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Sodium fast reactor evaluation: Core materials

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ABSTRACT

In the framework of the Generation IV Sodium Fast Reactor (SFR) Program the Advanced Fuel Project has conducted an evaluation of the available fuel systems supporting future sodium cooled fast reactors. In this paper the status of available and developmental materials for SFR core cladding and duct applications is reviewed. To satisfy the Generation IV SFR fuel requirements, an advanced cladding needs to be developed. The candidate cladding materials are austenitic steels, ferritic/martensitic (F/M) steels, and oxide dispersion strengthened (ODS) steels. A large amount of irradiation testing is required, and the compatibility of cladding with TRU-loaded fuel at high temperatures and high burnup must be investigated. The more promising F/M steels (compared to HT9) might be able to meet the dose requirements of over 200 dpa for ducts in the GEN-IV SFR systems.

1. Introduction

In the sodium fast reactors (SFR) envisioned in the Generation IV program, fuel pins are expected to be irradiated at a higher temperature to a higher burnup than in previously operated reactors. Core materials of concern here are fuel components such as the cladding and duct. Integrity of the fuel pins strongly depends on whether the cladding can withstand the irradiation environment. It is preferred that the cladding have a low swelling and high ductility throughout its lifetime. Sufficient creep strength at a high temperature is required. The materials for the duct must retain sufficient strength and toughness to allow duct handling after irradiation. This paper describes the main factors controlling the lifetime of these materials. The viability of the currently available materials is investigated for their utilization as the core materials of Generation IV SFR systems.

2. Fuel rod cladding

2.1. Austenitic steels

Austenitic steels were selected as the first materials for the cladding as well as for the duct of first generation fast reactors. Type 304 or 316 was used. These steels were chosen based on their corrosion resistance and good thermal creep resistance. Also, they have been favored in the sense of their high-temperature mechanical strength, good fabrication technology, and abundant experience. However, they exhibited excessive swelling at doses above 50 dpa. This swelling was decreased by adding stabilizing elements, by adjusting chemical composition, and by introducing cold work [1,2].

The irradiation behaviors of the austenitic steels have been summarized [2]. Radiation-induced void swelling is a life limiting factor for these steels when used for the fuel rod cladding of fast breeder reactors. The swelling increases with the neutron fluence at a given temperature. There is an incubation period after which swelling begins, as shown in Fig. 1. After the incubation period, steady-state swelling rate is almost constant, at 1%/dpa for austenitic steels [3]. The swelling depends on the irradiation temperature, neutron flux, and applied stress [2]. As shown in Fig. 2, significant improvements have been made to reduce swelling by increasing the incubation period through adding stabilizing elements, varying chemical composition, and applying cold work [4]. Stainless steels are alloyed with Ti, B and P and cold worked by 20% for the PNC 316, 15-15Ti, and D9 austenitic steels. The chemical compositions are shown in Table 1. As the swelling increases, however, its influence on the ductility becomes significant. Fig. 3 shows that, in the swelling range above ~6%, these steels become too brittle to be handled [3,5,6].

The status of the development is summarized in Ref. [5]. Abundant irradiation experience has been accumulated. Titanium-stabilized and cold worked steels have been used; D9 in the US, 15-15Ti in France, DIN 1.4970 in Germany. These steels have good creep strength such that MOX fuel with D9 cladding was irradiated in

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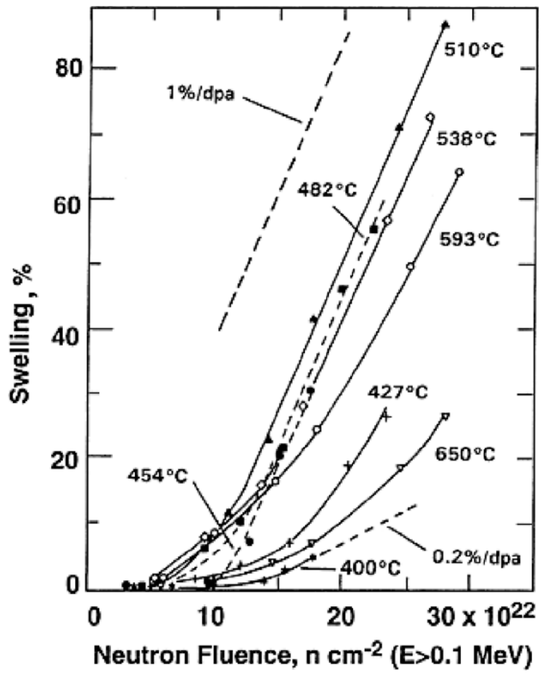

Fig. 1. Swelling as a function of the neutron fluence for Type 316 stainless steel [3].

