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Structural materials for Gen-IV nuclear reactors: Challenges and opportunities

K.L. Murty [a,*], I. Charit [b]

[a] North Carolina State University, Raleigh, NC 27695, USA
[b] University of Idaho, Moscow, ID 83844, USA

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ABSTRACT

Generation-IV reactor design concepts envisioned thus far cater toward a common goal of providing safer, longer lasting, proliferation-resistant and economically viable nuclear power plants. The foremost consideration in the successful development and deployment of Gen-IV reactor systems is the performance and reliability issues involving structural materials for both in-core and out-of-core applications. The structural materials need to endure much higher temperatures, higher neutron doses and extremely corrosive environment, which are beyond the experience of the current nuclear power plants. Materials under active consideration for use in different reactor components include various ferritic/martensitic steels, austenitic stainless steels, nickel-base superalloys, ceramics, composites, etc. This paper presents a summary of various Gen-IV reactor concepts, with emphasis on the structural materials issues depending on the specific application areas. This paper also discusses the challenges involved in using the existing materials under both service and off-normal conditions. Tasks become increasingly complex due to the operation of various fundamental phenomena like radiation-induced segregation, radiation-enhanced diffusion, precipitation, interactions between impurity elements and radiation-produced defects, swelling, helium generation and so forth. Further, high temperature capability (e.g. creep properties) of these materials is a critical, performance-limiting factor. It is demonstrated that novel alloy and microstructural design approaches coupled with new materials processing and fabrication techniques may mitigate the challenges, and the optimum system performance may be achieved under much demanding conditions.

1. Introduction

Most operating commercial nuclear reactors world-wide are of Generation-II category. The Generation-III reactors have just started to be deployed, and Gen-III+ reactors are at the advanced stage of commercialization. Although the safety and reliability of these reactors are very good, it has been widely recognized that the nuclear energy has a crucial role to play in mitigating the ever-increasing world energy needs. Hence, in 2000 the US Department of Energy launched a new program, called Generation-IV Initiative, to broaden the opportunity of nuclear energy utilization by making further advances in nuclear energy systems design. An instructive pictorial chronology of nuclear power reactor evolution has been illustrated elsewhere. Gen-IV Initiative is an international effort between ten countries and the European Union, and the number of the participating countries is on the rise [1]. This initiative calls for new nuclear energy systems that will significantly improve safety and reliability, sustainability, useful reactor life (60 years or more), proliferation-resistance and profitability, setting them apart from the current nuclear power reactors.

Six designs have been selected over others for further research and development, and subsequent deployment. These are summarized in Table 1 along with other relevant information. It is important to note that most commercial reactors operating today hardly see a coolant temperature exceeding 350 °C (i.e. temperature in the SI unit can be by adding 273 to the temperature in °C. However, in the rest of the paper °C is used to maintain consistency of temperature units throughout). Hence, the service environments predicted for the Gen-IV systems pose significant challenges to materials selection and qualification efforts. The structural components will undergo varied service conditions which can be summarized as follows: (a) exposure to higher temperatures (as apparent in Table 1), (b) higher neutron doses, and (c) extremely corrosive environment. The commonality of service conditions makes some cross-cutting opportunities possible. However, it is also important to remember that one material found suitable in one Gen-IV design may not be suitable for similar application in other designs depending on the reactor-specific service conditions.

Some desirable characteristics for the Gen-IV structural materials are noted below:

* Corresponding author.
E-mail address: murty@ncsu.edu (K.L. Murty).

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Table 1
Different Gen-IV nuclear reactor systems [2]

Reactor system	Coolant	Neutron spectrum	Core outlet temperature (°C)
Gas-cooled fast reactor (GFR)	Gas (e.g. He)	Fast	~850
Lead-cooled reactor (LFR)	Liquid metal (e.g. Pb, Pb–Bi)	Fast	550–800
Molten salt reactor (MSR)	Molten salt (fluoride salts)	Thermal	700–800
Sodium-cooled fast reactor (SFR)	Liquid metal (Na)	Fast	~550
Very high temperature reactor (VHTR)	Gas (e.g. He)	Thermal	>900
Super critical water-cooled reactor (SCWR)	Water	Thermal/fast	350–620

(1) Excellent dimensional stability against thermal and irradiation creep, void swelling, etc.
(2) Favorable mechanical properties such as strength, ductility, creep rupture, fatigue, creep–fatigue interactions, etc.
(3) Acceptable resistance to radiation damage (irradiation hardening and embrittlement) under high neutron doses (10–150 dpa or displacements per atom), helium embrittlement, etc.
(4) High degree of chemical compatibility between the structural materials and the coolant as well as with the fuel. In this regard, stress corrosion cracking (SCC), irradiation-assisted stress corrosion cracking (IASCC) and many other issues are important.

