Motivation for Utilizing New High-performance Advanced Materials in Nuclear Energy Systems

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Abstract

Despite the very demanding operational environment in nuclear reactors, there have been relatively few advanced high-performance materials introduced into fission reactors during the past 50 years. Some of the regulatory and operational barriers to the introduction of high performance materials are briefly discussed, and several examples of potential improvement in current and planned fission reactor systems that could be enabled by advanced structural materials for in-core applications are outlined.

1. Introduction

Current and proposed nuclear energy systems represent a very challenging operating environment for materials due to the presence of high ionizing and displacement damage radiation fields along with moderate to high mechanical stress, moderate to high operating temperatures, and the potential for interaction with corrosive coolants and/or volatile fission products [1-6]. Remarkable improvements in power plant operational reliability have been achieved for commercial light water reactors (LWRs) since the 1970s. As summarized in Figure 1, the current annualized fleet-averaged availability for US nuclear power reactors is >90% of their ideal capacity, compared to ~50 to 60% for fossil energy systems and ~25 to 35% for utility-scale hydroelectric and renewable energy electrical systems [7]. This impressive average capacity factor for US nuclear power plants is largely associated with improvements in operational procedures and scheduled maintenance, along with avoidance of unplanned shutdowns due to improvements in fuel and steam generator performance including evolutionary development of robust fuel cladding materials [8].

Since the term “advanced materials” is subjective (any material with properties superior to natural ore is arguably “advanced”), for the purposes of this article we have adopted a working definition for advanced materials to encompass any structural material with key properties (e.g., strength, thermal creep-fatigue, fracture toughness and/or resistance to stress corrosion cracking) that are significantly superior to structural materials commercially available in 1970 (corresponding to the onset of the “Generation II” LWR era). Of course, in many cases conventional commercial materials may provide fully adequate performance, and often the most expedient and cost-effective approach to resolve a material performance deficiency is to switch to another existing commercial material that is known to have improved resistance to the particular degradation phenomenon. However, in the long term there can be substantial benefits from R&D to create a select number of specifically tailored advanced materials to enable improved safety margins, better economics, etc. In this article we advocate that there are important benefits to be gained in nuclear energy systems by switching to advanced materials in a few particularly demanding components.

As summarized in Figure 2, with the exception of systematic improvements in Zr-based fuel cladding and the switch from Alloy 600 to Alloy 690 in steam generators, remarkably few new materials (or modifications of existing code-qualified materials) have been introduced into commercial LWRs during the past 50 years. There have been several important incremental structural material replacements (e.g., Type 304L for Type 304 austenitic stainless steel in BWR steam lines to improve weldability and resistance to stress corrosion cracking) and modified heat treatments to reduce residual stresses or sensitization (e.g. modified heat treatment of Alloy X-750 to suppress stress corrosion cracking susceptibility) [2, 5]. However, it is remarkable that new “generation III” LWRs under construction in the U.S. and elsewhere worldwide are largely being constructed using 1960s-era (or earlier) materials for the reactor pressure vessel (tempered bainitic low alloy steels containing ~2.5% Mn, Mo, Si, Cr and C solute) [9] and major core internal structural components (e.g., Types 304, 316, 321 and 347 stainless steels) and high strength springs and fasteners (e.g., Fe- and Ni-base alloys such as A286 and X-750) [5]. Somewhat paradoxically, the demanding operating environment associated with LWRs has fostered an incremental approach to introduction of new materials. Unanticipated materials problems early in the development of nuclear power (often associated with corrosion and stress corrosion cracking) and the large economic consequences associated with unplanned reactor outages (~1 M\$/day loss in revenue) have driven the nuclear reactor industry to be very conservative with respect to adopting new higher performance materials. In addition, there are perceived high regulatory obstacles (in terms of cost and time to obtain relevant engineering code committee and regulatory agency approval for a new material) for any reactor components associated with public safety such as the reactor pressure vessel.

These perceived regulatory approval obstacles, along with the perception of marginal benefit but potentially large economic risk associated with introduction of an unproven material (e.g., unanticipated shutdowns or appearance of new degradation phenomena), have stifled much of LWR component material innovation despite the existence of several promising high performance materials options that offer significant advantages. This is particularly of concern for components in contact with primary coolant due to potential unanticipated negative effects on the carefully controlled primary loop water chemistry. As discussed in Section 2, the low alloy steel currently used for LWR reactor pressure vessels has moderate strength and is embrittled by neutrons at a relatively more rapid rate compared to higher strength modern low alloy steels [10, 11]. Concerns about more pronounced neutron irradiation embrittlement of weldments has led the pressurized water reactor industry to switch to large ring forgings of the reactor vessel [12], which eliminates longitudinal welds in the core beltline region but requires forging of large components that can only be performed at a few facilities worldwide, with long waiting times and high cost. In other words, the selected approach to remedy the inferior radiation performance of the historical RPV steel (accentuated in the welds) was to eliminate the welds in the historical steel (requiring expensive ring forgings) rather than to pursue a higher-performance (radiation-resistant) low alloy steel that would exhibit sufficient resistance to weld embrittlement. Reluctance to consider advanced radiation resistant, more fabricable low alloy steels stems from concerns that introduction of an unproven material could potentially cause unanticipated operational issues or a reduction in plant lifetime (due to unanticipated degradation due to dose rate effects, etc.). However, advanced nuclear

reactor materials and construction techniques (e.g., fewer manual weldments) could directly reduce construction costs and also enable improved safety margins, which in turn could lead to easing of some quality assurance requirements that negatively impact reactor construction costs.

It is interesting to contrast the reluctance to introduce new materials in the nuclear fission power industry with that of the aircraft or fossil energy industries. For current fission energy reactors, there is no broad movement to significantly improve fuel/thermodynamic efficiency (unlike the case for fossil energy [13] or aerospace [14] industries): nuclear fuel is viewed as relatively cheap and sufficiently plentiful, and the technological hurdles associated with higher thermodynamic efficiency (requiring a host of new materials to accommodate the higher operating temperatures) are viewed as being too risky from an operational perspective. In contrast, despite the intense regulatory oversight and high attention to public safety within the aircraft industry, commercial aircraft have evolved from being constructed largely (>75%) from aluminum alloys (with ~1% composite materials) in the 1960s to ~25% aluminum alloys and ~50% composite materials in new aircraft such as the Boeing 787 [15]. This continuous introduction of new materials and design modifications has been central to the documented steady improvement in the operating parameters (operational cost, thermodynamic efficiency, reliability, etc.) for these industries.

Dramatic advances in the speed to design new high-performance structural alloys are now available due to improvements in computational thermodynamics databases and modeling tools [16, 17]. These new tools are being exploited to rapidly design and develop improved alloys for fossil energy [18-22] and automotive [23-25] structural alloy applications, and promising new high-performance steels for nuclear energy applications have been designed and experimentally tested although none of these new nuclear energy alloys have been fully qualified for reactor applications by the appropriate regulatory authority [26-30]. As discussed for several specific examples later in this article, enhanced use of these so-called integrated computational materials engineering (ICME) design tools to specifically tailor their performance for demanding nuclear energy applications could lead to expedient development of new high performance structural alloys. In other cases, there may be opportunities to leverage recent advances in high temperature ceramic composites technology [31] or advanced manufacturing approaches such as additive manufacturing [32, 33]. In all cases, obtaining relevant engineering code qualification and approval by the relevant nuclear energy regulatory authorities remains as a major hurdle.

In the following we focus on key in-core structural materials issues for LWR, sodium-cooled fast reactor (SFR) and very high temperature reactor (VHTR) fission power systems, since these are the nuclear energy systems either currently operating or most likely to complete development, licensing and deployment in the next 10-20 years. Assessments of materials challenges for other proposed Generation IV fission and fusion energy systems are given elsewhere [2-5, 11]. Analogous arguments can be made regarding the benefits associated with consideration of advanced structural materials for key out of core components such as heat exchangers, although they are not specifically addressed here.