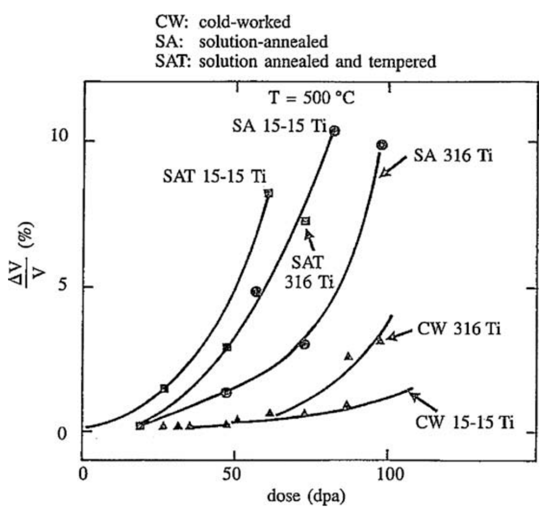

Fig. 2. Improvement of the swelling characteristics for austenitic stainless steels by varying the material conditions [4].

Table 1
Chemical composition of the austenitic stainless steels for a SFR.

	C	Cr	Ni	Mo	Si	Mn	Ti	Nb	P	B, ppm
15-15Ti	0.1	15	15	1.2	0.6	1.5	0.4		0.03	50
DIN1.4970	0.1	15	15	1.2	0.4	1.5	0.5			50
D9	0.05	14	15	1.5	0.9	1.7	0.23			50
PNC 316	0.05	16	14	2.5	0.8	1.7	0.1	0.1	0.025	40
PNC1520	0.05	15	20	2.5	0.8	1.7	0.25	0.1	0.025	40

the FFTF at a peak cladding temperature of 675 °C. However, use of these alloys is limited by the degree of swelling. Although the maximum dose attained without a failure was well above 130 dpa for an experimental sub-assembly, these steels have insufficient ductility above ~100 dpa. An optimized version of 15-15Ti steels, AIM1 (Austenitic Improved Material number one) has been

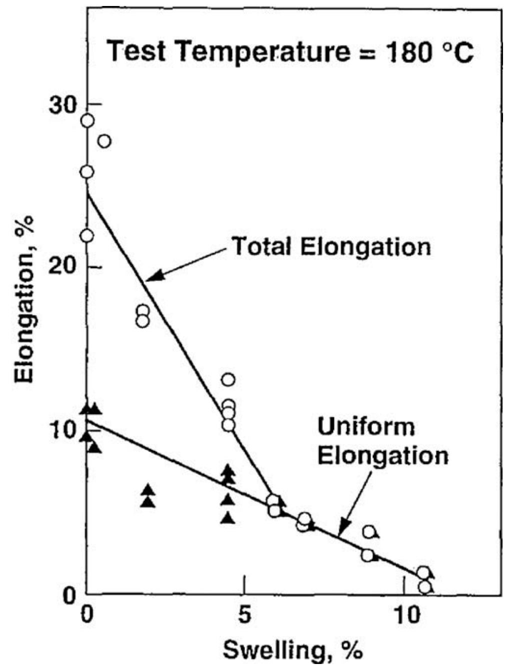

Fig. 3. Variation of the elongation as a function of the swelling for 20% cold worked Ti stabilized Type 316 steel irradiated in PHENIX [6].

developed within the framework of European Collaboration. Its use is probably limited to about 130 dpa, although several fuel assemblies are still being irradiated in Phénix [7]. Also, advanced austenitic cladding material 12-15Cr/20-25Ni was proposed to reduce the swelling in French and Japanese reactors. The swelling resistance of these alloys is much better than PNC 316 and 15-15Ti steels, respectively [5,8]. Fig. 4 shows the dose dependence of swelling in cold-worked austenitic steels irradiated at 405 °C in FFTF. These advanced austenitic steels can be used up to around 160 dpa [7,9,10].

2.2. Ferritic/martensitic steels

Ferritic/martensitic (F/M) steels are considered primary candidates for SFR cladding and duct materials of several Gen-IV

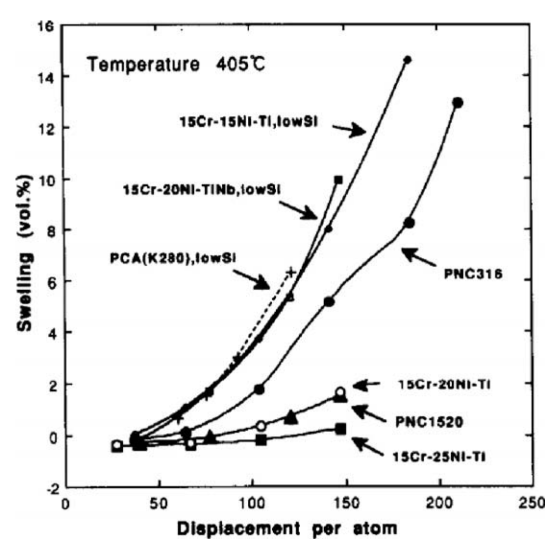

Fig. 4. Dose dependence of the swelling in 16–20% cold-worked austenitic steels irradiated at 405 °C in FFTF/MOTA [8].