Finally, workability, weldability, cost, etc. are other important aspects that need to be looked into during the materials selection process. All these requirements are related to the fundamental high temperature degradation mechanisms such as phase instability, oxidation, radiation-induced segregation and so forth.

1.1. Limitations of the current state-of-the-art

Because of the stringent requirements noted above, the materials employed in today's commercial reactors are not suitable for use in Gen-IV reactors. For example, zirconium alloys (Zircaloy-2 and -4, Zr–2.5Nb) have been used routinely as fuel cladding and other reactor internals in both light and heavy water reactors because of their low neutron capture cross-section, acceptable mechanical and corrosion resistance in high temperature (probably never exceeding 350–380 °C under normal service conditions) aqueous environment. However, higher temperatures envisioned in Gen-IV reactors would limit the use of zirconium alloys because of increased susceptibility to hydrogen embrittlement due to severe hydride formation, allotropic phase changes at higher temperatures (α–β phase), poor creep properties and oxidation. It is instructive to note that some high performance zirconium alloys may be of possible use in relatively low temperature Gen-IV reactor design (such as, SCWR).

Further, the out-of-core components (pressure-vessel, piping, etc.) may need to be made from materials other than the low alloy ferritic steels (e.g. A533B) currently employed primarily because similar components in Gen-IV reactors are expected to withstand much higher temperatures [3] and neutron doses. Some of the fabrication difficulty involved in the VHTR construction demands mention here. For example, the pressure vessel for VHTR reactor as shown in Fig. 1 is about double the size of the currently operating PWRs. Heavy component forgings will be needed in the construction of these huge pressure vessels.

2. Candidate materials

Several candidate materials have been suggested for structural applications in Gen-IV reactors as evident in Table 2. Primary options are the materials which have a reasonable database present,

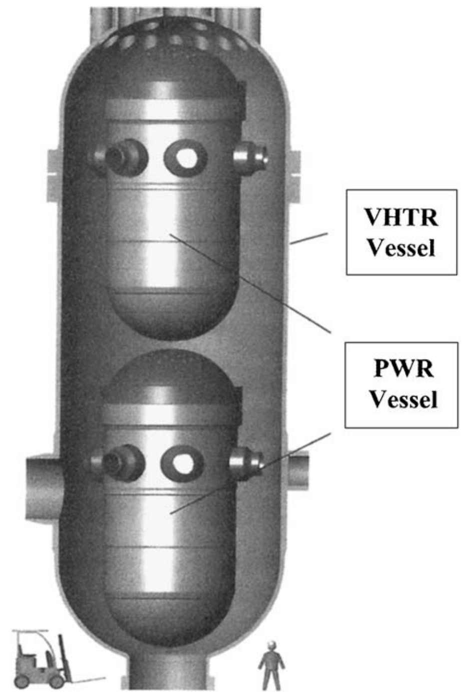

Fig. 1. The pressure vessel construction envisioned for VHTR as opposed to current typical pressurized water reactors (shown inside the VHTR vessel) [4].

and only qualifications need to be carried out. Secondary options refer to promising materials but need extensive research and development for database generation and subsequent qualification. Because of the longer reactor life coupled with tight time-schedule and resources to develop design methodologies, materials strategy involves accelerated testing followed by extrapolation to service conditions as well as microstructure-property modeling. The latter, however, is still in a budding stage to be fully capable of explaining various likely phenomena occurring in the material systems. Due to the limited space, here we only focus on different aspects of candidate metallic materials only.

2.1. Ferritic/martensitic (F–M) steels

Generally, the microstructure of F/M steels (9–12%Cr steels) is designed by suitable balancing of ferritic and austenitic stabilizing alloying elements in order to produce 100% austenite upon austenitization, and 100% martensite upon quenching or normalizing following austenitization. A tempering step at ~760 °C transforms much of the martensite to ferrite resulting in a tempered martensite structure. Here a brief account of the irradiation effects on the properties of F/M steels is described. From Table 2, it is clear that F–M steels may be used in a number of Gen-IV reactors. Initially, these steels were developed as structural materials for fossil-fuel power plants. Klueh [3] has summarized various F–M steels that were developed in the last sixty years or so. Continuous develop-

Table 2
Summary of various candidate structural materials for Gen-IV reactors [5]

Reactor system	F–M steel	Austenitic S.S.	ODS steel	Ni-base alloys	Graphite	Refractory alloys	Ceramics
GFR	P	P	P	P	–	P	P
Pb-LFR	P	P	S	–	–	S	S
MSR	–	–	–	P	P	S	S
SFR	P	P	P	–	–	–	–
SCWR	P	P	S	S	–	–	–
VHTR	S	–	–	P	P	S	P

