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IWSMT-9

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Mechanical properties and microstructural stability of 11Cr-ferritic/martensitic steel cladding under irradiation

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The in-reactor creep rupture tests of 11Cr-0.5Mo-2W, V, Nb F/M steel were carried out in the temperature range from 823 to 943 K using Materials Open Test Assembly in the Fast Flux Test Facility and tensile and temperature-transient-to-burst specimens were irradiated in the experimental fast reactor JOYO at temperatures between 693 to 1013 K to fast neutron doses ranging from 11 to 102 dpa. The results of post irradiation mechanical tests showed that there was no significant degradation in tensile and transient burst strengths even after neutron irradiation below 873 K, but that there was significant degradation in both strengths at neutron irradiation above 903 K. On the other hand, the in-reactor creep rupture times were equal or greater than those of out-reactor creep even after neutron irradiation at all temperatures. This creep rupture behavior was different from that of tensile and transient burst specimens.

Keywords:

Fast Reactor Materials, Mechanical Properties, Neutron Irradiation, Ferritic/Martensitic steels

1. Introduction

One of the world's most pressing problems is to reduce carbon dioxide and greenhouse gas emissions; in this regard, nuclear energy systems are receiving new attention as energy sources because large amounts of power can be produced by nuclear reactors without the adverse environmental effects that accompany the use of coal or oil products. As an energy strategy, fast reactor (FR) cycle systems as a major energy supply for the future have been studied for their economic competitiveness towards light water reactor cycle systems and other electric power supplies.

High swelling resistance and stable mechanical properties at elevated temperature under fast neutron irradiation conditions are very important issues for development of long-life FR core materials. Austenitic steels such as 304 and 316 stainless steels have been used as the first generation FR core materials, and then advanced austenitic steels which are superior to swelling characteristics have been developed and used; but in Generation IV fission reactors [1], 9-12 % chromium ferritic/martensitic (F/M) steels such as HT-9 are being considered as sodium FR core materials because of their excellent void swelling resistance although their high temperature strength is inferior to that of austenitic steels.

The most primary concern in designing FR plants is to avoid failure of fuel pin claddings during plant operation. Therefore, in-reactor creep rupture strength data are required in order to establish design criteria. There are, however, few public reports of in-reactor creep rupture properties [2-4] although there are many published reports [5-12] on creep strain properties. This is because it is very difficult to detect creep rupture time during actual FR operation. The above studies on creep rupture behavior were concerned with austenitic steels [2-4], but there are no reports on in-reactor creep rupture lifetimes of F/M steels, even though neutron-irradiated data on F/M steels is required for design purposes.

Based on this background, an experimental program was conducted to determine the in-reactor creep rupture properties of 11Cr-0.5Mo-2W, V, Nb F/M steel (PNC-FMS) in the temperature range from 823 to 943 K using the Materials Open Test Assembly (MOTA) in the Fast Flux Test Facility (FFTF). The tensile and temperature-transient-to-burst tests were also carried out for PNC-FMS claddings irradiated in the experimental FR JOYO at temperatures between 693 to 1013 K to fast neutron doses ranging from 11 to 102 dpa. This paper describes the evaluation of the effects of neutron irradiation on mechanical properties for PNC-FMS claddings.

2. Experimental procedures

2.1. Specimens

The PNC-FMS claddings studied here are one of the F/M steels developed for application as a core material for advanced FRs. Specimen lots of 61FS, 61FSF and 1FK were prepared. Their chemical compositions and heat treatment conditions are listed in Table 1. The outer and inner diameters of both lots 61FS and 61FSF claddings were 6.5 and 5.56 mm, respectively. And those of lot 1FK were 6.86 and 5.74 mm, respectively. Development details of the PNC-FMS claddings were reported elsewhere [13-14].

2.2. Thermal and in-reactor creep rupture tests

The creep rupture tests were performed in air atmosphere in order to obtain the fundamental characteristics of PNC-FMS claddings. In these tests, 200 mm long cladding segments were filled with argon gas and sealed to keep a constant internal pressure inside them. The in-air creep rupture test conditions are shown in Table 2.