SFR designs. They have high thermal conductivity and low thermal expansion. The US fast reactor program adopted HT9. Similar types of steel have been chosen in Europe and Japan (EM12, DIN 1.4914, and PNC-FMS). The chemical composition of the F/M steels for the SFR applications are shown in Table 2. All these steels have shown excellent swelling resistance up to 200 dpa with a steady-state swelling rate of ~0.2%/dpa [3]. Fig. 5 demonstrates the excellent swelling behavior of the F/M steels compared with the austenitic steels. However, most of these steels have low creep strength at temperatures above 650 °C, the temperature desired in the design of GEN-IV SFR systems. In addition, fabrication and joining are remaining issues due to the formability and weldability required when fuel rods are manufactured with F/M steels.

Characteristics of HT9 steel are summarized in Ref. [11] including their irradiation experience in the EBR-II and FFTF reactors. This steel is used with a tempered martensite microstructure produced by a normalized and tempered heat treatment. Strengthening mechanisms include solid-solution strengthening and the interaction of dislocations with microstructural features. At the operating temperature of cladding, thermal creep is dominant and irradiation creep is of secondary importance. Below 570 °C, the creep deformation of HT9 was less than for CW316 stainless steel under neutron irradiation. Above 600 °C, the dislocation density is reduced and the $M_{23}C_6$ precipitates coarsened, leading to a lower creep strength and large creep strain. Irradiation can accelerate coarsening, causing enhanced creep deformation as shown in Fig. 6 [12]. Moreover, creep deformation is influenced by swelling at higher doses [11]. In addition, heat-to-heat variation in creep and swelling behavior must be expected, and is one of the issues to be addressed.

The status of development of the F/M steels is summarized in Ref. [5]. EM 12 has been tested in the PHENIX reactor where two subassemblies reached a maximum exposure of 120–130 dpa at a moderate peak cladding temperature (600–630 °C max). In the former USSR, EP 450 alloy (13%Cr–2Mo–Nb–P–B–V) was also used as duct material, and has been successfully irradiated in a BOR 60 demountable subassembly up to about 180 dpa (June 1996) at an initial peak cladding temperature of 680 °C. For HT9 alloy irradiated in EBR-II and FFTF, the highest exposure doses were achieved with FFTF oxide fuels at a limited peak cladding temperature (600 °C), with a record level of about 200 dpa. Furthermore, some of the lead tests were performed at cladding temperatures in the range of 640–660 °C [13]. Post irradiation results confirmed the inherent characteristics of this type of material: very good resistance to void swelling, very small diameter changes (0.5% at 120–130 dpa) except for, in some pins, a peak cladding deformation (up to 1.7% at 120–130 dpa) towards the top of the fuel column, associated with a brittle lanthanide-rich layer at an inner cladding surface and a reduced creep strength at high temperatures in this upper part of the pins. This limited high temperature strength implies stringent design limits and raises some concerns with regard to pin failure behavior, especially in the context of the advanced commercial reactor design conditions (peak cladding temperature of about 650 °C).

F/M steels have been substantially developed for conventional power plants, especially for improvements in their creep resistance and oxidation at higher temperatures [14,15]. For example, a new grade of 9–12% Cr martensitic steel equivalent to T92 has long-time stability and creep properties much better than HT9. These new steels have creep stress rupture strengths similar to austenitic stainless steels. HT9 contains 0.2% C which is a strong austenite stabilizer. The main precipitate, $M_{23}C_6$, is prone to coarsen at high temperatures. Due to δ ferrite formation, 9% Cr steels with lower carbon content were favored over 12% Cr steel. In T91, a remarkable increase in creep strength was achieved by lowering the C content and adding Nb and N to promote fine MX precipitates. V

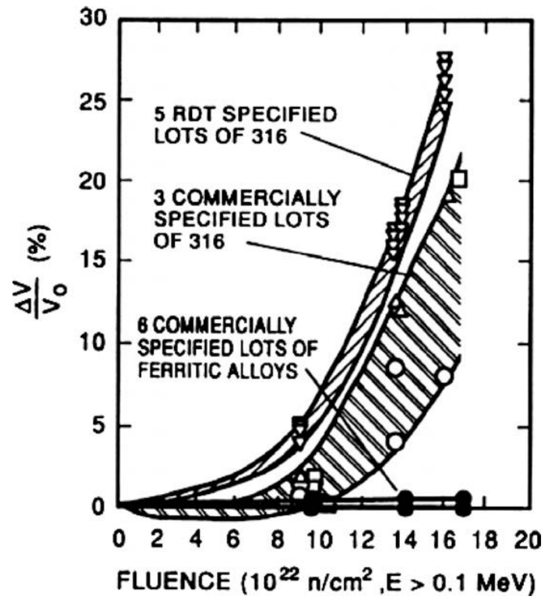

Fig. 5. Swelling behavior of commercial stainless steels at 420 °C (D.S. Gelles, unpublished research in [15]).

