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Journal of Nuclear Materials 278 (2000) 320–327

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

In-reactor creep rupture properties of 20% CW modified 316 stainless steel

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Received 24 May 1999; accepted 1 September 1999

Abstract

The in-reactor creep rupture tests of 20% cold worked modified 316 stainless steel were conducted in the temperature range from 878 to 1023 K using MOTA of FFTF, and were compared with the out-of-reactor tests. In-reactor creep rupture, lives become shorter than those of the out-of-reactor tests. In-reactor creep strain rate was significantly accelerated, and sufficient ductility appears to be maintained even under the irradiation. Considering 0.2% proof strength after neutron irradiation, sodium exposure or aging, the degraded rupture lives of in-reactor creep are ascribed to the enhanced dislocation recovery due to the neutron irradiation as well as to the solute elements dissolution into sodium under the sodium exposure environment. © 2000 Elsevier Science B.V. All rights reserved.

PACS: 62.20.Hg

1. Introduction

In prototype fast breeder reactor Monju, 20% cold-worked modified 316 stainless steel has been used for the fuel pin cladding. This material was developed to improve the high temperature creep rupture strength and swelling resistance far beyond the standard type 316 stainless steel by controlling the minor elements such as phosphorus, boron, titanium and niobium within a range of 0.1 mass%. The typical composition is Fe–16Cr–14Ni–0.05C–2.5Mo–0.7Si–0.025P–0.004B–0.1Ti–0.1Nb and 20% cold-working is added, which is designated PNC316 [1,2]. Out-of-reactor tests of PNC316 including creep rupture testing as well as fuel assembly irradiation test of Monju fuel in Fast Flux Test Facility (FFTF) have been conducted by Japan Nuclear Cycle Development Institute [2] [3].

A primary concern of designers is to avoid the fuel pin failure during service, and creep rupture characterization has proven to be a useful criterion for a design basis. It was found, however, that in-reactor creep rupture strength is different from the results of out-of-reactor tests. For instance, Lovell, Gilbert and Puigh suggested that in-reactor rupture lives were equal to or greater than those of out-of-reactor for 20% cold-worked AISI 316SS [4,5]. On the other hand, it was reported that neutron irradiation eventually caused the degradation of creep rupture strength [6–8].

Therefore, an experimental program was initiated to determine the in-reactor creep rupture properties of PNC316 in the temperature range from 878 to 1023 K using Materials Open Test Assembly (MOTA) [5] in FFTF. As a comparison, creep rupture tests were conducted in air atmosphere and in sodium environment.

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PII: S0022-3115(99)00233-0

The tensile tests were also carried out for PNC316 after aging, sodium and neutron exposures to evaluate effects of neutron irradiation and sodium exposure on high temperature mechanical properties. This paper describes the results obtained in this program for PNC316 cladding tubes.

2. Experimental procedure

2.1. Specimens

The creep rupture and tensile tests were conducted using two heats of PNC316; 55MK and 55MS. The chemical compositions of both heats are given in Table 1. In PNC316, minor elements of titanium, niobium, phosphorus and boron are added within the specification of Japan Industrial Standard of 316 stainless steel. The cold-working conditions of 20.0% and 19.2% after the solution treatment at around 1355 K were provided for each heat of 55MK and 55MS, respectively. The creep rupture and tesile specimens were prepared from cladding segments having the dimension of 6.5 mm diameter and 0.47 mm thickness.

2.2. In-reactor creep rupture tests

Pressurized tubes were used to provide in-reactor creep rupture data in the FFTF-MOTA experiment. The specimens cut from the cladding segments of both 55MK and 55MS heats were electron beam welded to the end caps. After gas filling with helium to the pressure required to produce the desired stress in the specimens at the intended irradiation temperature, the specimens were sealed with a laser welding of the fill hole located at the specimen top end cap. Small amount of unique isotopic mixtures of krypton and xenon gases were also filled to identify the ruptured specimens and their rupture times through analysing the released gas species by means of Cover Gas Monitoring System during FFTF operation.

The pressurized tubes 28.1 mm in length had been irradiated in the FFTF cycles fifth and sixth for some specimens and cycles nineth and tenth for another specimens using the MOTA irradiation vehicle. Irradiation conditions are given in Table 2. Three irradiation temperatures were selected; 878, 943 and 1023 K, which were monitored by thermocouples and controlled within accuracy of ±5 K during irradiation. Several levels of hoop stress were set for each temperature to obtain the ruptured time varied in the order of 100–10 000 h. Corresponding maximum fast neutron fluence is approximately 20×10^{26} n/m^2 ($E > 0.1$ MeV). After completion of creep rupture tests under irradiation, specimen diameter changes were measured at five locations along axial direction by means of the laser profilometry equipment.