Pressurized cladding creep specimens were used to obtain in-reactor creep rupture data in the MOTA. End caps were welded by an electron-beam technique to the cladding bodies; this work was done by Westinghouse Hanford Company. The specimens were then filled with the desired pressure of helium gas to generate the desired hoop stresses in them at the irradiation temperatures. The specimens were sealed by laser welding of the fill hole located in the specimen end caps. Small amounts of unique isotopic mixtures of krypton and xenon gases were included in the helium to allow identification of the individual ruptured specimens and their rupture times by analyzing the released gas species using the FFTF reactor cover gas monitoring system during operation [3].

These specimens were irradiated in the FFTF for three reactor cycles (from the 10^{th} to the 12^{th}) using the MOTA irradiation vehicle. Irradiation and in-reactor creep rupture test conditions are shown in Table 3. The irradiation temperatures were monitored by thermocouples and controlled within an accuracy of ±5 K.

2.3. Tensile tests

Tensile tests were carried out in air using a screw-driven tensile testing machine at a strain rate of 5.0×10^{-5} /s, which was changed to 1.3×10^{-3} /s after yielding. The test temperatures were 693 to 1013 K corresponding to the irradiation temperatures. Yield strength (YS) was determined as 0.2 % offset proof stress. The tensile specimens were irradiated in JOYO at irradiation temperatures from 693 to 1013 K to neutron doses from 11 to 99 dpa.

2.4. *Temperature-transient-to-burst tests*

The cladding specimen was internally pressurized by high purity argon gas, and then it was heated by direct electric current with temperatures increasing linearly until rupture in order to study rupture strength during ramp heating. The temperature was measured by a Type R thermocouple. The heating rate was kept at 5 K/s because of the importance of this heating rate during operational transients in the prototype fast breeder reactor MONJU. The axial temperature distribution during the transient heating was confirmed to be within ±25 K in a 20 mm region around the axial center of the specimen. The hoop stress conditions, which were estimated by the thin walled tube approximation, were 49, 98, 120 and 196 MPa. The neutron irradiation of the temperature-transient-to-burst specimens was done in JOYO at irradiation temperature ranging from 693 to 1013 K to neutron doses from 19 to 102 dpa.

3 Results and Discussions

Figure 1 (a) shows the thermal control data for the as-received PNC-FMS claddings in air atmosphere as a function of a Larson-Miller Parameter (LMP) plot [2-4] which is most appropriate way to estimate trend curves of time to rupture at each test temperature. The specific formula for the LMP that was used in this work is given by

$$\mathrm{LMP} = T\,[34.3 + \log t_R] \text{ --- } (1)$$

where T is the temperature in Kelvin and t_R is the time to rupture in hours. The thermal control experimental results and creep-rupture trend curves at each test temperature that were estimated from LMP are shown in Fig. 1 (b). The creep rupture strength of lots 61FS, 61FSF and 1FK specimens are located on the same lines with only small data scatter (shown in Fig. 2 (b)), therefore each lot appears to have similar creep rupture properties.

To compare the data of pressurized tube specimens with F/M steel specimens, the effective stress was used. For a pressurized tube specimen with biaxial stress, the effective stress is given by

$$\sigma_{eff} = (\sqrt{3}/2)\sigma_h \quad \text{--} \quad (2)$$

where σ_{eff} is the effective uniaxial stress and σ_h is the hoop stress. This correlation between uniaxial and hoop stresses is based on the Von Mises theory [6, 12]. Figure 2 shows the estimated 10^5 h uniaxial rupture stresses at 823, 873 and 923 K with the data for the other F/M steels [15]. PNC-FMS creep rupture strength that converted hoop stress to uniaxial stress was superior to the creep rupture strength of other F/M steels at all test temperatures.