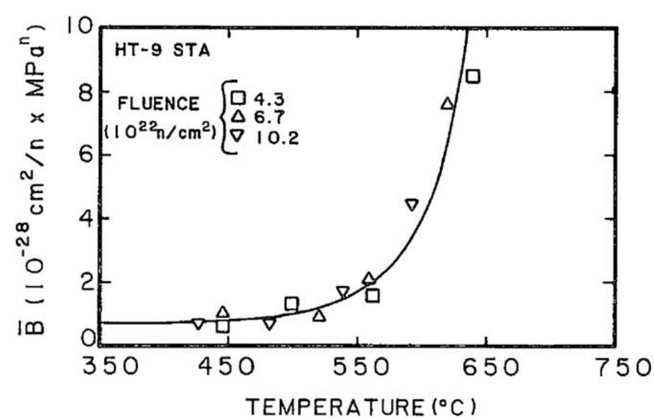

Fig. 6. Temperature dependence of the in-reactor creep coefficient for the HT9 cladding [12]. The creep coefficient, $\overline{B}$ is defined by the equation $\overline{\varepsilon} = \overline{B}\overline{\sigma}^n \phi t$ where $\overline{\varepsilon}$ is the effective strain, $\overline{\sigma}$ is the effective stress, n is the stress exponent, ϕ is the flux, and t is the time.

Table 2
Chemical composition of the ferritic-martensitic stainless steels for a SFR.

	C	Cr	Ni	Mo	W	V	Nb	Si	Mn	N	B(ppm)
EM12	0.10	9.0	0.30	2.0		0.40	0.50	0.40	1.00		
DIN1.4914	0.14	11.3	0.70	0.50		0.30	0.25	0.45	0.35	0.029	70
HT9	0.20	12.0	0.6	1.0	0.5	0.3		0.38	0.60		
PNC-FMS	0.15	11.0	0.5	0.5	2	0.2	0.05	0.05	0.5	0.05	
T91	0.10	9.0		1.0		0.2	0.08	0.4	0.4	0.05	
T92	0.07	9.0		0.5	1.8	0.2	0.05	0.06	0.45	0.06	40

and Nb have been found to exhibit optimal contents, about 0.2% and 0.05%, respectively [17]. Further improvement was accomplished by partly substituting W for Mo and adding B. This is designated as T92. Mo equivalent (Mo + 0.5 W) of 1.5% is the most effective for creep strengthening. Boron is effective in stabilizing the microstructure by being incorporated in $M_{23}C_6$. To utilize T92 for nuclear applications, ^{11}B needs to be used instead of natural boron, to avoid the ~20% ^{10}B which undergoes an (n,α) reaction [15]. Fig. 7 shows comparison of the creep rupture strength of these steels. T91 and T92 have creep strength much better than 12Cr1MoV steel [16]. Moreover, the difference between T91 and T92 is clear with increasing time. New next generation F/M steels for the conventional power-generation industry are still in their developmental stage. These contain cobalt to suppress the adverse effect of nickel on creep. However, Co-containing steels are not an option for cladding and duct materials. Considering that HT9 in the fast reactor program had adequate performance, new steels with improved creep properties will have an advantage over HT9 at around 650 °C. Also it is expected that these steels will have irradiation-resistance equivalent to HT9 so that swelling at these temperatures should not be a factor [14].

Japanese PNC-FMS is one of the steels that was developed by reflecting these advances relative to HT9. Table 2 shows that the chemical composition of PNC-FMS is similar to T91 and T92. It exhibits high temperature strength superior to conventional high Cr steels. Creep rupture strength of this steel at 650 °C is the highest among similar F/M steels as shown in Fig. 8 [18]. Also it has been shown that the creep rupture strength of the PNC-FMS cladding is not degraded by the irradiation environment [19]. Irradiation test has been performed in JOYO (present maximum exposure: 150 dpa) [11]. However, due to rapid coarsening of the carbides and nitrides at higher temperatures, it is required to limit application to 650 °C [20].

2.3. Oxide dispersion strengthened steels

To extend the use range of F/M steels to temperatures well above 650 °C, oxide dispersion strengthened (ODS) steels are under development in several programs. Oxide dispersion strengthening enables the use of cladding in this temperature range. ODS steels have been considered in the planning for Generation IV reactors, and a performance database needs to be established for them. In addition, most challenging issues that should be addressed are the fabrication of fuel cladding through the powder metallurgical process, and the joining of cladding to end plugs.