P = primary option, S = secondary option.

ment with alloy and microstructural modifications has made their likely use in some Gen-IV reactors. F–M steels with 9–12% Cr are considered for use; some examples being HT-9, T-91, NF12, etc. because they tend to have better corrosion/oxidation resistance than the low-Cr ones. Previously, some advanced F–M steels have been considered for fusion reactor applications (first wall and blanket applications) because of their noted reduced-activation (RA) property that refers to a quick radioactive decay after neutron irradiation, allowing shallow burial of the components after component replacement or plant decommissioning. This property will be helpful in Gen-IV reactors, if not the sole guiding factor. Further, they have good void swelling resistance and relatively good creep resistance. However, there are concerns regarding their low long-term creep rupture strength at higher temperatures and irradiation embrittlement at or less than 400 °C.

Here we describe one example from 9Cr–1MoVNb steel (T91) on the radiation effect in F/M steels [2]. Irradiation exposure dose of 9 dpa resulted in appreciable radiation hardening due to the formation of wide range of radiation-produced defects in a temperature range of 425–450 °C (Fig. 2(a)). Fig. 2(b) shows the corresponding ductility as a function of test temperature. Hardening causes a decrease in ductility at the lowest temperature. However, interestingly the ductility for the irradiated alloy increases at 450 °C compared to the aged alloy. It is interesting to note that the aged alloy shows a maximum strength at a temperature where the irradiated alloy shows much higher ductility. However, note that the ductility data of the unirradiated alloy are not available at 450 °C. Radiation hardening saturates by around 10 dpa. For irradiation above 425–450 °C, there may be enhanced softening due to increased recovery and coarsening. Although the temperature range to be endured in proposed Gen IV systems is higher than 400 °C, the material behavior in this regime should be obtained to address the overall reliability aspects. The effect of alloying elements on the radiation-induced mechanical property changes needs to be carefully evaluated.

Irradiation hardening also affects other properties like toughness. Fig. 3 shows the Charpy impact curve (impact energy vs. temperature) for a 12Cr–1MoVW (HT9) alloy as has been profusely observed for other low alloy ferritic RPV steels. It clearly shows that there is an increase in ductile–brittle transition temperature (DBTT) accompanied with a drop in the upper shelf energy. The magnitudes of the shift and energy drop, however, depend on the irradiation temperature and neutron flux spectrum. In this case, radiation embrittlement saturates at around 10 dpa (Fig. 3).

Phase stability under neutron irradiation is also a major concern and radiation-induced segregation (RIS) affects many useful properties. Furthermore, RIS leads to eventual formation of precipitates such as, M_6C, χ-phase, α', etc. in irradiated F/M steels all of which enhance embrittling effects. Thus, microstructural evolution as a result of a combination of irradiation and high temperature affects the creep rupture properties. A stress–LMP plot for various F–M steels is shown in Fig. 4(a). It is interesting to note that the reduced-activation F–M steels have much better creep rupture properties as opposed to HT-9 steel [6]. Klueh [3] has adequately described this effect.

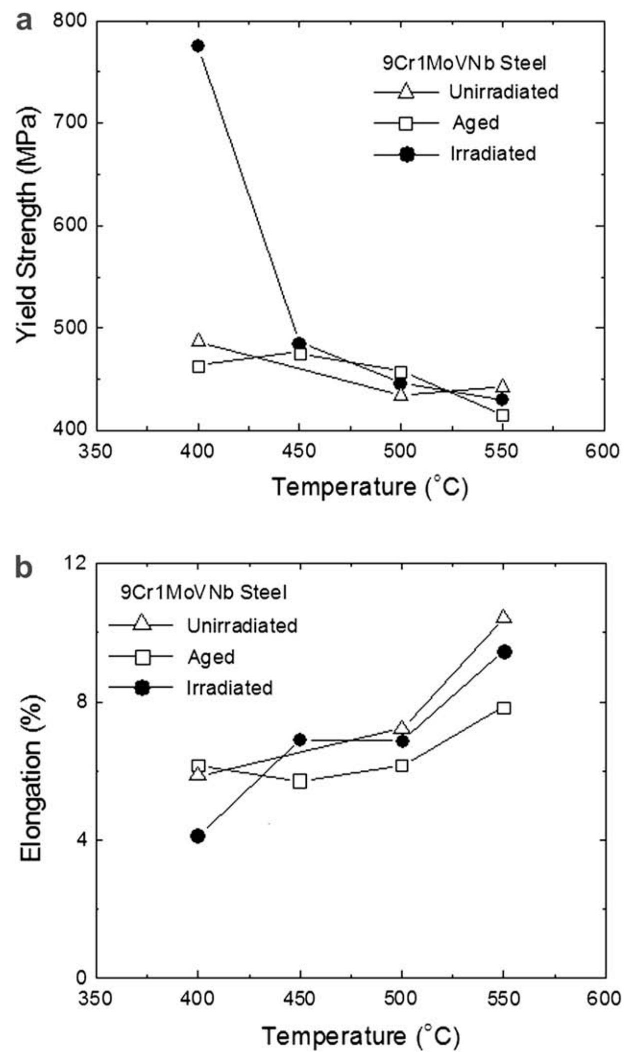

Fig. 2. Variation of (a) yield strength and (b) total elongation as a function of test temperature.