The in-reactor creep rupture properties of the PNC-FMS claddings are shown in Fig. 3. A previous study by Puigh and Hamilton [4] suggested that the in-reactor creep lifetimes decreased with increasing exposure time over approximately 2,000 h in austenitic stainless steels although they were nearly equal to out-reactor creep lifetimes under 2,000 h. Ukai et al. [2] also reported that in-reactor creep rupture lifetimes were shortened in 20% cold worked modified 316 stainless steel as compared with the results of out-of-reactor creep rupture tests. On the other hand, in Fig. 3, the in-reactor creep rupture times were equal or greater than those of out-reactor creep even after neutron irradiation, although the creep rupture times of PNC-FMS claddings were much longer than 2,000 h. This result suggested that there was no significant degradation in creep rupture strength comparison with out- and in-reactor creep rupture tests and that the irradiation effects on the tendency for creep rupture strength of F/M steels were different from those of austenitic steels.

Figures 4 (a) and (b) show the YS and ultimate tensile strength (UTS) for PNC-FMS claddings before and after irradiation as a function of neutron dose, respectively. The relationship between neutron dose and failure temperature (transient burst strength) is shown in Fig. 5. At the irradiation temperature of 693 K, irradiation caused an increase in both YS and UTS in Fig 4. A significant degradation in tensile strength could not be clearly observed at irradiation temperatures between 763 and 873 K [16-17] and thus no irradiation effects on tensile strengths were recognized in this irradiation temperature range. On the other hand, at irradiation temperatures over 903 K, irradiation caused a decrease in both YS and UTS [17]. In Fig. 5, essentially no change in transient burst strength occurred at irradiation temperatures below 853 K [16-17] and there were no irradiation dependent effects in this irradiation temperature range. On the other hand, at irradiation temperatures over 943 K, there was significant degradation in transient burst strength due to the effects of neutron irradiation and/or thermal aging [17].

The microstructural observation of PNC-FMS cladding lot 61FS after neutron irradiation in a previous work by Yamashita et al. [18] showed the following results. When the specimens were irradiated at low irradiation temperatures between 673 and 723 K to doses up to 94.6 dpa, both dislocation loops and cavities (less than 30 nm in diameter) were formed in the region of the ferrite phase and estimated void swelling was 0.05% at the most. On the other hand, it was observed that the precipitates of $M_{23}C_6$ and M_6C grew and entirely covered the prior austenitic grain boundaries and also lath martensite structures were slightly recovered during neutron irradiation at moderate irradiation temperatures between 773 and 878 K to doses up to 101.5 dpa. Furthermore at high irradiation temperatures above 923 K to doses up to 103 dpa, formation and growth of equi-axial grains occurred in addition to the complete recovery of martensite laths, and there was the huge growth of carbides.

As described previously, the tensile and transient burst strengths of PNC-FMS claddings degraded due to neutron irradiation at high temperatures, and it was suggested that such significant strength degradation was related to a drastic microstructural change during irradiation, especially recovery of martensite lath structures and precipitation behavior [16-18]. However it was still not clarified why in-reactor creep rupture behavior was similar to out-reactor creep behavior at 943 K. Among possible explanations, it is considered that thermal aging rather than neutron irradiation would affect the degradation of in-reactor creep rupture strength, namely the contribution of point defects introduced during neutron irradiation could be negligible because of their rapid annihilation at high temperatures. It is also considered that there is difference of test specimen conditions with microstructural changes, namely in-reactor creep rupture tests were in-situ neutron irradiation, while post-irradiation examination tests were ex-situ, viz. microstructural observation after neutron irradiation.

It is suggested that the effects of neutron irradiation on the mechanical properties of PNC-FMS depended on the irradiation temperatures rather than neutron doses. Furthermore it is necessary to study microstructural influence for in- and out-reactor creep strengths in order to clarify in-reactor creep rupture behavior.

4. Conclusion

In-reactor creep rupture, tensile and temperature-transient-to-burst tests were conducted on PNC-FMS claddings. The results are summarized as follows.

(1) The in-reactor creep rupture times were equal or greater than those of out-reactor creep rupture times even after neutron irradiation below 943 K.

(2) The results of post irradiation mechanical tests showed that there was no significant degradation in tensile and transient burst strengths even after neutron irradiation below 873 K, but that there was significant degradation in both strengths at neutron irradiation above 903 K.