The 14% Cr ODS alloy MA957, manufactured by INCO, has been irradiated in FFTF [21,24]. Pressurized tubes were irradiated up to ~115 dpa in a temperature range of 400–600 °C. Creep behavior of

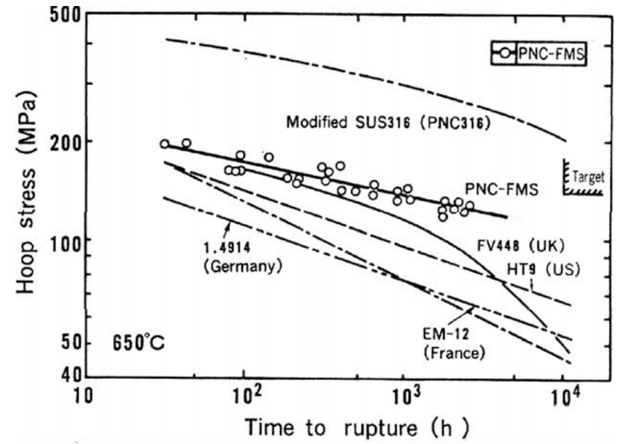

Fig. 8. Creep rupture strength for the cladding steels for a SFR [18].

MA957 was comparable to HT9 in a lower temperature range. In Fig. 9, at 600 °C the creep rate of MA957 is about one-half the value for HT9, and the magnitude of the creep transient for MA957 is much lower than for HT9 [21]. Creep-rupture behavior was compared using the Larson–Miller Parameter [24], and the creep-rupture properties of MA957 are superior to T92, as shown in Fig. 10.

JAEA since 1987 has developed two types of ODS steel for use as cladding. One is a martensitic 9Cr-ODS steel with chemical composition Fe–0.13C–9Cr–2 W–0.2Ti–0.35Y_2O_3. The other is a ferritic 12Cr-ODS steel with chemical composition Fe–0.03C–12Cr–2W–0.3Ti–0.23Y_2O_3 [11,22]. The 9Cr-ODS steel was developed to improve the radiation resistance and the ferritic 12Cr-ODS steel focused on corrosion resistance. The cold rolling process to manufacture ODS cladding causes extensive grain growth along the rolling direction, resulting in a strength anisotropy. Techniques to control the grain structure were devised to solve this problem; it included α to γ phase transformation for 9Cr-ODS steel and recrystallization processing for 12Cr-ODS steel. These steels showed improved tensile strength and uniform elongation. In Fig. 11, the creep rupture strength of these ODS steels at 700 °C is compared to PNC-FMS and PNC316 [23]. These curves were predicted by the Larson-Miller parameter method. Creep rupture strength of both 9Cr-ODS and 12Cr-ODS steels meet the target of 120 MPa for 10,000 hr at 700 °C [23]. This strength is well above that of PNC-FMS, and better than that of PNC316 beyond 1,000 hr at 750 °C. ODS fuel pin irradiation tests have been conducted in BOR 60 of RIAR in Russia [32]. The irradiation tests are planned to reach a target burnup of 15 at % and a dose of 75 dpa. Also, ODS fuel pins are scheduled to be irradiated in JOYO. These

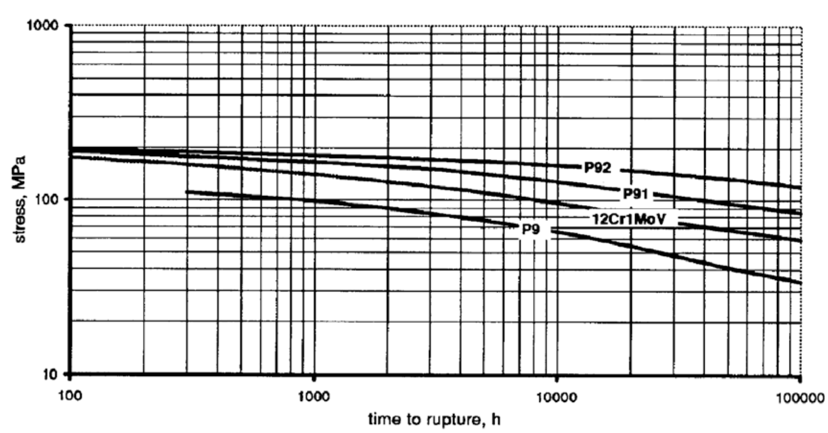

Fig. 7. Comparison of the creep rupture strength for 12Cr–1MoVW (HT9), P91, and P92 at 600 °C [16]. Steels P91 and P92 are different designations for T91 and T92.

