concern at temperatures below 350 °C. The ITER operating conditions for the initial phase have been chosen in a dose-temperature regime where the impacts of these phenomena are manageable [15]. Similarly for the SNS, the prescribed initial operating displacement doses will be held to low values [11,12], so that the changes in the properties of the stainless steel container will be tolerable. However, these phenomena will affect the performance of SCWR components operating at temperatures in the range 280–350 °C and to doses beyond 5 dpa. Clearly, the very extensive design data base, design equations and design rules generated for 316LN under the ITER project could provide a valuable resource of direct relevance to the SCWR project. For operating temperatures >350 °C in the SCWR core internals, other phenomena become important in stainless steels such as void swelling, effects of grain boundary segregation and helium generation on ductility and rupture life and the effects of off-normal

Table 2
Austenitic stainless steels: spallation, fusion, and Generation IV fission applications

System (working fluid)	Component	T, °C	Maximum dose, dpa	Maximum He, appm	Candidate alloys
SNS (mercury)	Spallation target module	80–150	5*	200*	316LN
ITER (water)	First wall/blanket	100–300	3	75	316LN
SCWR	Fuel assembly	280–620	15	200	Advanced low
(SC water)	Core support/internals	280–500	0.1–20	250	swelling steels: D9, PNC316, HT-UPS

Asterisks indicate conditions set for removal of first target. Later targets may be subjected to higher doses and transmutation levels with experience gained in conditions specific to SNS [11,12].

Table 3
Ferritic/martensitic steels: fusion, and Generation IV fission applications

System (working fluid)	Component	T, °C	Maximum dose, dpa	Maximum He, appm	Candidate alloys
SSTR (water)	First wall/blanket	300–550	100	>1000	Low activation
HCLL (He)		270–550	100	>1000	8–9% Cr ferritic–
HCPB (He)		300–550	100	>1000	martensitic steels
SCWR (SC water)	Fuel assembly	280–620	15	20	Advanced 8–12%
	Core support/internals	280–500	0.1–20	20	ferritic–martensitic steels
LFR (Pb–Bi, Pb)	Fuel assembly	300–550	150	15	

temperature excursions on helium bubble coarsening and overall sink strengths. In this regime, stainless steels micro-alloyed with Ti, B and P for swelling resistance and higher creep strengths are required; examples include the Japanese PNC 316 [16], the French 15-15Ti alloy [17], and the US HT-UPS alloy [18]. However, the stainless steels have no application in fusion systems at high temperatures because of low thermal conductivity and helium embrittlement at grain boundaries.

To meet the high dose-high helium generation rate conditions projected for fusion DEMO systems and beyond, efforts are focused world-wide on variants of the 8–9 Cr ferritic/martensitic steels, modified for reduced activation properties and for improved resistance to hardening-induced shifts in fracture properties [4,5,19,20]. This alloy class is also being evaluated for LFR fuel assemblies and core structures and for SCWR core internals. For the fission reactor applications there is a strong interest in higher strength ferritic/martensitic steels such as NF 616, E911 and HCM 12A, which represent a group of materials code approved for operation at temperatures up to 620 °C [21].

Table 3 summarizes the operating ranges. Temperature regimes overlap strongly, and although there is an order of magnitude higher He/dpa ratio for fusion than fission, performance of structural materials is limited by essentially the same radiation-induced phenomena. For temperatures below ~400 °C, radiation hardening occurs, with reductions in uniform strain and flow localization. Additionally, significant changes in fracture behavior occur, manifested in upward shifts in ductile-to brittle transition. The dose and temperature dependence of these phenomena are dependent on both alloy composition and microstructure. These effects may be exacerbated in the case of fusion by higher helium. For spallation neutron sources for neutron scattering, where the operating temperatures are <200 °C and helium generation rates are up to an order of magnitude higher than for fusion, ferritic/martensitic steels are not considered to be good candidates. The radiation-induced ductile to brittle transition temperature, exacerbated by helium, may shift upward to the operating or shutdown range. These steels are, however, candidates for higher temperature spallation source targets for radioactive waste transmutation. At temperatures above ~400 °C, radiation-induced grain boundary segregation, long-term microstructural instabilities and their possible effects on fracture behavior are important issues. Effects of helium on rupture life and ductility of the ferritic/martensitic steels are relatively unexplored, and could affect performance of components operating above 550 °C. Finally, off-normal temperature excursion effects on microstructure and properties must be considered, particularly if there is a possibility of exceeding the austenite transformation temperature for a significant time.