Fig. 4(b) shows the creep rate as a function of stress for a modified 9Cr–1Mo (P91) alloy. At higher stresses, the stress exponent value (n) is ~10 and at lower stresses, n value is ~1. Higher stress exponent may imply the operation of dislocation creep with threshold stress created by various second phase particles, whereas lower stress exponent refers possibly to diffusion creep. It reflects a transition in creep mechanism. It has a great practical significance in that blind extrapolation of the high stress data to lower service stresses leads to non-conservative estimates of the strain-rates (see the dotted line). In addition, temperature gradients resulting from start-ups, shut-downs and in-service fluctuations induce stress reversals. Creep–fatigue interactions thus

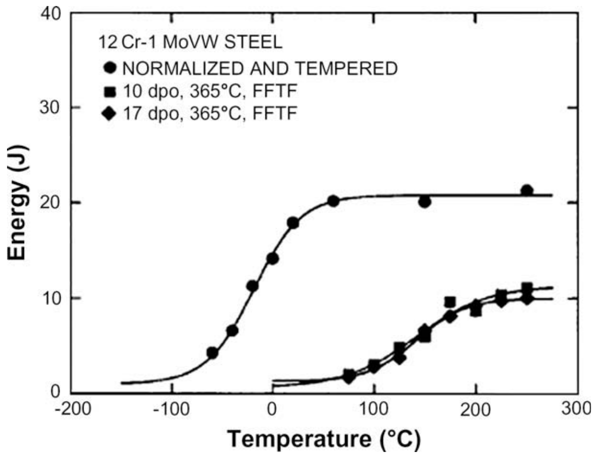

Fig. 3. Charpy impact energy vs. temperature for unirradiated and irradiated 12Cr1MoVW steels [3].

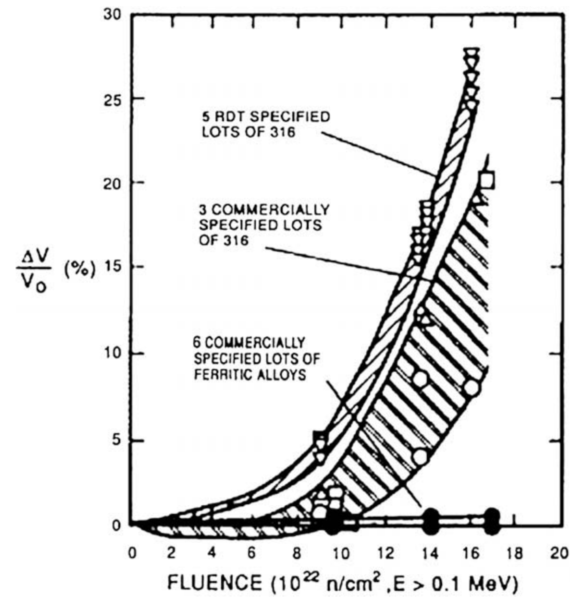

Fig. 5. Volumetric swelling vs. neutron fluence in austenitic and ferritic steels [3].

become important in the next generation reactors. Hence, database pertaining to those effects is a must to qualify a material for Gen-IV application.

2.2. Austenitic stainless steels

Austenitic stainless steels have good creep resistance to higher temperatures coupled with reasonable corrosion/oxidation resistance. Alloys like 316LN, D-9, etc. are good examples.

However, relatively large amount of void swelling at moderate neutron doses remains a major performance-limiting issue as depicted in Fig. 5, the volumetric swelling of various steels as a function of neutron fluence. It is important to note that the extent of swelling is much higher in different lots of an austenitic stainless steel (316 SS) compared to ferritic or F–M ones [2]. In some applications, their low thermal conductivity may adversely affect the reactor efficiency. Radiation-induced segregation (RIS) and phase stability issues will also play a major role. For example, radiation-assisted depletion of Cr from the grain boundaries may render the austenitic steels susceptible to corrosion in water or lead–alloy cooled systems. The interactions between RIS and void swelling may also be important [7].