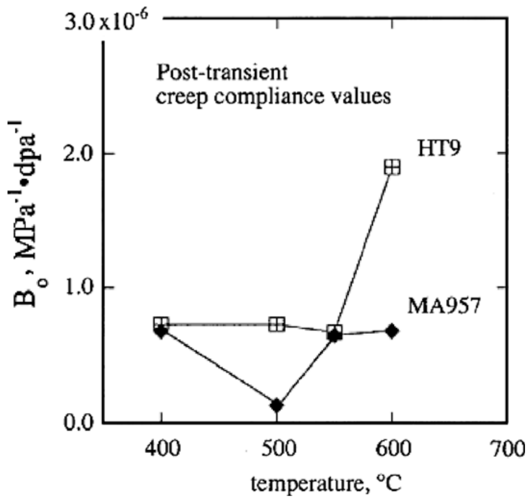

Fig. 9. Temperature dependency of the creep compliance for MA957 and HT9 during irradiation [21]. The creep compliance, B_0 is defined by the equation $\dot{\bar{\varepsilon}}/\bar{\sigma} = B_0 + D\dot{S}$ where $\dot{\bar{\varepsilon}}$ is the effective strain rate per dpa, $\bar{\sigma}$ is the effective stress, D is the creep-swelling coupling coefficient, and $\dot{S}$ is the volumetric swelling rate per dpa.

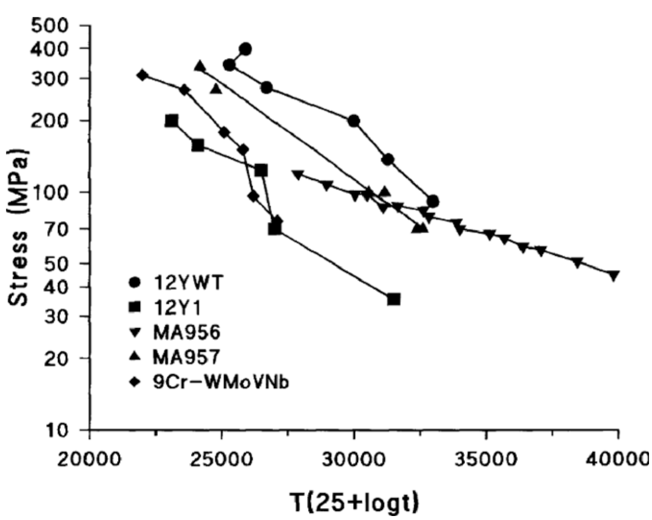

Fig. 10. Creep rupture stress vs. Larson–Miller parameter for MA957 and T92 (9Cr–WMoVNb) [24].

irradiation test data will be applied to the licensing of ODS driver fuels in the MONJU up-grade core.

2.4. Cladding performance in GEN-IV SFR systems

The design of a GEN-IV SFR system demands an advanced cladding capable of high temperature-high burnup operation. Specifically, a maximum cladding temperature higher than 650 °C is desirable, which will allow for a higher core outlet temperature to achieve higher thermal efficiency. To achieve a higher average discharge burnup, it is also necessary to develop claddings with good swelling resistance to more than 200 dpa.

Austenitic steels have excellent material properties at high temperature and acceptable swelling capability up to ~160 dpa, satisfying the requirements for the cladding of the current SFR systems. To achieve a higher burnup in the GEN-IV SFR system, steels with superior swelling-resistance need to be used. To this end, the creep strength of F/M steels should be improved, and an ODS steel with a high temperature strength needs to be qualified.

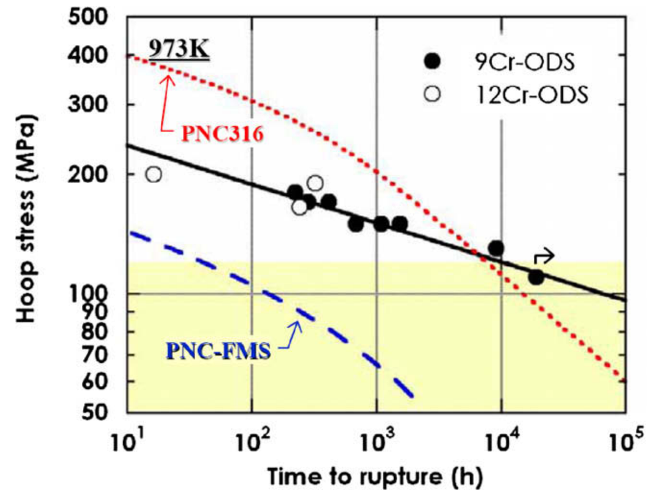

Fig. 11. Creep rupture strength of the ODS steels compared with those of PNC-FMS and PNC316 [23].