(3) The effects of neutron irradiation on the mechanical properties of PNC-FMS depended on the irradiation temperatures rather than neutron doses.

Acknowledgements

Execution of this experimental program required the active cooperation of many people in JAEA (successor to JNC) and the US, and we wish to gratefully acknowledge their support and encouragement. We are pleased to thank the staff of Pacific Northwest National Laboratory (former Westinghouse Hanford Company) for their cooperation in the irradiation test at FFTF. We also wish to thank Dr. T.

Yoshitake, Mr. S. Mizuta, Mr. T. Endo, Mr. T. Inoue, Mr. Y. Shigeto, Mr. S. Sato and Mr. M. Sekine of JAEA for specimen preparation and testing.

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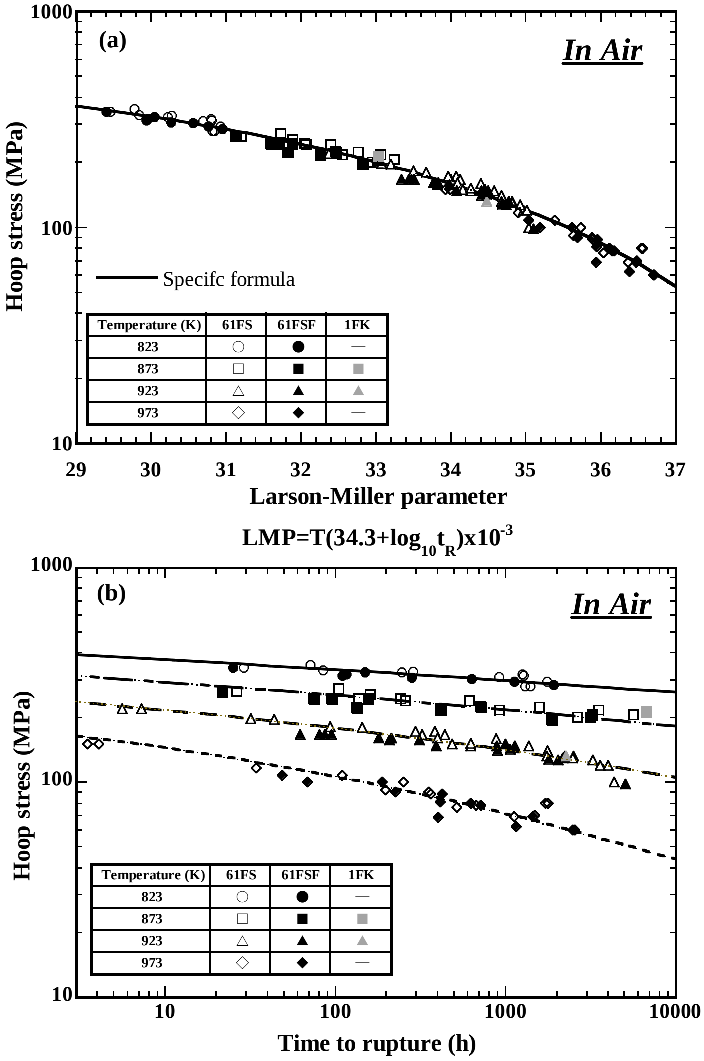

Figure 1 Summary of the thermal stress rupture behavior of as-received PNC-FMS cladding. (a) The rupture time t_R is in hours and the temperature T is in Kelvin in the Larson-Miller parameter. (b) Relationship between hoop stress and time to rupture at the temperature from 823 to 973 K. The trend curve indicates the fitting from LMP.