Further, irradiation creep and thermal creep may have important implications on the dimensional stability and performance of the reactor. Deformation mechanism maps (DMM) depicting the rate-controlling deformation mechanisms in stress–temperature space prove to be useful. Figs. 6(a) and (b) show the DMMs for a 316 stainless steel in unirradiated and irradiated (dose of 10 dpa) conditions, respectively [8]. It is to be noted that a separate irradiation creep regime appears in the DMM of the irradiated material. However such information is currently not available for many metals of interest.

Kurata et al. [9] have studied corrosion behavior of ferritic and austenitic steels in Pb–Bi eutectic for their possible applications in LFRs. Fig. 7 shows corrosion depth as a function of Cr content in

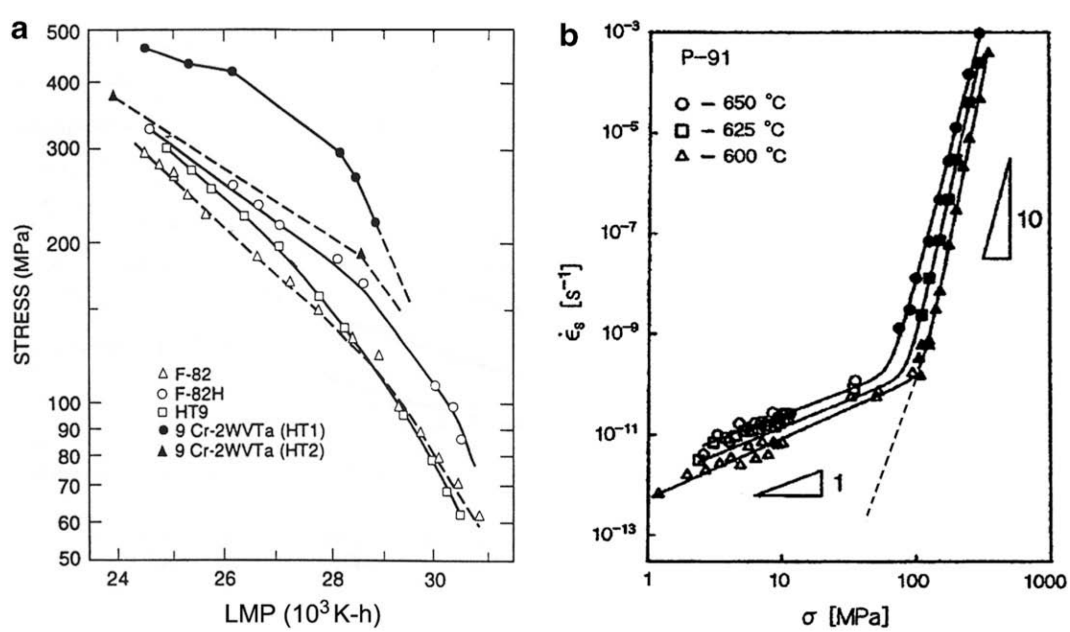

Fig. 4. (a) Stress vs. LMP plots for F–M steels [5], and (b) creep rate vs. stress plot for P91 alloy [3].

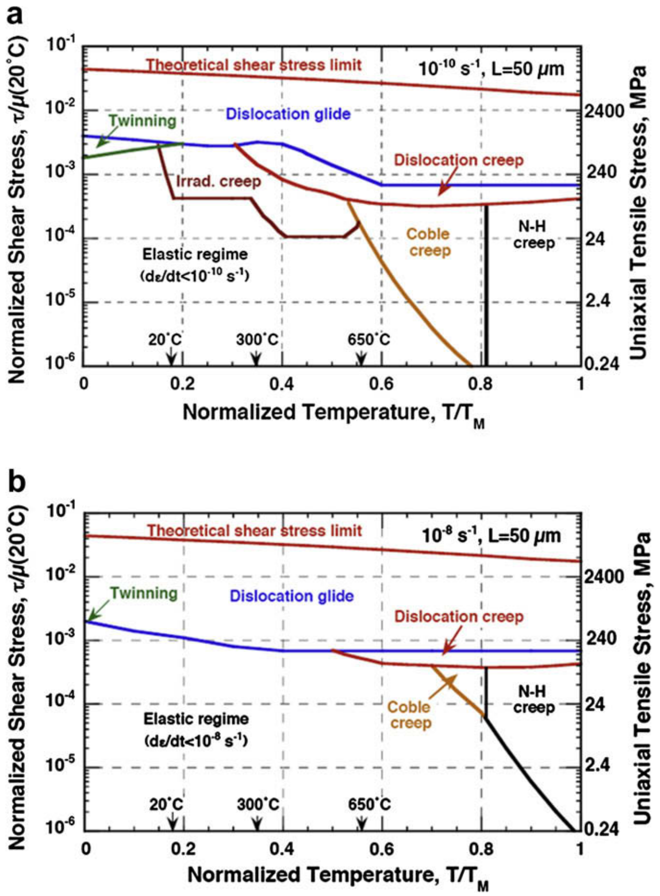

Fig. 6. Deformation mechanism maps for (a) unirradiated and (b) irradiated 316 stainless steel [8].