2. Light water reactors

Internationally, the commercial LWR dominates nuclear power production with about 440 reactors in 31 countries providing ~11% of the world's electricity. Additionally there are about 180 shipboard reactors. The dominance of LWR technology in today's nuclear energy arena was leveraged and put on its design path in large part from development of naval reactors that began conceptually in the mid 1940's and emerged in 1955 with the first nuclear-powered submarine, the USS-Nautilus. That first naval reactor utilized platelet fuel of uranium and zirconium in a carbon steel pressure vessel. Building upon this R&D infrastructure, the first civilian prototype reactor (Shippingport) came online in 1957. The fuel for the blanket region of that reactor was UO_2 pellets clad in a zirconium alloy (Zircaloy-2), similar to some existing power reactor fuels.

Within the LWR arena, numerous examples exist where the nuclear materials technology has not advanced beyond its originally adopted 1950s-1960s materials (cf. Fig. 2). Amongst the most conspicuous examples are the continued use in new builds of an historic and relatively simple low alloy steel for the reactor pressure vessel (RPV) and utilization of UO_2 pellets with zirconium alloy cladding for nuclear fuel. A review of the former example and a more detailed discussion of the latter are offered in this section. Other key LWR materials issues such as research to identify life-limiting phenomena and determining the optimized power plant lifetime extension by judicious replacement of selective components are discussed elsewhere [34]. Nuclear power has essentially been investing in improved engineering safety systems (often with multiple redundancy to provide "defense in depth") rather than advanced materials that may obviate the need for some of these systems.

On the perceived basis that economics of scale govern nuclear electricity production costs (more recent analyses have found costs are often dominated by other factors [35-39]), the typical LWR plant size was designed and constructed to operate at a relatively large size of ~1 GWe. Plants of this size require very large RPVs (~ 4 m diameter and 22 cm thick). These vessels were historically made from plate segments of low alloy steel (A302B in 1st-generation reactors, and then alloys A508 and A533) welded to one another. Current design requirements necessitate shell rings instead of plates to avoid axial welds near the core region where the neutron flux on the RPV is highest. These rings are forged from ingots weighing up to 500 tons and require exceptionally large capacity hydraulic presses (up to 15000 tons). Only a handful of places exist worldwide (none in the US) with the capability to perform this costly process. This showcases the detrimental effects of the inability of nuclear power industry to adopt new materials. If a transition away from current pressure vessel steels were made to adopt steels such as 3Cr-3WV with the appropriate bainitic microstructure [40, 41], major benefits in cost and reliability could be realized. Specifically, the higher strength in these alloys would translate into a meaningful (~40%) reduction in thickness necessary for these vessels. This reduction in thickness, and therefore ingot mass, would greatly reduce fabrication costs while enabling access to numerous smaller capacity vendors (existing in the US and elsewhere). Furthermore, safety and reliability may be enhanced for this critical component given the anticipated improved irradiation resistance in these alloys [42-44]. For example, the unirradiated fracture toughness for 3Cr-3WV steel is similar or superior to conventional 1Cr RPV steels [40], and the irradiation-induced shift in ductile to brittle transition temperature and reduction in upper shelf toughness is expected to be markedly less for the 3Cr-3WV steels due to

reduced radiation hardening [10, 11], although additional experimental data are needed to confirm this expectation.

Along similar lines of reasoning, several other historic permanent core components (e.g., based on 304 or 316 SS) should be considered for replacement by improved radiation- and stress corrosion cracking-resistant materials in new build reactors. One of the most challenging issues for in-core LWR structural materials is irradiation assisted stress corrosion cracking [45-47]. As noted elsewhere [48], LWRs have been plagued by numerous key structural components that were constructed using materials with significant susceptibility to stress corrosion cracking. Research and development activities such as the jointly coordinated Electric Power Research Institute and U.S. Department of Energy “Advanced Radiation Resistant Materials” program [49] are examining alternatives to existing LWR core materials and potential structural material candidates for Generation IV reactors. Numerous appropriate alternatives to historic stainless steels are being examined, although it is notable essentially all of the candidate materials being considered in the ARRM program (except for some “innovative materials” such as SiC and metallic glass) were developed more than 25 to 50 years ago. It would be useful to consider new high performance structural alloys specifically designed to be highly resistant to stress corrosion cracking (rather than limiting the candidate options to existing materials).

Zr alloys for LWR fuel cladding have been continuously advanced with over six decades of active R&D for this narrow application in LWRs and therefore are highly optimized for normal reactor operations. Zr is highly transparent to neutrons, and this attribute drove the development of Zr alloy cladding for very compact submarine reactors where neutron economy was paramount [50] (with an accompanying major influence on development of commercial LWR fuel cladding). Owing to the fortunate discovery that unintentional impurities from stainless steels during melting of Zr ingots significantly improved their corrosion resistance in high temperature water [50, 51], Zircaloy series of alloys were deployed in the 1950s in the US and Europe. The alloying additions consisted of Sn to getter nitrogen in the alloys [52] and stainless steel constituents for corrosion resistance (Fe and Cr in Zircaloy-4 utilized in PWRs and Ni, Fe and Cr in Zircaloy-2 for use in BWRs). The Russian alloys (E-series) used in the Russian pressurized reactors instead used Nb as the sole alloying element [53]. In almost all cases the alloying element additions totaled <2.5wt%. For PWRs today, the alloying strategy is a hybrid of the early approaches where Nb, Fe, and Cr are added for corrosion resistance and Sn is also retained in these alloys for creep resistance [51, 54]. The BWRs continue to utilize Zircaloy-2 with a high purity Zr inner liner [55] while occasionally alloys with slightly higher Fe contents are also used [56].

The continued evolutionary improvements in Zr alloy cladding for commercial power reactors have resulted in significant improvements in oxidation resistance and hydrogen pickup under normal operating conditions [54]. This provides a significant advantage for durable operational cycles with fuel ^{235}U enrichment capped below the arbitrarily set 5% limit. Aided by extensive and effective fuel reliability programs, higher reactor power and higher fuel burnups have been achieved over the past several decades (enabling enhanced economics of electricity generation). However, fuel operation to beyond 60-70 MWd/kgU average rod burnup for the Zr alloy/ UO_2 fuel system faces performance issues such as excessive irradiation growth (anisotropic stress-free deformation), hydrogen pickup, and cladding oxidation [57]. An additional high burnup degradation concern involves hydrogen pickup in the Zr alloy cladding. The high affinity of Zr for hydrogen results in its dissolution

and subsequent precipitation of hydrides that can result in significant embrittlement [58-60]. Given a fortuitous combination of factors, namely cladding texture and state of stress, during reactor operation these hydride platelet precipitates manifest in the circumferential direction. As the hydrogen content increases at high burnups or during the initial stages of postirradiation storage of the fuel rods in dry casks, these hydrides may reorient in the radial direction [61]. This greatly exacerbates the embrittlement issues in the cladding and is currently a major operational storage and shipping challenge in many countries for ultimate safe disposition of used nuclear fuel from LWRs.