Table 3 shows the main parameters related to the cladding performance for the SFRs under development by JSFR, KAERI-SFR and ABR. Metal and/or oxide fuels are likely be employed as a result of their technical maturity relative to other fuel types. As for the cladding materials, the ODS steel or the advanced F/M steel are primary candidates. Maximum temperature determines which type of cladding is selected; the ODS steel up to 700 °C, and the F/M steel limited to around 650 °C. Peak dose is designed to be more than 200 dpa, and could reach 250 dpa.

Cladding temperature is regarded as the dominant factor among those parameters. Although more effort is required to clarify their material stability and creep behavior during long service times, the F/M steels for the conventional, non-nuclear plants have been substantially improved relative to those used in the current SFR operations. Moreover, the life time of fuel pin is 50,000 hours which is much shorter than 300,000 hours for the conventional plants. In this regard, for the Generation IV SFR cladding materials, it is evident that an improved F/M steel can be used by restricting the peak temperature to around 650 °C, and the ODS steel has no limitation for its utilization up to 700 °C. In both steels, swelling would not be a limiting factor relative to creep deformation for the cladding integrity as both have a common swelling-resistant structure.

While the manufacturing technologies for the F/M steel cladding are well established, heat-to-heat variation in the material properties and uncertainty on the irradiation behavior still exists. Quality assurance system should be developed to ensure more uniform material properties. Compared to F/M steels, further research on ODS steels should be devoted to their technology maturity in fields such as an optimization of their production technology in terms of cost and production efficiency, and joining technology [25]. More irradiation tests for F/M steels and ODS steels are inevitable to establish the Generation IV performance requirements.

High cladding temperature may worsen the fuel-cladding chemical interaction (FCCI), especially at the high burnup needed for TRU-bearing fuels. For oxide fuels, cladding corrosion due to

Table 3
Main parameters related to the cladding performance for the SFRs under development.

	JSFR	KAERI-SFR	SMFR (ABR)
Fuel (reference)	MOX	Metal	Metal / MOX
Cladding (reference)	ODS	FMS	FMS/ODS
Cladding temperature limit, °C	700	650	650~700
Peak dose, dpa	250	250	200

FCCI is accelerated for a high O/M ratio (>1.98) at high temperature and high burnup [1]. The corrosion rate must be measured for the range of new F/M steel claddings. Understanding of the FCCI at high burnup and in the presence of TRU is essential. In the case of metallic fuels, the temperature limit at their inner cladding surface is determined by the eutectic reaction among U, Pu, and Fe; it is 650 °C for U-26Pu-10Zr with HT9 [26]. This means that there is a need to broaden the understanding of the eutectic temperature and the reaction kinetics for the TRU-bearing metallic fuel pins manufactured with improved cladding materials. Also, a lanthanide and cladding constituent interdiffusion has caused the formation of a brittle layer [13]. As the FCCI occurs for F/M steels and ODS steel, the development of a coating technology for the inner surface of cladding [27], or the introduction of a barrier cladding [28] would possibly overcome such limitations.

Compatibility of cladding with sodium coolant has been shown to be excellent by maintaining oxygen levels below 10 ppm. Transfer of carbon through the sodium loop may result in carburization or decarburization. There is very little data on the compatibility with sodium for the advanced F/M steel or ODS. Also, advanced steels with lower chromium content might exhibit lower strength and corrosion resistance. It is required to identify these concerns with extensive experiments.

3. Duct materials

Ducts are used in SFRs to contain fuel pins and provide a flow channel for sodium. To meet these functions, it must have the proper characteristics. The first generation of ducts was manufactured with austenitic steels [5]. Although these steels have been progressively improved, these ducts exhibited low resistance to irradiation swelling at the higher dose required of cladding. In later cores of these reactors, F/M steels with 9–12% Cr have been used for the duct material [5]. These steels are EM10, PNC-FMS, and HT9 which are the same designations as the cladding materials. However, different heat-treatment conditions were adopted for the cladding and the duct. These were chosen because fuel cladding requires high creep rupture strength while a duct needs greater tensile strength and fracture toughness. For instance, PNC-FMS was normalized at 1100 °C for 10 min followed by tempering at 780 °C for 1 h for cladding, and 1050 °C for 10 min and 700 °C for 1 h for ducts, respectively [19]. The effect of irradiation on the toughness can be affected by the normalizing-and-tempering treatment and by the processing used for the steel during manufacturing. Increasing the austenitizing temperature generally increases the prior-austenite grain size, resulting in an increase in the ductile-brittle transition temperature (DBTT). The toughness is degraded by the addition of minor elements such as S, P, and Si. Unlike fusion applications, transmutation helium in F/M steels does not seem to be an issue for the SFR ducts.