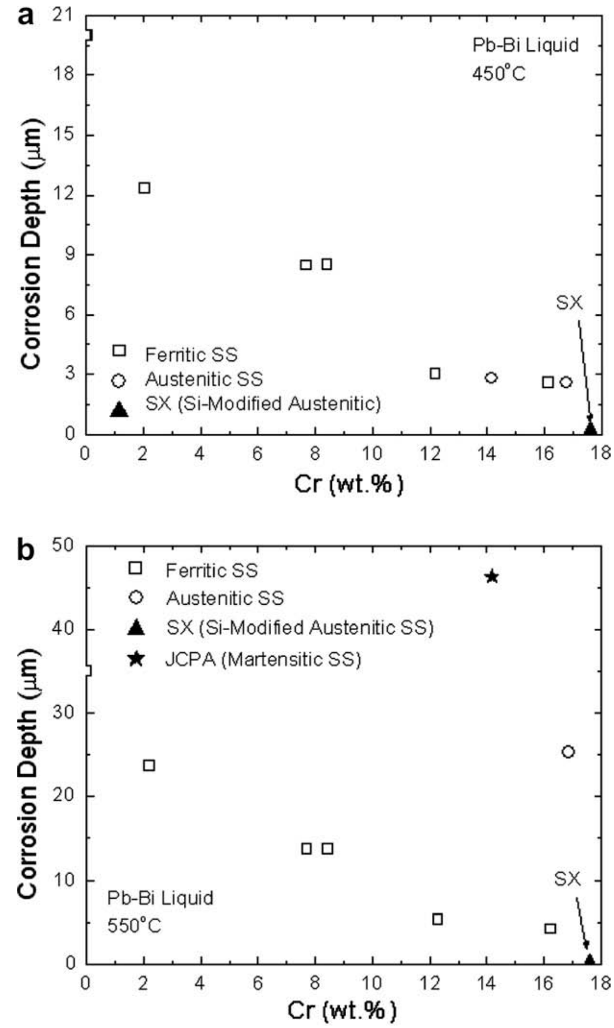

Fig. 7. Corrosion depth as a function of Cr content at (a) 450 and (b) 550 °C for ferritic and austenitic steels [9].

these steels. As expected, the extent of corrosion decreases with increasing Cr content in ferritic steels in temperature range 450–550 °C. There is not enough data for austenitic steels. It is clear that at lower temperature (450 °C), austenitic steels have similar corrosion resistance as ferritic ones. However, at higher temperatures (550 °C) austenitic steels show a poor performance. Si-additions seem to increase corrosion resistance in Pb–Bi medium and needs to be further investigated.

2.3. Oxide dispersion strengthened (ODS) steels

ODS steels are made through mechanical alloying process. These alloys have good high temperature properties, radiation-resistance in terms of swelling and radiation embrittlement. One example is 12YWT (12YWT: Fe–12.29Cr–3W–0.39Ti–0.248Y_2O_3). It has been found that small nanoclusters of Y–Ti–O particles hinder dislocation motion effectively, and can act as effective sinks for radiation-induced defects. These are called nanostructured ferritic alloys (NFA). Fig. 8(a) shows the atom probe compositional maps of the 12YWT steel [10]. It clearly shows clustering of Y, Ti and O. It has also been found that the presence of Ti and W help in creating a much smaller oxide particles with average size of 5–6 nm. In Fig. 8(b), the effects of these particles are evident from the better creep test strength of 12YWT at 700 °C compared with 12Y1 (no Ti, W) and 12YW (no Ti) [11].

Kimura et al. [12] investigated various ODS steels and pronounced radiation hardening was noted at lower temperatures while at higher temperatures, the hardening got reduced drastically. However, the total strain (or ductility) of the irradiated steels did not have any effect at any temperature. However, more research at higher dose rates will be needed in this direction. Nano-scale design of structural materials may have major implications in future nuclear reactor systems.