In addition to the operational and fuel disposition concerns, utilization of Zr alloys puts a significant burden on the capacity and capability of safety systems, namely emergency core cooling system (ECCS) in LWRs. The ECCS capabilities in LWRs are determined by evaluation of two types of postulated design-basis accidents: reactivity insertion and loss of coolant accidents (RIA and LOCA) [62]. For RIAs and LOCAs, the extent of enthalpy deposition and peak cladding temperature, respectively, are limited by regulatory guidelines to avoid excessive oxidation of the cladding and subsequent damage that could compromise coolability or controllability of the core. These limits tend to get more restrictive as fuel burnup increases due to the detrimental effects of hydrogen in the cladding. Beyond these postulated scenarios, improbable “black swan” events [63] could occur that are referred to as beyond design-basis accidents or severe accidents. An example of such an event is the 2011 Fukushima Daiichi accident where an earthquake-induced loss of offsite power coincided with eventual loss of emergency onsite power and station blackout. Under such a scenario the ability to adequately remove decay heat from the core is compromised and the increasing core temperatures sequentially surpass the onset of various chemical reactions that result in core degradation. The most consequential of these reactions is the Zr-steam reaction exhibiting very rapid kinetics above 1200°C accompanied by a large enthalpy of oxidation (resulting in core heating). Since steam is the oxidant, hydrogen gas is produced at the same rate that in turn increases the core internal pressure and becomes a source of explosive hazard. To fully appreciate this point, consider that there are ~25-40 tonnes of Zr in LWR cores and each kg of this metal when fully oxidized generates 6.6 MJ of heat and 22 moles of explosive hydrogen (equivalent to 33-53 tonnes of dynamite).

To alleviate the significant burden the uncontrolled oxidation of Zr places on the cooling systems, one may consider alternative cladding materials with reduced oxidation kinetics [64]. Figure 3 compares the short-term high temperature steam oxidation behavior of Zircaloy-4 with two proposed accident-tolerant cladding materials, alumina forming ferritic alloys (FeCrAl) [8] and SiC fiber reinforced SiC matrix ceramic composites [65]. The poor high temperature oxidation behavior of Zircaloy-4 is not improved by switching to newer Zr alloys: Recent high temperature oxidation studies have found strong differences in Zr alloy oxidation behavior occur up to ~1000°C, but only minor differences are observed above 1100°C since here the Zr alloy oxidation is governed by bulk diffusion of oxygen [66]. Both of the proposed new cladding materials exhibit steam oxidation kinetics at least 100× slower than Zr alloys over a broad temperature range up to at least 1400°C [67]. These proposed new cladding materials are by no means fundamentally immune to degradation under LWR severe accidents since under near adiabatic conditions the core temperature will continuously rise to either result in eventual oxidation or melting of these materials. However, the reduced oxidation rate in these materials can be enormously

beneficial since it goes hand-in-hand with a reduced rate of heat and hydrogen generation in the core as a result of this reaction. This leads to a slower rate of temperature rise in the core that is beneficial in two distinct ways. Firstly, it delays the onset of melting and degradation in the core, providing additional coping time that may be exploited to inject coolant into the core and mitigate further progression of the accident. Secondly, the reduced heat generation rate in the core may be small enough that even a limited coolant injection capability could prove sufficient to stabilize or cool the system. These two beneficial effects have been demonstrated using various severe accident analysis tools [68]. Similarly, additional benefits in accident tolerance may be achieved by alternative fuel forms such as triply encased particle fuel in a high conductivity inert matrix [69] or other fuel forms [64]. In combination with enhanced safety features in next-generation LWRs, utilization of accident-tolerant fuels and cladding could potentially eliminate the need for emergency planning public evacuation zones due to miniscule risks of large radiation releases under accident scenarios.

It is mandatory that any proposed accident tolerant fuel system must demonstrate the potential for excellent operational performance under normal operating conditions [70]. Ongoing research is investigating a variety of performance issues including long-term corrosion behavior in LWR water chemistries [71, 72] and potential wear [73] and failure mechanisms [74, 75] under normal operating conditions for several advanced cladding material concepts. Since introduction of any new material to the reactor core might produce unanticipated negative consequences such as enhanced or localized corrosion due to cross-contamination, or lower availability due to initially higher fuel cladding failure rates, a sustained R&D program is important prior to widespread insertion into commercial reactors.

3. Sodium-cooled fast reactors

Sodium-cooled fast reactors (SFRs) offer the potential for significant improvements in fuel utilization and thermodynamic efficiency compared to light water reactors due to their efficient conversion of nonfertile isotopes and higher fuel burnup levels (10-20% vs. <5% for LWRs) and higher operating temperatures (~550°C vs. ~300°C) [3, 76]. Typical uranium utilization in SFRs is ~50-80% compared to ~1% for current light water reactors [3, 76, 77]. Economic competitiveness of this advanced reactor system requires high availability and thermodynamic efficiency to offset anticipated higher construction and fuel recycling costs [76, 78]. This requires the use of very high performance structural materials in the SFR core, where the operational environment is significantly more severe compared to light water reactors.

The two most significant materials issues involve design of the fuel cladding and wrapper/duct materials to simultaneously achieve good thermal creep strength up to 650°C, good radiation resistance (void swelling, irradiation creep, phase stability) up to ~150 to 200 displacements per atom (dpa), and good chemical compatibility with sodium [79] up to ~650°C for in-core residence times of 3 to 7 years [80]. Typical design targets are thermal creep rupture strengths >90-140 MPa for 20,000h operation at ~650°C and <5-10% volumetric void swelling after 150-200 dpa [3, 81-83]. It is noteworthy that amongst all fission reactor designs, the fast reactor has the greatest challenge with respect

to neutron displacement damage. Even higher creep strength and radiation damage resistance would be required for proposed ultra-high burnup reactor concepts such as the Traveling Wave Reactor that would operate for decades between refueling in order to minimize or eliminate reprocessing of spent fuel [80, 84]. The fuel cladding and wrapper/duct must maintain dimensional stability up to high radiation damage levels and avoid fracture during several years of full power operation. The operating temperature and stresses are highest for the fuel cladding, but also represent a significant challenge for the wrapper/duct [83]. In addition to distortions associated with irradiation creep and void swelling, significant operational stresses in the fuel cladding arise due to fuel-cladding mechanical interaction associated with fuel swelling at high burnups along with increased hoop stress due to internal pressurization from release of fission gas [80, 82, 83]. Currently no commercial structural material has been identified that simultaneously satisfies the dual requirement of high thermal creep strength and void swelling/flow localization resistance [82, 83]. Figure 4 compares thermal creep [82, 85, 86] and neutron-induced void swelling behavior [5] for Ti-modified austenitic stainless steel (D9) and for two ferritic/martensitic steels containing 9 and 12%Cr (P91 and HT-9, respectively). Austenitic stainless steels in general exhibit acceptable creep strength but do not provide sufficient high dose radiation resistance. Several commercial ferritic/martensitic steels appear to provide sufficient void swelling resistance to 150 to 200 dpa but do not exhibit acceptable high temperature thermal creep strength.

The most promising approach to meet or exceed the demanding SFR fuel cladding and wrapper design requirements is to utilize computational thermodynamics modeling to design new high performance steels (fabricated using conventional steelmaking processes) that are specifically tailored for improved high temperature thermal creep and radiation resistance [18, 21, 26-30, 87]. Limited experimental data obtained to date on 9Cr ferritic/martensitic steels demonstrate it is possible to double the thermal creep strength at 650°C compared to current conventional ferritic/martensitic steels such as 9Cr-1Mo [18, 21, 26-30, 87] while providing improved radiation resistance and good ductility and fracture toughness [87]. In the long term, oxide dispersion strengthened ferritic/martensitic steels have the potential to provide even better creep strength and radiation resistance [87-90], but reliable and robust joining procedures need further development and fabrication costs associated with the powder metallurgy manufacturing process are currently much higher than for conventional alloy fabrication methods. Oxide dispersion strengthened steels may provide thermal creep strengths that are ~3x that of conventional ferritic/martensitic steels [87, 89], and superior radiation resistance [89, 90].