one- column figure

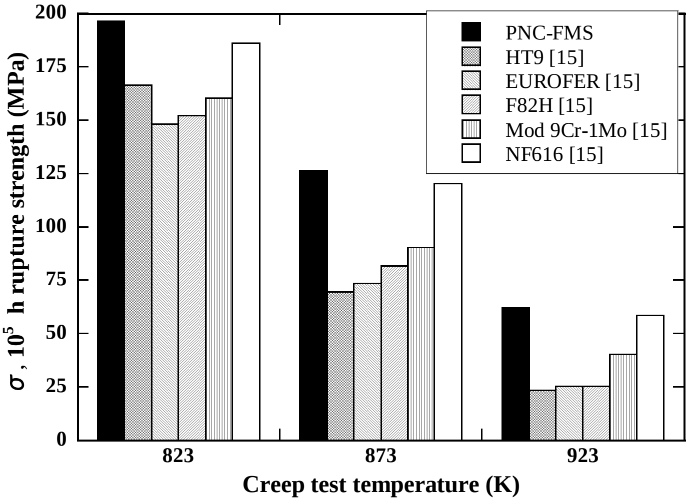

Figure 2

Comparison of the 10^5 h rupture strengths for PNC-FMS, Sandvik HT9, EUROFER, F82H, modified 9Cr-1Mo and NF616 at 823, 873 and 923 K.

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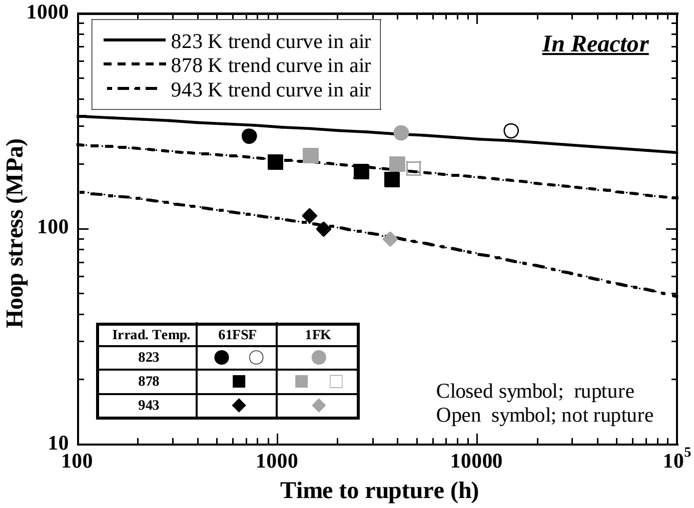

Figure 3 Results of in-reactor creep rupture tests of PNC-FMS at temperatures of 823, 878 and 943K.

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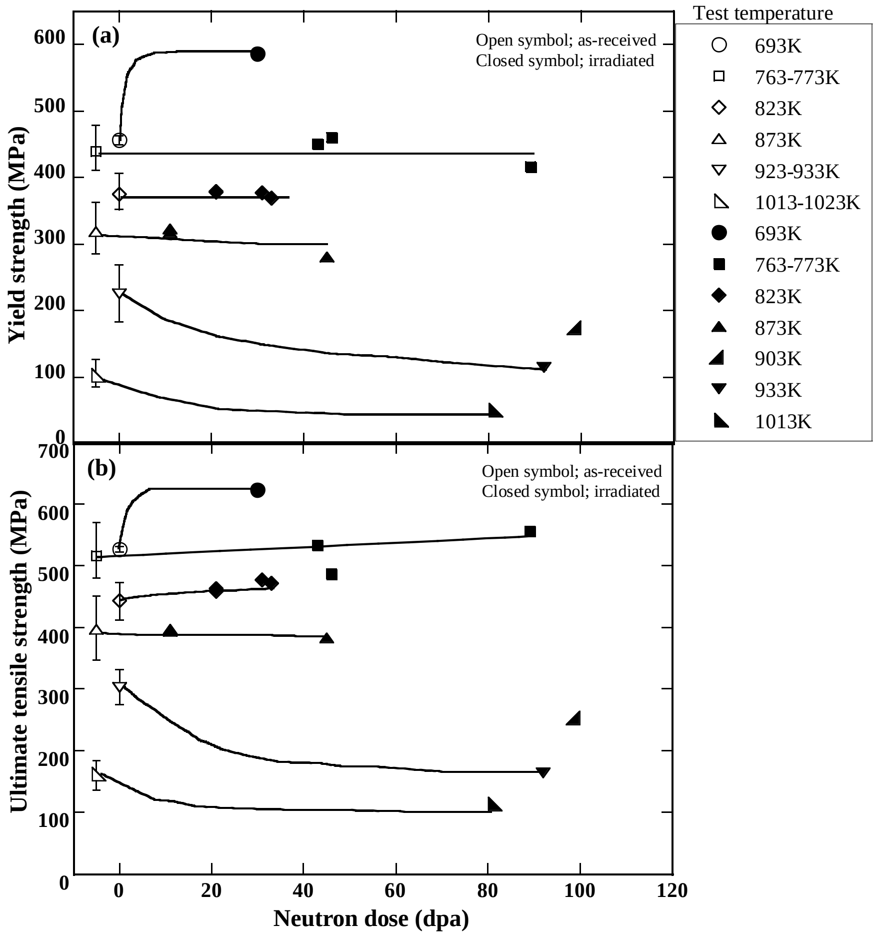