2.4. Ni-base Alloys

Ni-base alloys have traditionally been used for high temperature applications. Therefore, it is only prudent to study their viability in Gen-IV reactor applications. New Ni-base superalloys (such as, IN740: Ni–2Fe–24Cr–20Co–2Nb–0.5Mo–2Ti–1C, wt%) have good creep rupture properties (Fig. 9(a)) and high temperature strength (Fig. 9(b)) [13]. The main problems with Ni-base alloys would be the radiation embrittlement, swelling and phase instability under neutron radiation environment. Their applicability in balance-of-plant features (turbines, steam-generators, etc.) where radiation effects are minimal is possible. However, high temperature He embrittlement is an issue to be looked at for applications in GFR/VHTR reactors. More research is needed to better judge their viability for Gen-IV reactors. Currently, a solid solution strengthened Ni-base superalloy (Alloy 617) is being considered for heat exchanger applications for next generation nuclear plant (NGNP) that incorporates VHTR reactor concept.

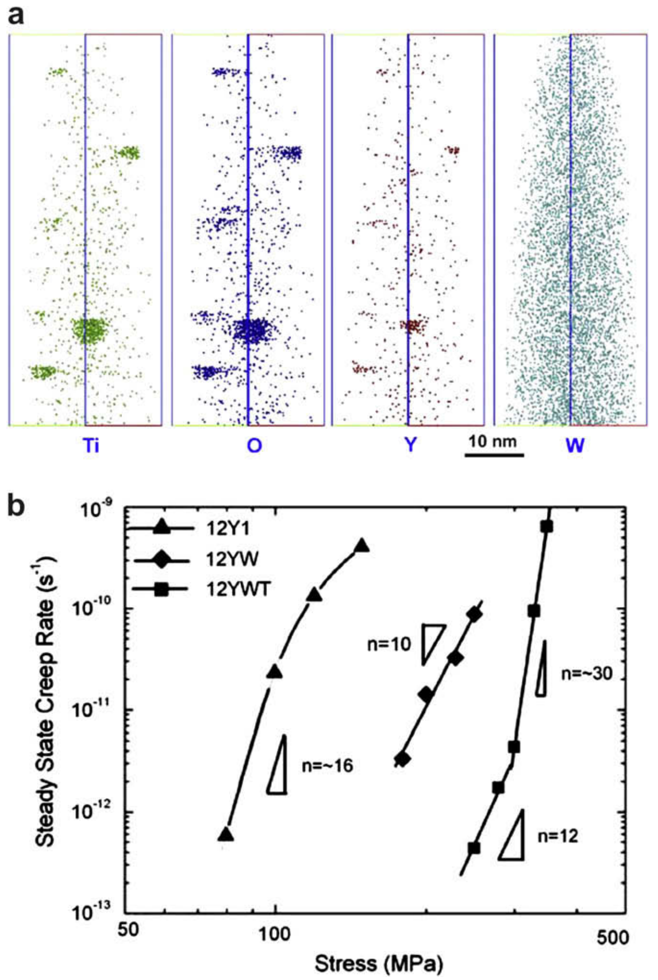

Fig. 8. (a) Compositional maps produced by atom probe study in 12YWT even after annealing at 1300 °C for 24 h [10], and (b) steady-state strain rate as a function of stress for various ODS steels [11].

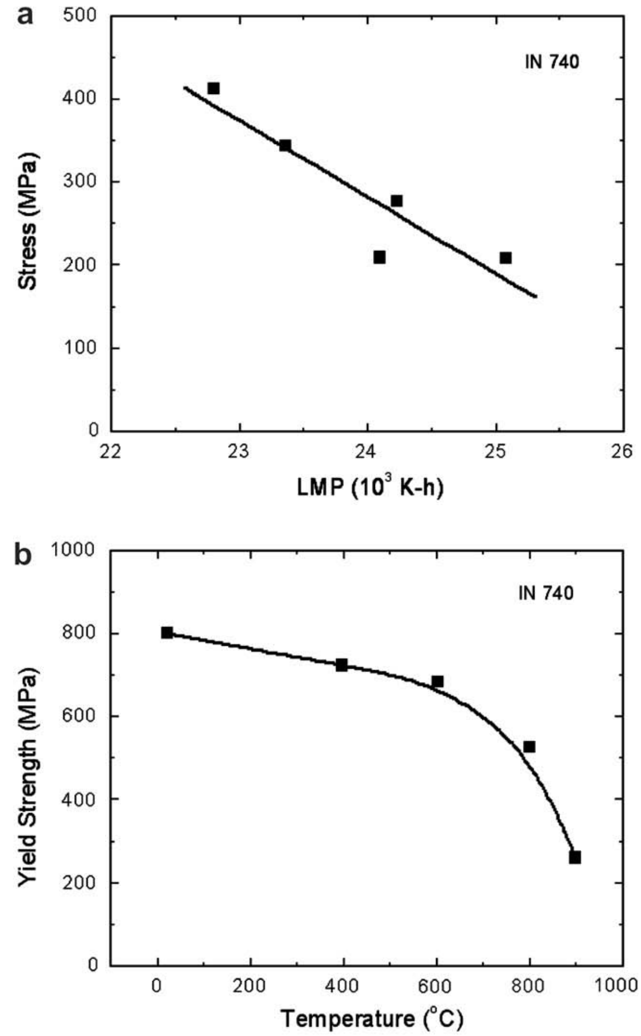

Fig. 9. (a) Stress vs. LMP plot, and (b) yield strength as a function of temperature for IN740 alloy.