Additional research is needed on creep-fatigue synergistic damage mechanisms in irradiated steels [91, 92], as well as more detailed studies on mechanical deformation of weldments (in particular Type IV cracking in the heat affected zones) [93, 94]. Cyclic fatigue can occur in SFRs due to (low-cycle) start up and shut down transients as well as (high cycle) coolant thermohydraulic instabilities known as “thermal striping” [95, 96] and other mechanical vibrations. Historically, low cycle fatigue has been governed by ductility and high cycle fatigue has been correlated with strength [91], but the impact of irradiation on cyclic softening and other creep-fatigue processes is not well understood. There is some experimental evidence that traditional post-irradiation tensile, fatigue and creep rupture tests may produce greater degradation compared to in situ tests [97-100], but possible physical mechanisms to explain these effects are still under development.

Additional key SFR materials R&D issues are largely associated with improved thermodynamic efficiency or reliability in order to enhance economic competitiveness [78]. These issues include identification of potential heat exchanger materials for supercritical CO₂ Brayton power conversion systems that have good creep strength and corrosion resistance to sodium and CO₂ [101, 102]. High performance materials to enable advanced steam generators with improved reliability are also an important R&D area [78].

4. Very high temperature reactors (VHTRs)

The first conceptual design for a high temperature (>1000°C) gas cooled reactor (HTGR) was published in 1947 [103]. The design proposed very high temperatures (>1000°C) and a lower power density than today's commercial LWRs, thereby offering significant intrinsic passive system safety advantages but daunting high temperature materials development challenges. Over the subsequent decades a number of commercial system evolutions have occurred in graphite moderated reactors, each slowly approaching the safety and system efficiency promised by that original HTGR design.

Figure 5 summarizes the operating temperatures and construction and operation dates for gas-cooled reactors worldwide. The first commercial graphite-block moderated reactors were the British Generation-1 Magnox design: CO₂-cooled reactors utilizing a magnesium alloy "pin-type" cladding encasing natural uranium fuel. To improve safety and thermal efficiency, the 99Mg-0.8Al magnesium-based cladding was replaced by a higher-temperature (and higher neutron absorbing) 20Cr-25Ni stainless steel, allowing reactor exit gas temperature to increase from ~360°C to ~630°C. The design temperature limit was controlled by excessive oxidation by the CO₂ coolant rather than steel softening considerations. To compensate for the additional neutron scavenging induced by the steel compared to the magnesium alloy the fuel in these reactors was enriched in fissile U-235. These Gen-2 plants, known as Advanced Gas Cooled Reactors (AGCR's), along with the original Gen-1 plants, provided decades of base-load power in the UK and elsewhere in Europe.

Transition from AGCR to HTGR and ultimately VHTR is limited by availability of appropriate engineering materials for the pressure vessel, intermediate heat exchanger, and core internals, as well as the associated code qualification of those materials and high-temperature design methodologies allowing engineers to carry out appropriate design. While the operating temperature and vessel size for the gas-cooled reactors are larger than for the LWR or SFR, the benefits in adopting new high performance RPV steels are essentially identical with the other reactor platforms. Continuing the trend toward higher efficiency power plants (Fig. 5), the first Generation-III HTGR demonstration plants emerged with the Peach Bottom reactor (1966-1974) operating at ~750°C being the first example. That reactor utilized stainless steel pin-type fuel containing graphite compacts of "BISO" (bistructural isotropic) particle fuel consisting of two concentric layers of pyrolytic carbon surrounding the fuel kernel. Subsequently a number of experimental HTGRs followed in the UK (Dragon), Germany (AVR), Japan (HTTR) and China (HTR-10), each moving from the non-encapsulated BISO fuel to a tri-structural "TRISO" fuel that utilized a SiC micro-pressure vessel surrounding the fuel kernel (between two layers of pyrolytic carbon) for primary fission product containment. These early experimental HTGRs further

pushed the operating temperature envelope, entirely eliminating metallic pin-type fuels, opting for either spherical graphite pebbles of entrained TRISO (the Pebble Bed Modular Reactor Design) or cylindrical graphite compacts of entrained TRISO (the Prismatic Design) within interlocked graphite blocks. The majority of the core was graphite and ceramic fuel, essentially limiting the use of metallic structures within the nuclear core to the control system and the pressure vessel.

The research carried out in these experimental Generation-III HTGRs led to a few demonstration power plants (the Ft. St. Vrain reactor in the US and the THTR in Germany), though neither operated very long due in large part to engineering issues with the plants. Since that time a number of international research programs have considered Generation-III HTGRs and Generation-IV VHTRs (characterized by helium outlet temperatures of greater than $\sim 900^{\circ}\text{C}$) for the production of electricity and process heat. One such program has been the International Gen IV nuclear energy program. Other, purely commercial enterprises include the now defunct South African Pebble Bed Modular Reactor (PBMR) project (Gen III HTGR) and the ongoing Chinese HTR-PM HTGR reactor, which has been under construction since 2012.

With the elimination of metallic fuel cladding, the remaining VHTR metallic core internals include control rods, control-rod guides, upper plenum shroud, and hot-duct liner. In addition to these in-core materials, a suitable high-performance creep-resistant heat exchanger material would be needed in order to achieve desired high operating temperatures. In all cases, use of computational thermodynamics tools to guide an expedited high-performance high-temperature alloy development program would be valuable. In order to further increase plant efficiency, becoming true HTGRs or VHTRs, either new, higher-temperature metallic alloys or structural ceramics would become necessary. However, due in part to the current lack of qualification for “building blocks” of the gas-cooled reactors, such as the core graphite and the TRISO fuel, programs such as the U.S. Next Generation Nuclear Plant (NGNP) program and the former South African pebble bed reactor project focused significant resources on these primary needs (although neither project completed qualification of the fuel or graphite). As a compromise, poorly performing *historic* structural alloys such as a SA-508 Grade 3 (pressure vessel) or Alloy 800H (reactivity control rods, heat exchanger and other applications) were selected for the reference design and investigated for incremental performance extension.

The continued attraction of materials such as Alloy 800H [104], an oxidation resistant and higher creep strength variant of the Fe-32.5Ni-21Cr Incoloy 800 (circa 1965) follows a similar theme as that for the Zircaloy-2 and 4 (circa 1958) cladding for the LWR, the HT-9 (circa 1960's) fuel clad of fast reactors and the SA-508 Grade 3 (circa 1965) steel utilized for RPVs: They are code-qualified historic alloys of proven engineering application, though arguably of inferior performance to modern engineering alloys. As mentioned, the historic Alloy 800H is being evaluated for possible extension in HTGR and VHTR concepts to allow use at temperatures above the current code limits [104]. Currently, the alloy is qualified to 760°C for nuclear applications, though a draft German standard [105] allowed use to 1100°C . By inspection of Figure 6, for the operating temperature regime of the HTGR and VHTR ($> 700^{\circ}\text{C}$) the yield and creep rupture strength of alloy 800H is a fraction of alternative alloys such as Alloy X (XR), Alloy 230, or Alloy 617 [106]. It is noted that some of these alternative alloys are being investigated for potential VHTR applications (e.g., Alloy X for hot duct liner in Japanese HTTR program and Alloy 617 for intermediate heat

exchangers for the NGNP program). However, code qualification efforts of these alternative alloys is sporadic.

One drawback of utilizing the historic materials, as suggested in Figure 5, is that the current development path for gas-cooled reactors has essentially approached an asymptotic performance temperature limit defined by the code qualification limits of the in-core and RPV alloys. Higher performance VHTR materials are being explored at a relatively modest effort including performance testing and engineering design code work. As example the PBMR program fabricated and carried out a testing program on carbon fiber reinforced composite (CFRC) [107, 108] reflector tie rods and lateral restraint straps and the International Generation IV program has examined SiC/SiC composites and CFRCs for a number of applications such as core support and stabilizers and control-rod guides [109-112]. These targeted introductions of composites for high-value components are similar to the approach used for introducing composites into commercial aircraft. The code qualification of such nuclear composite materials is ongoing, at a low level, through an ASME working group and in a number of associated testing standards development under ASTM Committee C28 on advanced ceramics [113].