The effect of irradiation on the tensile behavior of F/M steels shows that the yield strength increases and the ductility decreases [11,14]. Such trends saturate with increasing fluence. For irradiation above 425–450 °C, these properties are generally unchanged after some amount of irradiation, although there may be some softening, depending on the fluence and temperature. The effect of irradiation hardening on the toughness is shown as an increase in the DBTT and a decrease in the upper-shelf energy. Laves phase, which forms during thermal aging and for irradiation at 400–600 °C, can also cause embrittlement. This phase does not form if the irradiation is above ~600 °C. Instead $M_{23}C_6$ and MX coarsen during elevated-temperature irradiation.

HT9 has an irradiation record up to 200 dpa in FFTF [5,29]. The irradiation results show that the dimensional stability is very good. The maximum swelling rate is less than 2% at all temperatures. At temperatures around 400 °C and up to approximately 450–470 °C, these steels are susceptible to brittle failure due to irradiation hardening. Mechanical tests show that irradiation hardening occurs mainly at temperatures below 500 °C, and that this effect saturated at a low dose. Some hardening is still observed at higher temperatures, but it is reduced in magnitude and embrittlement is not observed. The mechanical behaviors is quite similar among various F/M steels. The uniform elongation remains around 1%. DBTT shift due to irradiation does not depend significantly on dose. The DBTT shifts are relatively small, so these steels can be employed as duct materials.

During the development of advanced F/M steels, their mechanical properties were improved along with an increase in the creep

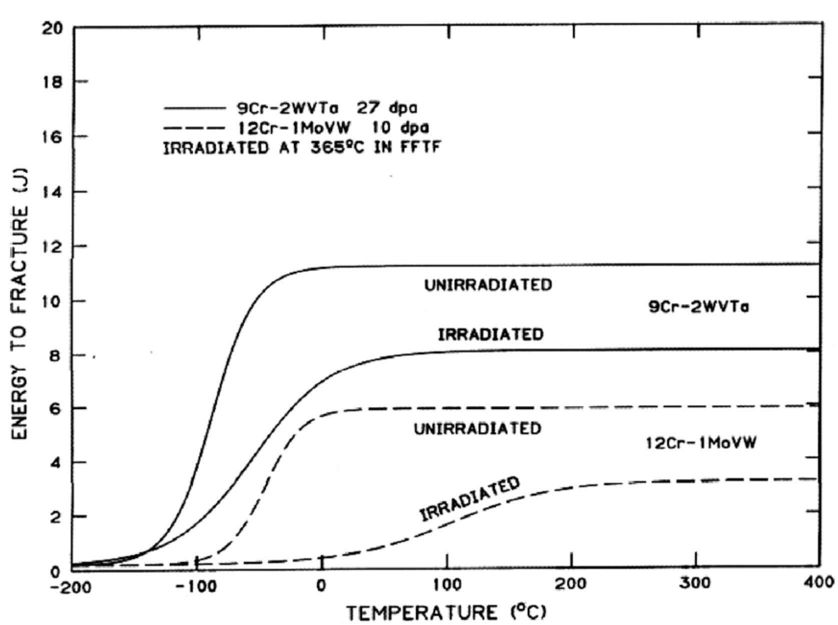

Fig. 12. Comparison of the unirradiated and irradiated Charpy curves for one-third-size specimens of 12Cr–1MoVW (HT9) and 9Cr–2WVTa steels irradiated in FFTF at 365 °C [31].

strength [14,15]. Studies of the embrittlement of HT9 and modified 9Cr–1Mo indicated that, for similar strengths, the shift in DBTT of the latter steel (54 °C) was about half that of the former (124 °C) after irradiation in EBR-II and FFTF at 375–390 °C. The difference was attributed to the larger amount of carbide in the HT9, which contains twice as much carbon as the modified 9Cr–Mo [30]. It was demonstrated that a further increase can be accomplished in the resistance to irradiation embrittlement; the reduced-activation steel 9Cr-2WVTa developed by ORNL exhibits a DBTT that is at least 25 °C less than that for T91 in the unirradiated condition. Fig. 12 shows that irradiation of this steel results in an even smaller increase in DBTT than HT9 at these temperatures [31]. Also lower Cr content in the 9Cr steels results in the elimination of δ ferrite, which can be a factor in their high toughness relative to HT9.

4. Conclusion

The status of available and developmental materials for SFR core cladding and duct applications was reviewed. To satisfy the Generation IV SFR fuel requirements such as higher temperature and higher burnup, an advanced cladding needs to be developed, based on the accumulated experience with these materials. The candidate cladding materials are austenitic steels, F/M steels, and ODS steels. A large amount of irradiation testing is required, and the compatibility of cladding with TRU-loaded fuel at high temperatures and high burnup must be understood. The more promising F/M steels (compared to HT9) might be able to meet the dose requirements of over 200 dpa for ducts in the GEN-IV SFR systems.

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