Table 1
Chemical composition and manufacturing condition of PNC316 cladding

Compsotion (wt%)	55MK	55MS
C	0.052	0.047
Si	0.82	0.92
Mn	1.83	1.74
P	0.028	0.03
S	0.009	0.004
Ni	13.84	13.74
Cr	16.52	16.5
Mo	2.49	2.5
B	0.0031	0.0039
N	0.003	0.008
Ti	0.08	0.097
Nb + Ta	0.079	0.07
V	0.01	0.002
Co	0.01	0.005
Solution treatment	1353 K × 2 min	1358 K × 1 min
Cold-work	20.00%	19.20%
ASTM G.S.No.	8.0–8.5	8.3–8.8

2.3. Thermal control creep tests

As the thermal control creep strain tests, the creep strains of pressurized tubes were measured using the same kind of specimens as in-reactor creep rupture tests. These test conditions are shown in Table 3. The exposed temperatures of 878, 943 and 1023 K and the loaded hoop stress at each temperature are completely identical with that of in-reactor creep rupture tests. The creep strains of 55MK heat were measured in the argon gas atmosphere at an appropriate interval in order to compare the in-reactor rupture strains.

As reference to the creep rupture strength for both heats of 55MK and 55MS, creep rupture tests were also conducted in-air atmosphere. In these tests, cladding segments with 220.0 mm in length were loaded with argon gas keeping constant internal pressure inside the cladding segments. As shown in Table 4, temperature range lies in 873–1023 K, and each temperature is not the same as the in-reactor creep rupture one. Large amount of reference creep rupture data were acquired by setting at different hoop stress levels at each temperature. After the creep rupture tests, diameter changes were measured using a micrometer.

2.4. In-sodium creep rupture tests

In-sodium creep rupture tests were conducted using 160.0 mm long cladding segments in the sodium test loop of Oarai Engineering Center of JNC. The test conditions are shown in Table 5, in which test temperatures were

Table 2
In-reactor creep rupture tests

Heats	Irradiation temp. (K)	Hoop stress (MPa)	Ruptured time (h)	Fluence (10^{26} n/m^2 $E > 0.1$ MeV)	Number of hoop stress level
55MK	878	200.0–327.6	309.3–15169.4	0.50–19.36	7
	943	110.0–178.5	2010.6–7985.0	3.17–9.82	6
	1023	70.0–85.3	864.1–1301.2	1.37–2.12	2
55MS	878	260.0	5263.0	5.90	1
	943	127.5	4678.0–4898.0	5.24–5.52	2

Table 3
Thermal control creep strain tests

Heats	Temperature (K)	Hoop stress (MPa)	Number of hoop stress level
55MK	878	200.0–327.6	6
	943	110.0–178.5	5
	1023	70.0–85.3	2

Table 4
In-air creep rupture tests

Heats	Temperature (K)	Hoop stress (MPa)	Ruptured time (h)	Number of hoop stress level
55MK	873	385.1–474.3	146.5–1426.0	5
	923	246.0–363.6	417–3402.0	7
	973	132.3–315.6	88.9–5234.9	8
	1023	88.2–239.1	26.8–2690.0	14
55MS	873	381.2–464.5	174.0–2740.0	5
	923	251.9–371.4	423–4884.9	7
	973	132.3–311.6	126.0–4490.9	8
	1023	87.2–205.8	70.0–2689.2	14

Table 5
In-sodium creep rupture tests

Heats	Temperature (K)	Hoop stress (MPa)	Ruptured time (h)	Number of hoop stress level
55MK	923	156.3–402.8	29.0–14506.0	10
	948	125.8–283.1	648.5–9509.7	12
	973	69.2–157.3	1261.3–12230.0	7
55MS	923	207.7–304.9	870.4–11818.0	5
	948	157.3–254.8	2120.2–9667.3	14
	973	69.2–248.5	240.0–16036.0	10

selected at 923, 948 and 973 K. The rupture time was determined by means of monitoring the pressure drop in the pressure gauge. The purity of the sodium in the test loop was maintained by controlling the cold-trap temperature at 393 K, which corresponds to 1 ppm of the oxygen content in the sodium. The flow rate of the sodium in this experiment is 0.01 m/s.