The motivation for moving to such composites, analogous with introduction of ceramic composites in today's aircraft engines (such as SiC composite compressor shroud in the GE LEAP engine and soon the inner and outer combustor liner for the GE9X engine), is their superior high-temperature performance as well as low mass. Furthermore, nuclear-grade SiC composites have another significant advantage over both historic and modern structural alloys. Specifically, Alloy 800H [114] and the general class of nickel-based superalloys [115] suffer from very low neutron dose embrittlement, essentially requiring expensive component replacement during the lifetime of the reactor. This contrasts with SiC composites, which have exceptional irradiation stability, allowing lifetime components [65, 116].

5. Recommendations

Advanced materials offer numerous performance, economic, environmental and safety advantages for current and future nuclear power systems. The perceived regulatory barriers and economic risks associated with the introduction of unproven materials can be largely eliminated through sustained (decadal) nonproprietary or federally funded research and development prior to their introduction in commercial power stations. Conversely, historical evidence from other industries suggests lack of introduction of improved materials (and corresponding design innovations) will eventually lead to obsolescence.

For current and next-generation light water reactors, R&D to identify suitable accident tolerant fuel systems and deep-burn fuel concepts offer significant promise to improve safety under low-probability accident conditions and to reduce waste disposal burdens. For next-generation reactor concepts, this could greatly reduce or possibly eliminate the need for emergency planning public evacuation zones around reactors. Introduction of new reactor pressure vessel steels that leverage the significant advances since the 1960s in design of high-performance low-alloy steels and the improved understanding of methods to suppress radiation damage degradation of fracture toughness could provide

simultaneous fabrication savings and improved performance. Similar advantages could be achieved with advanced in-core structural materials to replace early-generation stainless steels and nickel alloys.

For sodium-cooled fast reactors, new high-performance steels designed using computational thermodynamics or based on oxide dispersion strengthening offer a viable solution to the current capability gap in conventional austenitic and ferritic/martensitic steels of simultaneous high thermal creep strength and high-dose void swelling resistance that is needed for economic competitiveness of SFRs.

The low power densities in VHTRs provide significant safety advantages during loss of coolant accident scenarios, but require high-performance creep-resistant materials for the pressure vessel and core internal structural materials such as control rod drive mechanisms. Ceramic composites offer the best option for core internal structures that would enable realization of the initial vision of operating temperatures near 1000°C.

In contrast to the shortening time scales to develop and deploy materials in non-nuclear industries [117], the span of time required for the adoption of new materials in the nuclear industry has expanded significantly and typically is greater than the duration of most nuclear project development phases or projects themselves. For this reason, along with the regulatory complexities of the nuclear material qualification process, there is no clear path to advanced nuclear materials development. The result, as evidenced by the re-application of tried-and-true but marginally adequate materials, is incremental improvements in our nuclear energy systems and a hollowing-out of domestic nuclear industry and research laboratory ability to design and develop advanced nuclear materials into commercial application. To address this situation, one solution would be to identify a number of key enabling technologies and formulate a roadmap to field these technologies in public/private partnership separate from any reactor program. There have been some encouraging recent steps to enhance public-private partnerships for nuclear energy R&D such as small modular reactor development and innovative new reactor concepts that have attracted venture capital support. Example technologies to codify would include adoption of modern steels for pressure vessel application, adoption of modern ferritic martensitic steel for fast reactor application, and the general area of high temperature design methodology and designing with ceramic composite for gas-cooled reactor application. Such a program should necessarily include multidisciplinary teams including materials engineers, the regulatory agencies, reactor designers and systems engineering experts and have a narrow focus on formulating and executing structural engineering code cases.

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References

[1] Grimes RW, Konings RJM, Edwards L. Greater tolerance for nuclear materials. *Nature Mater.* 2008;7:683-885.

- [2] Raj B, Vijayalakshmi M, Rao PRV, Rao KBS. Challenges in materials research for sustainable nuclear energy. *MRS Bull.* 2008;33:327-37.
- [3] Yvon P, Carre F. Structural materials challenges for advanced reactor systems. *J Nucl Mater.* 2009;385:217-22.
- [4] Zinkle SJ, Busby JT. Structural Materials for Fission and Fusion Energy. *Mater Today.* 2009;12:12-9.
- [5] Zinkle SJ, Was GS. Materials challenges in nuclear energy. *Acta Mater.* 2013;61:735-58.
- [6] Yvon P, Le Flem M, Cabet C, Seran JL. Structural materials for next generation nuclear systems: Challenges and the path forward. *Nucl Eng Des.* 2015;294:161-9.
- [7] Hankey R, Cassar C, Peterson R, Wong P, Yildiz O. Electric Power Monthly with data for February 2016. U.S. Energy Information Administration, <https://www.eia.gov/electricity/monthly/pdf/epm.pdf>; 2016.
- [8] Terrani KA, Zinkle SJ, Snead LL. Advanced oxidation-resistant iron-based alloys for LWR fuel cladding. *J Nucl Mater.* 2014;448:420-35.
- [9] Odette GR, Lucas GE. Embrittlement of Nuclear Reactor Pressure Vessels. *JOM.* 2001;53:18-22.
- [10] Rensman J. NRG Irradiation Testing: Report on 300°C and 60°C Irradiated RAFM Steels. NRG report 20023/0568497/P. Petten, The Netherlands: NRG; 2005.
- [11] Stork D, Agostini P, Boutard JL, Buckthorpe D, Diegele E, Dudarev SL, et al. Developing structural, high-heat flux and plasma facing materials for a near-term DEMO fusion power plant: The EU assessment. *J Nucl Mater.* 2014;455:277-91.
- [12] Server WL, Nanstad RK. Reactor pressure vessel (RPV) design and fabrication: the case of the USA. In: Soneda N, editor. *Irradiation Embrittlement of Reactor Pressure Vessels.* Cambridge: Woodhead Publ Ltd; 2015. p. 3-25.
- [13] Viswanathan R, Henry JF, Tanzosh J, Stanko G, Shingledecker J, Vitalis B, et al. US Program on Materials Technology for Ultra-Supercritical Coal Power Plants. *J Mater Eng Perf.* 2013;22:2904-15.
- [14] Boyer RR, Cotton JD, Mohaghegh M, Schafrik RE. Materials considerations for aerospace applications. *MRS Bull.* 2015;40:1055-65.
- [15] Roeseler WG, Sarh B, Kismarton MU. Composite structures: The first 100 years. In: Kageyama K, Ishikawa T, Takeda N, Hojo M, Sugimoto S, Ogasawara T, editors. *16th International Conference on Composite Materials.* Kyoto, Japan: Japan Society for Composite Materials; 2007.
- [16] Olson GB. Computational design of hierarchically structured materials. *Science.* 1997;277:1237-42.
- [17] Andersson JO, Helander T, Hoglund LH, Shi PF, Sundman B. THERMO-CALC & DICTRA, computational tools for materials science. *Calphad-Comp Coupling Phase Diag Thermoch.* 2002;26:273-312.
- [18] Abe F. Precipitate design for creep strengthening of 9% Cr tempered martensitic steel for ultra-supercritical power plants. *Sci Technol Adv Mater.* 2008;9:013002:013002 pp.
- [19] Yamamoto Y, Santella ML, Brady MP. Effect of Alloying Additions on Phase Equilibria and Creep Resistance of Alumina-Forming Austenitic Stainless Steels. *Metall Mater Trans A.* 2009;40A:1868-80.
- [20] Abe F. Alloy Design of Creep- and Oxidation-resistant 9%Cr Steel for High Efficiency USC Power Plant. *Mater Sci Forum.* 2012;706-709:3-8.