Figure 4 Relationship between tensile strength and neutron dose of irradiated PNC-FMS claddings: (a) yield strength and (b) ultimate tensile strength

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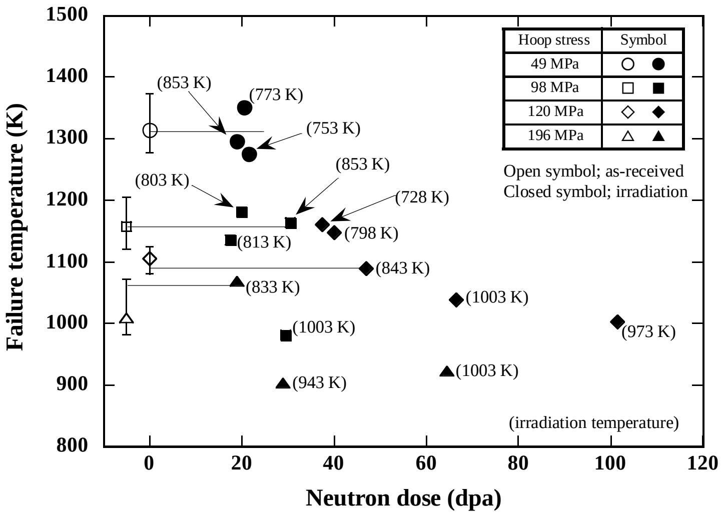

Figure 5

Relationship between transient burst strength and neutron dose of irradiated PNC-FMS claddings. Error bars with maximum and minimum data points are for as-received specimens.

one- column figure

Table 1 Chemical compositions and heat treatment conditions of PNC-FMS claddings (wt%)

Lot	C	Si	Mn	P	S	Ni	Cr	Mo	W	N	Nb	V	Fe
61FS	0.10	0.07	0.54	0.002	0.002	0.32	11.1	0.45	1.89	0.04	0.06	0.21	bal.
61FSF	0.12	0.06	0.69	0.002	0.002	0.82	11.0	0.09	2.11	0.04	0.06	0.20	bal.
1FK	0.11	0.02	0.49	0.003	0.003	0.01	11.05	0.48	1.94	0.045	0.05	0.20	bal.

Heat treatment condition:
normallized at 1373 K for 10 min and then tempered at 1053 K for 1h (lot 61FS and 1FK)
normallized at 1333 K for 10 min and then tempered at 1023 K for 10h (lot 61FSF)

Table 2 In-air creep rupture tests

Material	Temperature (K)	Hoop stress (Mpa)	Time to rupture (h)	Number of hoop stress level
PNC-FMS (61FS, 61FSF and 1FK)	823	280-353	25-1749	11
	873	196-272	21.6-6699.5	10
	923	98-220	5.6-5098	16
	973	60-150	3.5-2561	10

Table 3 In-reactor creep rupture tests

Material	Irrad. Temp. (K)	Hoop stress (MPa)	Time to rupture (h)	Neutron dose (dpa)	Number of hoop stress level	Number of not ruptured specimen
PNC-FMS (61FSF and 1FK)	823	270-285	720-4164	3.5-74.7	3	1
	878	170-285	974.9-3982	4.9-23.5	6	1
	923	90-115	1453.7-3657	7.4-19.3	3	0

two- column figures