2.5. Tensile tests

The tensile tests were conducted for the specimens cut from the cladding segments of both 55MK and 55MS heats to complementarily assess effects of neutron irradiation and sodium exposure on the high temperature creep rupture properties. The three temperature levels of 873, 923 and 973 K were selected for tensile tests after aging, sodium exposure and neutron irradiation. The tensile test conditions are shown in Table 6. The aging of tensile specimens was conducted in a vacuum of less than 0.1 Pa. The sodium exposure condition is the same as that for the in-sodium creep rupture tests. The duration for aging and sodium exposure ranges from about 2000–18 000 h. The neutron irradiation of the tensile specimens was conducted in the Core Material Irradia-

Table 6
Tensile test conditions

Heats	Temperature (K)	Aging time (h)	Sodium exposed time (h)	Irradiated time (h) (fluence [$\times 10^{26}$ n/m^2, $E > 0.1$ MeV])
55MK & 55MS	873	2000–18 000	2000–18 000	3850–14 520 (3.9–15.2)
	923	1000–18 000	5000–18 000	3850–14 520 (4.0–14.7)
	973	2000–18 000	2000–18 000	3850–18 740 (4.0–20.6)

tion Rig (CMIR) of JOYO up to the maximum irradiation time of 18,740 h and the fast neutron fluence of 20.6×10^{26} n/m^2. The tensile tests for these specimens were conducted in-air atmosphere at the strain rate of about 1×10^{-4} s^{-1}.

3. Results

The results of creep rupture tests in-air atmosphere are shown in Fig. 1, in which abscissa is time to rupture in hours and ordinate represents the hoop stress in MPa. The creep rupture data of 55MK and 55MS heats are not separately plotted in this figure, since both heats have similar creep rupture strength with small data scatter. The time to rupture covers 27–5235 h, as shown in Table 4. It is confirmed that during the creep rupture tests in-air atmosphere, oxide scale formed on the surface is confined within the thickness of about 5 μm, and that such oxide scale does not affect the creep rupture strength. Fig. 2 shows the in-sodium creep rupture data at temperatures of 923, 948 and 973 K. When compared with the in-air data of Fig. 1, there is no large difference at 923 K. At elevated temperature of 973 K, however, the creep rupture strength in sodium is significantly decreased with increasing rupture time above 1000 h. In-reactor creep rupture data are shown in Fig. 3 at the temperatures of 878, 943 and 1023 K. At temperatures of 878 and 1023 K, where it is possible to compare the data directly with those of the in-air at 873 and 1023 K, it is obvious that the creep rupture strength is degraded under the irradiation in wide range of rupture time of 309–15 169 h. In particular, decrease of creep rupture strength due to irradiation is remarkable at 1023 K.

In order to evaluate the shortened lives in time to rupture during irradiation, creep curves were measured. The typical in-reactor creep strain data are plotted for 878, 943 and 1023 K in Figs. 4–6, compared with the thermal control creep strain data. In these figures, hoop strain data of the in-reactor testing are limited only for the rupture point, whilst creep strains of the thermal control specimens were obtained at intervals of about 2000 h. It is presumed that the onset of tertiary creep becomes earlier and creep strains are significantly accelerated for the in-reactor creep, in contrast to the thermal control creep strains, although thorough tracing of the in-reactor creep strain could not be done. These results of enhanced in-reactor strain are consistent with the shortened time to rupture during irradiation that is shown in Fig. 3.

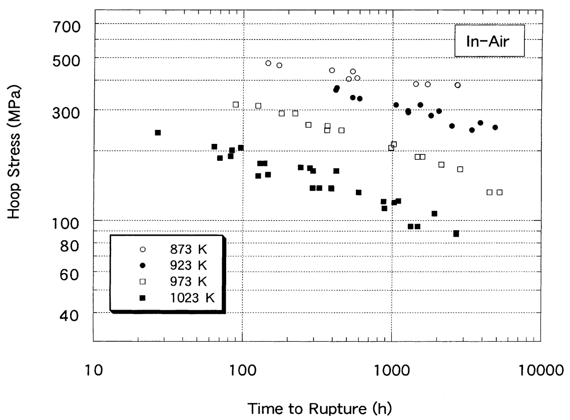

Fig. 1. The results of in-air creep rupture tests of PNC316 at the temperature ranging from 873 to 1023 K.