- [21] Sachadel UA, Morris PF, Clarke PD. Design of 10%Cr martensitic steels for improved creep resistance in power plant applications. *Mater Sci Technol*. 2013;29:767-74.
- [22] Tancret F. Computational thermodynamics, Gaussian processes and genetic algorithms: combined tools to design new alloys. *Model Simul Mater Sci Eng*. 2013;21:045013:045013 pp.
- [23] Raabe D, Ponge D, Dmitrieva O, Sander B. Designing Ultrahigh Strength Steels with Good Ductility by Combining Transformation Induced Plasticity and Martensite Aging. *Adv Eng Mater*. 2009;11:547-55.
- [24] Xu W, Rivera-Diaz-Del-Castillo PEJ, Yan W, Yang K, San Martin D, Kestens LAI, et al. A new ultrahigh-strength stainless steel strengthened by various coexisting nanoprecipitates. *Acta Mater*. 2010;58:4067-75.
- [25] Raabe D, Tasan CC, Springer H, Bausch M. From High-Entropy Alloys to High-Entropy Steels. *Steel Res Int*. 2015;86:1127-38.
- [26] Klueh RL, Hashimoto N, Maziasz PJ. New nano-particle-strengthened ferritic/martensitic steels by conventional thermo-mechanical treatment. *J Nucl Mater*. 2007;367-370:48-53.
- [27] Hollner S, Piozin E, Mayr P, Caes C, Tournie I, Pineau A, et al. Characterization of a boron alloyed 9Cr3W3CoVNbBN steel and further improvement of its high-temperature mechanical properties by thermomechanical treatments. *J Nucl Mater*. 2013;441:15-23.
- [28] Tan L, Busby JT, Maziasz P, Yamamoto Y. Effect of thermomechanical treatment on 9Cr ferritic-martensitic steels. *J Nucl Mater*. 2013;441:713-7.
- [29] Tan L, Byun TS, Katoh Y, Snead LL. Stability of MX-type strengthening nanoprecipitates in ferritic steels under thermal aging, stress and ion irradiation. *Acta Mater*. 2014;71:11-9.
- [30] Tan L, Snead LL, Katoh Y. Development of new generation reduced activation ferritic-martensitic steels for advanced fusion reactors. *J Nucl Mater*. 2016;478:42-9.
- [31] Alvin MA, Anderson I, Heidloff A, White E, Bhatt R, Grady J, et al. Development of Advanced Material Systems for Future Gas Turbine Applications. New York: Amer Soc Mechanical Engineers; 2015.
- [32] Scheithauer U, Slawik T, Schwarzer E, Richter HJ, Moritz T, Michaelis A. Additive Manufacturing of Metal-Ceramic-Composites by Thermoplastic 3D-Printing (3DTP). *J Ceram Sci Technol*. 2015;6:125-31.
- [33] Bourell DL. Perspectives on Additive Manufacturing. In: Clarke DR, editor. *Annual Review of Materials Research*, Vol 46. Palo Alto: Annual Reviews; 2016. p. 1-18.
- [34] Light Water Reactor Sustainability Program: Integrated Program Plan, INL/EXT-11-23452. Office of Nuclear Energy, US Dept. of Energy; 2012.
- [35] Christensen LR, Greene WH. Economies of scale in United States electric-power generation. *The Journal of Political Economy*. 1978;84:655-76.
- [36] Krautmann AC, Solow JL. Economies of scale in nuclear-power generation. *South Econ J*. 1988;55:70-85.
- [37] Cantor R, Hewlett J. The economics of nuclear-power - further evidence on learning, economies of scale, and regulatory effects. *Resources and Energy*. 1988;10:315-35.
- [38] Carelli MD, Garrone P, Locatelli G, Mancini M, Mycoff C, Trucco P, et al. Economic features of integral, modular, small-to-medium size reactors. *Prog Nucl Energy*. 2010;52:403-14.
- [39] Boarin S, Locatelli G, Mancini M, Ricottia ME. Financial case studies on small- and medium-size modular reactors. *Nucl Technol*. 2012;178:218-32.

- [40] Klueh RL, Alexander DJ, Maziasz PJ. Bainitic chromium-tungsten steels with 3 pct chromium. *Metall Mater Trans A*. 1997;28A:335-45.
- [41] Klueh RL, Evans ND, Maziasz P, Sikka VK. Creep-rupture behavior of 3Cr-3W-V bainitic steels. *Int J Pressure Vessels and Piping*. 2007;84:29-36.
- [42] Klueh RL, Nelson AT. Ferritic/martensitic steels for next generation reactors. *J Nucl Mater*. 2007;371:37-52.
- [43] El-Guebaly L, Huhn T, Rowcliffe A, Malang S. Design challenges and activation concerns for ARIES vacuum vessel. *Fusion Sci Technol*. 2013;64:449-54.
- [44] Blagoeva DT, Debarberis L, Jong M, ten Pierick P. Stability of ferritic steel to higher doses: Survey of reactor pressure vessel steel data and comparison with candidate materials for future nuclear systems. *Int J Pressure Vessels and Piping*. 2014;122:1-5.
- [45] Was GS, Ashida Y, Andresen PL. Irradiation-assisted stress corrosion cracking. *Corros Rev*. 2011;29:7-49.
- [46] Hojna A. Irradiation-Assisted Stress Corrosion Cracking and Impact on Life Extension. *Corrosion*. 2013;69:000173:11 pp.
- [47] McMurtrey MD, Cui B, Robertson I, Farkas D, Was GS. Mechanism of dislocation channel-induced irradiation assisted stress corrosion crack initiation in austenitic stainless steel. *Curr Opin Solid State Mat Sci*. 2015;19:305-14.
- [48] Gorman JA. 2015 Frank Newman Speller Award: Stress Corrosion Cracking and Nuclear Power. *Corrosion*. 2015;71:1414-33.
- [49] Pathania R. Critical Issues Report and Roadmap for the Advanced Radiation-Resistant Materials Program, Report 1026482. Palo Alto, CA: EPRI; 2012.
- [50] Rickover HG, Geiger LD, Lustman B. History of the development of zirconium alloys for use in nuclear reactors, NR:D:1975, TID-26740. US ERDA, Division of Naval Reactors; 1975.
- [51] Adamson RB. Zirconium Production and Technology: The Kroll Medal Papers 1975–2010. West Conshohocken, PA: ASTM International; 2010.
- [52] Garde AM, Pati SR, Krammen MA, Smith GP, Endter RK. Corrosion behavior of Zircaloy-4 cladding with varying tin content in high-temperature pressurized water reactors. In: Garde AM, Bradley ER, editors. *Zirconium in the Nuclear Industry: 10th International Symposium*, ASTM STP 1245. West Conshohocken, PA: American Society for Testing and Materials; 1994. p. 760-78.
- [53] Shishov V. The evolution of microstructure and deformation stability in Zr-Nb-(Sn, Fe) alloys under neutron irradiation. In: Limbäck M, Barberis P, editors. *Zirconium in the Nuclear Industry*. West Conshohocken: ASTM International; 2012. p. 37-66.
- [54] Motta A, Couet A, Comstock RJ. Corrosion of zirconium alloys used for nuclear fuel cladding. *Annu Rev Mater Res*. 2015;45:311-43.
- [55] Armijo JS, Coffin LF, Rosenbaum HS. Development of zirconium-barrier fuel cladding. In: Garde AM, Bradley ER, editors. *Zirconium in the Nuclear Industry: 10th International Symposium*, ASTM STP 1245. West Conshohocken, PA: American Society for Testing and Materials; 1994. p. 3-18.
- [56] Une K, Sakamoto K, Aomi M, Matsunaga J, Etoh Y, Takagi I, et al. Hydrogen absorption mechanism of zirconium alloys based on characterization of oxide layer. In: Limbäck M, Barberis P, editors. *Zirconium in the Nuclear Industry*. West Conshohocken: ASTM International; 2012. p. 401-30.