3.2. Advanced materials

The reduced activation FM steels being investigated for fusion DEMO plants are limited to maximum operating temperatures of ~550 °C and to meet the higher temperature operating conditions of subsequent fusion power plants entirely new structural materials will have to be developed. Possible materials systems under investigation include mechanically alloyed ferritic and ferritic–martensitic steels [22,23], refractory metal alloys and SiC/SiC composites [24,25]. There is also a range of possible applications for these advanced materials in the Generation IV concepts under consideration.

Obtaining improved structural materials for nuclear applications depends upon developing a physical understanding of underlying mechanisms to form a sound basis for alloy design. A past example of this process is the successful development of the swelling-resistant austenitic stainless steels for LMFBR applications [26]. Today, within the broad context of developing materials for extended performance in nuclear environments, the mechanically-alloyed, oxide-dispersion-strengthened (ODS) alloys provide a promising example. Programs to develop ODS ferritic and ferritic/martensitic steels for nuclear applications originated in LMFBR programs pursued in the 1970s–80s and have been pursued more recently in Japan [22] and in Europe [23]. Recently, the application of advanced microanalytical methods has shown the existence of remarkably stable dispersions of nano-sized clusters of oxygen atoms stabilized by elements such as Y and Ti in both commercial and experimental alloys [27].

Efforts are underway to understand interactions between composition and processing parameters that control nano-cluster composition, number density and stability, and relationships among microstructural size scales and mechanical behavior. Advances in fundamental understanding of the behavior of nano-composited materials could enable the design of alloy compositions and microstructures specifically to address radiation and corrosion phenomena, which presently pose severe limitations on alloy performance. For fusion DEMO first wall/blanket applications, dispersions of nano-scale clusters and particles are on the correct scale for effectively trapping large concentrations of helium, which could minimize grain boundary embrittlement and extend swelling incubation periods to lifetime neutron fluences. For those fission systems that demand operating temperatures in the 600–1000 °C regime, nano-composited materials have the demonstrated potential for very high creep resistance. Moreover, they exhibit remarkable resistance to severe over-temperature events without destruction of microstructural components responsible for creep resistance. In principle, the matrix containing the nano-cluster dispersion could be modified to incorporate elements required for enhanced corrosion resistance in various working fluids. This is clearly an area where the resources and techniques available to world-wide fusion and fission programs could be very effectively combined to accelerate progress.

4. Conclusions

1. Existing engineering materials have significant applications in fission, fusion and spallation systems. However, highly irradiated components for advanced systems in general require improved or new structural materials, with microstructures and compositions designed to withstand specific chemical and irradiation environments coupled with advanced methods of design and regulation to ensure safety and reliability.
2. Advanced designs for fusion, fission and spallation systems encompass a wide diversity of conditions in terms of particle flux, spectra, temperature, mechanical loading conditions and chemical environments. However in both fcc and bcc alloys there is a broad underlying commonality in terms of the fundamental radiation-induced phenomena and associated radiation effects such as swelling, phase instabilities, hardening, flow localization, hardening and non-hardening embrittlement.
3. Coordinated efforts are needed among the three technologies to expand understanding of mechanisms and to apply this knowledge to develop alloys that can resist the effects of damaging phenomena. These efforts will build a scientific basis with which to design materials for specific environments in advanced fusion, fission and spallation systems.
4. World-wide efforts to understand the processing-microstructure-property relationships for mechanically-alloyed ferritic and ferritic/martensitic steels could lead to the development of materials with exceptional high temperature microstructural stability and creep strength, coupled with effective trapping of transmutant gases. Well coordinated research programs that fully benefit from rapidly advancing knowledge both within and outside the nuclear field could yield advanced materials with expanded operating regimes for the aggressive environments of future nuclear systems.

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