2.5. Refractory alloys

Refractory metals (such as, Nb, Mo, Ta, etc.) have melting temperatures in excess of 2000 °C. Hence, they should have potential applications at high temperatures. Table 3 shows various aspects of refractory metals on a 10-point scale (1 being the worst and 10 being the best) [14]. Although refractory metals possess good creep resistance and swelling resistance up to high burnups, they have poor oxidation resistance coupled with low temperature radiation embrittlement and fabrication (joining) difficulties. That is why refractory alloys are not being considered for Gen-IV reactor applications.

3. New materials development

Although currently the existing materials databases are being utilized for possible materials selection, new materials development may be necessary. Instead of going through time-consuming and expensive new alloy design, it is possible to use innovative processing techniques to tailor the properties of the existing alloys to suit Gen-IV applications.

Watanabe [15] proposed the grain boundary engineering (GBE) concept. GBE is a means of tailoring grain boundary microstructure (grain boundary character distribution, etc.). The GBE treat-

Table 3
Summary of various engineering aspects of refractory metals [14]

Technology category	Nb–1Zr	Ta–10W	Mo–0.5Ti–0.1Zr	W–Re	Re
Fabricability	8	7	4	3	4
Weldability	7	7	4	3	7
Creep strength	6	8	8	8	9
Oxidation resistance	1	1	3	3	7
Alkali metal compatibility	8	9	9	9	8
Radiation effects	6	6?	5	4	4?
Cost (2 mm sheet)	4	3	4	3	2

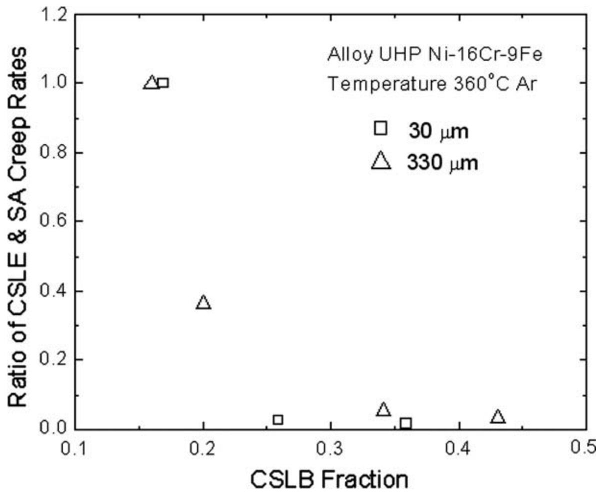

Fig. 10. Ratio of the creep rate of grain boundary engineered material and solution-annealed Ni–16Cr–9Fe alloy as a function of CSL boundary fraction [16].

ment involves suitable combination of annealing and cold working (i.e. thermal mechanical) treatments. There is now overwhelming evidence that certain coincident site lattice boundaries (CSLBs) with Σ value $\leqslant 29$ (i.e. special CSLBs) possess certain unique grain boundary properties, such as less susceptibility to impurity segregation, lower diffusivity, better resistance to grain boundary sliding, etc. Therefore, the existence of higher number of CSLBs leads to greater resistance to intergranular degradation against fracture, cavitation and localized corrosion, higher creep resistance and possibly higher radiation damage resistance.

An example of the success of the GBE concept in practice is shown in Fig. 10 where the increase in the CSLB fraction significantly reduced the creep rate in a Ni–16Cr–9Fe alloy irrespective of grain size [16]. GBE, at least in principle, can be applicable to any kinds of materials. However, its applicability in commercial applications still needs to be substantiated. It is further expected that microstructure-property modeling will be of great importance in Gen-IV materials development efforts. Use of modern computational techniques (molecular dynamics simulation, kinetic Monte Carlo methods, etc.) will greatly help in this direction.

4. Concluding remarks

In this paper, we have covered many issues related to structural materials to be considered for use in Gen-IV reactors. We did not cover various fabricability aspects of these materials. Also, many non-metallic candidate structural materials (graphite, SiC–SiC composites, etc.) could not be discussed. However, it is true that extensive material qualification efforts will be required. Lack of fast spectrum irradiation facility and high temperature testing facilities restrict appropriate evaluation of structural materials. However, innovative approaches (like GBE) along with extensive experimental and modeling work are essential in developing high temperature, radiation-resistant structural materials amenable for Gen-IV reactor systems.

Acknowledgements

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