- [57] Ledergerber G, Valizadeh S, Wright JL, Limbäck M, Hallstadius L, Gavillet D, et al. Fuel performance beyond design–Exploring the limits. LWR Fuel Performance Meeting Top Fuel/WRFPM American Nuclear Society; 2010. p. 513-24.
- [58] Garde AM, Smith GP, Pirek RC. Effects of hydride precipitate localization and neutron fluence on the ductility of irradiated Zircaloy-4. In: Bradley ER, Sabol GP, editors. Zirconium in the Nuclear Industry: 11th International Symposium, ASTM STP 1295. W Conshohocken, PA: American Society Testing and Materials; 1996. p. 407-30.
- [59] Kammenzind BF, Franklin DG, Peters HR, Duffin WJ. Hydrogen pickup and redistribution in alpha-annealed Zircaloy-4. In: Bradley ER, Sabol GP, editors. Zirconium in the Nuclear Industry: 11th International Symposium, ASTM STP 1295. W Conshohocken, PA: American Society Testing and Materials; 1996. p. 338-70.
- [60] Motta AT, Chen LQ. Hydride Formation in Zirconium Alloys. JOM. 2012;64:1403-8.
- [61] Colas K, Motta A, Daymond MR, Almer J. Mechanisms of Hydride Reorientation in Zircaloy-4 Studied in Situ. In: Comstock R, Barberis P, editors. Zirconium in the Nuclear Industry. West Conshohocken: ASTM International; 2015. p. 1107-37.
- [62] 10CFR Part 50–Domestic licensing of production and utilization facilities. US Nuclear Regulatory Commission.
- [63] Taleb NN. The Black Swan: The Impact of the Highly Improbable. New York: Random House; 2007.
- [64] Zinkle SJ, Terrani KA, Gehin JC, Snead LL. Accident tolerant fuels for LWRs: A perspective. J Nucl Mater. 2014;448:374-9.
- [65] Katoh Y, Ozawa K, Shih C, Nozawa T, Shinavski RJ, Hasegawa A, et al. Continuous SiC fiber, CVI SiC matrix composites for nuclear applications: Properties and irradiation effects. J Nucl Mater. 2014;448:448-76.
- [66] Steinbrück M, Ver N, Grosse M. Oxidation of Advanced Zirconium Cladding Alloys in Steam at Temperatures in the Range of 600-1200 degrees C. Oxid Metals. 2011;76:215-32.
- [67] Pint BA, Terrani KA, Yamamoto Y, Snead LL. Material Selection for Accident Tolerant Fuel Cladding. Metallurgical and Materials Transactions E. 2015;2E:190-6.
- [68] Robb KR. Analysis of the FeCrAl Accident Tolerant Fuel Concept Benefits During BWR Station Blackout Accidents. Proc 16th Intern Topical Meeting on Nuclear Reactor Thermal Hydraulics, NURETH-162015. p. 1183-95.
- [69] Terrani KA, Snead LL, Gehin JC. Microencapsulated fuel technology for commercial light water and advanced reactor application. J Nucl Mater. 2012;427:209-24.
- [70] Carmack J, Goldner F. Forward for special JNM issue on accident tolerant fuels for LWRs Preface. J Nucl Mater. 2014;448:373-.
- [71] Terrani KA, Yang Y, Kim YJ, Rebak R, Meyer HM, Gerczak TJ. Hydrothermal corrosion of SiC in LWR coolant environments in the absence of irradiation. J Nucl Mater. 2015;465:488-98.
- [72] Terrani KA, Pint BA, Kim Y-J, Unocic KA, Yang Y, Silva CM, et al. Uniform corrosion of FeCrAl alloys in LWR coolant environments. J Nucl Mater. 2016;479:36-47.
- [73] Lee YH, Byun TS. A comparative study on the wear behaviors of cladding candidates for accident-tolerant fuel. J Nucl Mater. 2015;465:857-65.
- [74] Ben-Belgacem M, Richet V, Terrani KA, Katoh Y, Snead LL. Thermo-mechanical analysis of LWR SiC/SiC composite cladding. J Nucl Mater. 2014;447:125-42.

- [75] Stone JG, Schleicher R, Deck CP, Jacobsen GM, Khalifa HE, Back CA. Stress analysis and probabilistic assessment of multi-layer SiC-based accident tolerant nuclear fuel cladding. *J Nucl Mater.* 2015;466:682-97.
- [76] Locatelli G, Mancini M, Todeschini N. Generation IV nuclear reactors: Current status and future prospects. *Energy Policy.* 2013;61:1503-20.
- [77] A Technology Roadmap for Generation IV Nuclear Energy Systems. Generation IV International Forum; 2002.
- [78] GIF R&D Outlook for Generation IV Nuclear Energy Systems. Generation IV International Forum; 2009.
- [79] Furukawa T, Yoshida E. Material Performance in Sodium. In: Konings RJM, editor. *Comprehensive Nuclear Materials, Vol 5: Material Performance and Corrosion/Waste Materials.* Amsterdam: Elsevier; 2012. p. 327-41.
- [80] Nakae N, Ozawa T, Ohta H, Ogata T, Sekimoto H. An approach for evaluating the integrity of fuel applied in Innovative Nuclear Energy Systems. *J Nucl Mater.* 2014;446:1-9.
- [81] Bennett JW, Horton KE. Materials Requirements for Liquid Metal Fast Breeder Reactors. *Metall Trans A.* 1978;9A:143-9.
- [82] Cheon JS, Lee CB, Lee BO, Raison JP, Mizuno T, Delage F, et al. Sodium fast reactor evaluation: Core materials. *J Nucl Mater.* 2009;392:324-30.
- [83] Martin P, Pelletier M, Every D, Buckthorpe D. French and United Kingdom experience of high-burnup mixed-oxide fuel in sodium-cooled fast breeder reactors. *Nucl Technol.* 2008;161:35-44.
- [84] Weaver KD, Gilleland J, Ahlfeld C, Whitmer C, Zimmerman G. A Once-Through Fuel Cycle for Fast Reactors. *J Eng Gas Turbines Power-Trans ASME.* 2010;132:102917:6 pp.
- [85] Kimura K. 9Cr-1Mo-V-Nb steel. In: Yagi K, Merckling G, Kern TU, Irie H, Warlimont H, editors. *Materials.* Berlin: Springer; 2004. p. 126-33.
- [86] Latha S, Mathew MD, Parameswaran P, Rao KBS, Mannan SL. Creep behaviour of 14Cr-15Ni-Ti stainless steel at 923 K. *Mater Sci Eng A.* 2010;527:5167-74.
- [87] Zinkle SJ, Boutard JL, Hoelzer DT, Kimura A, Lindau R, Odette GR, et al. Development of Next Generation Tempered and ODS Reduced Activation Ferritic/Martensitic Steels for Fusion Energy Applications. *Nucl Fusion.* 2016;in press.
- [88] Dubuisson P, De Carlan Y, Garat V, Blat M. ODS Ferritic/martensitic alloys for Sodium Fast Reactor fuel pin cladding. *J Nucl Mater.* 2012;428:6-12.
- [89] Ukai S. Oxide Dispersion Strengthened Steels. In: Konings RJM, editor. *Comprehensive Nuclear Materials, Vol 4: Radiation Effects in Structural and Functional Materials for Fission and Fusion Reactors.* Amsterdam: Elsevier; 2012. p. 241-71.
- [90] Odette GR. Recent Progress in Developing and Qualifying Nanostructured Ferritic Alloys for Advanced Fission and Fusion Applications. *JOM.* 2014;66:2427-41.
- [91] Hoffelner W. Damage assessment in structural metallic materials for advanced nuclear plants *J Mater Sci.* 2010;45:2247-52.
- [92] Raj B, Choudhary BK. A perspective on creep and fatigue issues in sodium cooled fast reactors. *Trans Indian Inst Met.* 2010;63:75-84.
- [93] Li YK, Hongo H, Tabuchi M, Takahashi Y, Monma Y. Evaluation of creep damage in heat affected zone of thick welded joint for Mod.9Cr-1Mo steel. *Int J Pressure Vessels and Piping.* 2009;86:585-92.
- [94] Raj B, Vijayalakshmi M. Ferritic Steels for Sodium-cooled Fast Reactors: Design Principles and Challenges. *JOM.* 2010;62:75-83.

- [95] Lee JI, Hu LW, Saha P, Kazimi MS. Numerical analysis of thermal striping induced high cycle thermal fatigue in a mixing tee. *Nucl Eng Des.* 2009;239:833-9.
- [96] Chellapandi P, Velusamy K. Thermal hydraulic issues and challenges for current and new generation FBRs. *Nucl Eng Des.* 2015;294:202-25.
- [97] Lovell AJ, Chin BA, Gilbert ER. In-reactor creep rupture of 20% cold-worked AISI 316 stainless steel. *J Mater Sci.* 1981;16:870-6.
- [98] Marmy P. In-beam mechanical testing of CuCrZr. *J Nucl Mater.* 2004;329:188-92.
- [99] Peacock AT, Barabash V, Danner W, Rodig M, Lorenzetto P, Marmy P, et al. Overview of recent European materials R&D activities related to ITER. *J Nucl Mater.* 2004;329:173-7.
- [100] Bloom EE, Busby JT, Duty CE, Maziasz PJ, McGreevy TE, Nelson BE, et al. Critical questions in materials science and engineering for successful development of fusion power. *J Nucl Mater.* 2007;367-370:1-10.
- [101] Rouillard F, Charton F, Moine G. Corrosion Behavior of Different Metallic Materials in Supercritical Carbon Dioxide at 550 degrees C and 250 bars. *Corrosion.* 2011;67:095001:7 pp.
- [102] Pint BA, Keiser JR. Initial Assessment of Ni-Base Alloy Performance in 0.1 MPa and Supercritical CO₂. *JOM.* 2015;67:2615-20.
- [103] McCullough CR. Summary Report on the Design and Development of High Temperature Gas-Cooled Power Pile. Oak Ridge, TN: Clinton Laboratory; 1947. p. 1-313.
- [104] Ren WJ, Swindeman R. Status of Alloy 800H in Considerations for the Gen IV Nuclear Energy Systems. *J Press Vessel Technol-Trans ASME.* 2014;136:054001:12 pp.
- [105] Schubert F, Breitbach G, Nickel H. German Structural Design Rule KTA 3221 for Metallic HTR-Components. High Temperature Service and Time-Dependent Failure, ASME-PVP. 1993;262:9-18.
- [106] Ren W, Swindeman RW. A Review of Alloy 800H for Applications in the Gen IV Nuclear Energy Systems. *Proceedings of the ASME Pressure Vessels and Piping Conference 2010, Vol 6.* New York: Amer Soc Mechanical Engineers; 2010. p. 821-36.
- [107] Fazluddin S, Smid K, Slabber J. The use of advanced materials in VHTR's, paper E06. 2nd International Topical Meeting on High Temperature Reactor Technology. Beijing: Institute of Nuclear and New Energy Technology, Tsinghua University; 2004.
- [108] Fazluddin S, Mitchell M. Application of Composite Materials in the PBMR. In: Anandjiwala RD, John MJ, editors. *Fibre Reinforced Composites Conference 2007.* Port Elizabeth, South Africa: South Africa Dept. of Science & Technology; 2007.
- [109] Corwin WR. U.S. Generation IV reactor integrated materials technology program. *Nucl Engr Technol.* 2006;38:591-618.
- [110] Corwin WR, Ballinger R, Majumdar S, Weaver KD. Next Generation Nuclear Plant Phenomena Identification and Ranking Tables (PIRTs): Volume 4: High-Temperature Materials PIRTs, NUREG/CR-6944. 2007.
- [111] Corwin WR, Burchell TD, Halsey WG, Hayner GO, Katoh Y, Klett JW, et al. Updated Generation IV Reactors Integrated Materials Technology Program Plan, Revision 2, ORNL/TM-2005/556. Oak Ridge National Lab; 2005.
- [112] Corwin WR, Burchell TD, Ghoniem NM, Katoh Y, McGreevy TE, Morgan D, et al. Generation IV Reactors Integrated Materials Technology Program Plan: Focus on Very High Temperature Reactor Materials, ORNL/TM-2008/129. 2008.
- [113] ASTM. Carbon-carbon composite specification–Draft 1, E-10. 2013.

- [114] Thiele BA, Diehl H, Ohly W, Weber H. Investigations into the irradiation behavior of high-temperature alloys for high-temperature gas-cooled reactor applications. Nucl Technol. 1984;66:597-606.
- [115] Boothby RM. Radiation Effects in Nickel-Based Alloys. In: Konings RJM, editor. Comprehensive Nuclear Materials. Amsterdam: Elsevier; 2012. p. 123-50.
- [116] Snead LL, Katoh Y, Windes WE, Shinavski RJ, Burchell TD. Ceramic composites for near term reactor application. Proceedings of the 4th International Topical Meeting on High Temperature Reactor Technology - 2008, Vol 2. New York: Amer Soc Mechanical Engineers; 2009. p. 1-10.
- [117] Schafrik RE. Materials for a Non-Steady-State World. Metall Mater Trans B. 2016;47:1505-15.

List of figures

Fig. 1. Comparison of average U.S. capacity factors for utility-scale generators in the years 2013-2015 (most recent available data) [7].

Fig. 2. Development timeline for structural materials in light water reactors. The filled diamond symbols denote the approximate date of alloy commercialization, and the horizontal bars denote the approximate duration of use in light water reactors.

Fig. 3. Current and proposed advanced nuclear fuel cladding after 4 h of exposure in flowing steam (0.5 m/s): a) Zircaloy-4 at 1400°C, b) Fe-10Cr-6Al-0.3Y at 1400°C, and chemical vapor infiltrated SiC/SiC composite at 1600°C. While the entire 0.6 mm-thick Zr-based cladding has undergone full oxidation, a few-micrometer-thick scale of alumina and silica protects the ferritic alloy and the ceramic composite, respectively at these high temperature test conditions.

Fig. 4. a) Comparison of thermal creep rupture behavior of Ti-modified austenitic stainless steel [86] and 9-12%Cr ferritic/martensitic steels at 650°C [82, 85]. b) Comparison of the dose dependence of neutron-induced volumetric void swelling in cold-worked stainless steel, Ti-modified austenitic stainless steel and 9-12%Cr ferritic/martensitic steels [5]. The typical desired design lifetime conditions for SFR fuel cladding [3, 81, 82] are shown in both figures. The relationship between operating time and displacement damage assumes a typical core damage rate of 1×10^{-6} dpa/s.

Fig. 5. Evolution in operating temperature for gas-cooled reactors.

Fig. 6: Average yield strength and 100,000 hour rupture strength of candidate in-core gas-cooled reactor structural alloys (from ref. [106] and vendor-supplied property data).

Figure 1

Figure 2

Figure 3

(a)

Zircaloy-4

(b)

FeCrAl

(c)

SiC/SiC

Figure 4a

Figure 4b

Figure 5

Figure 6