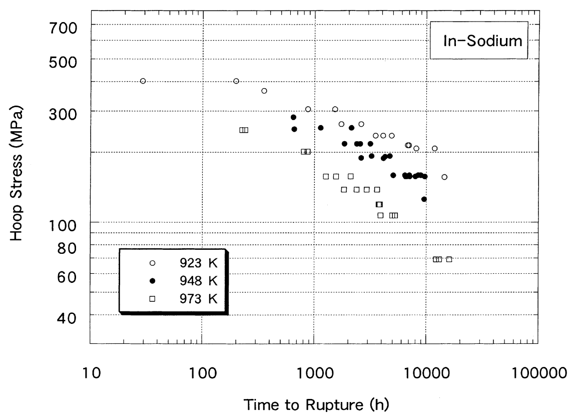

Fig. 2. The results of in-sodium creep rupture tests of PNC316 at the temperature of 923, 948 and 973 K.

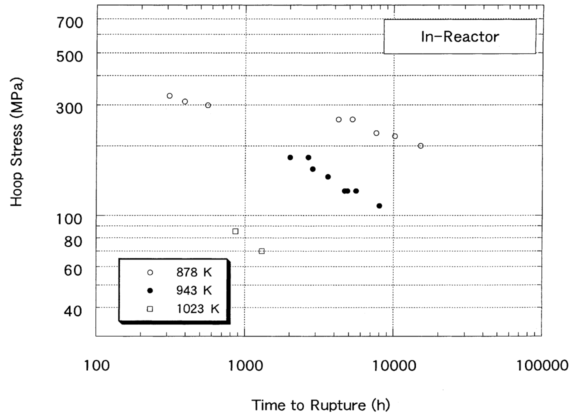

Fig. 3. The results of in-reactor creep rupture tests of PNC316 at the temperature of 878, 943 and 1023 K.

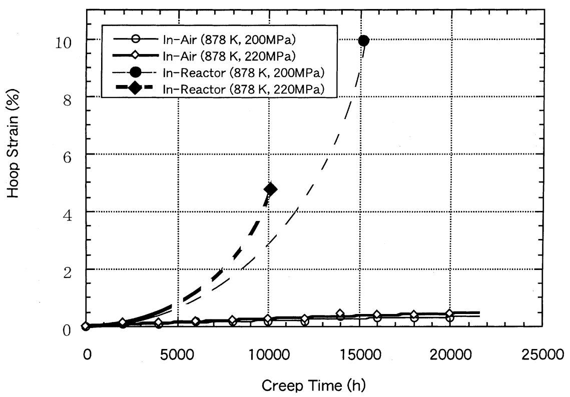

Fig. 4. Comparison of creep strains between in-reactor and thermal control at 878 K for two stress levels.

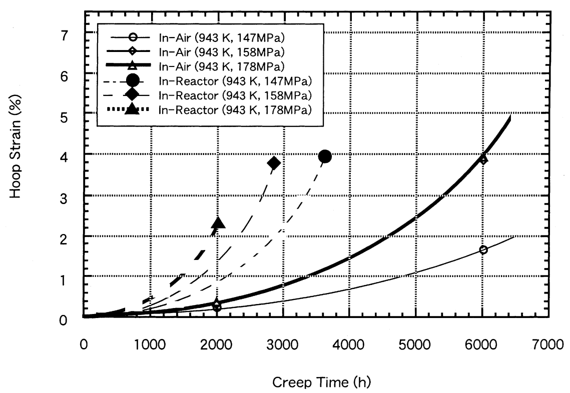

Fig. 5. Comparison of creep strains between in-reactor and thermal control at 943 K for three stress levels.

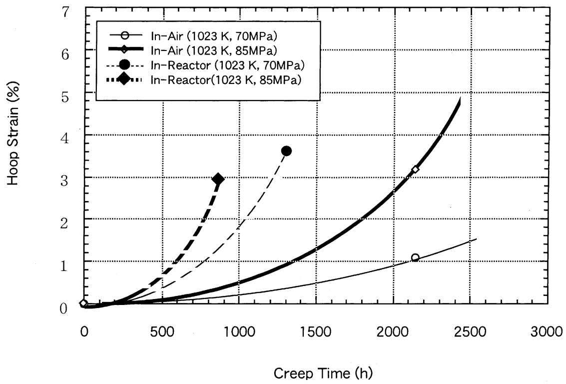

Fig. 6. Comparison of creep strains between in-reactor and thermal control at 1023 K for two stress levels.

4. Discussion

Concerning in-reactor creep properties, Lovell et al. [4] found that in-reactor rupture lives were equal or greater than the in-air creep rupture tests for 20% cold-worked AISI 316SS, based on the irradiation tests of instrumented subassembly in EBR-II. Puigh and Schenter [5] also reported as the first experiment of in-reactor creep rupture test using MOTA in FFTF that in-reactor rupture lives agreed well with the in-air creep rupture ones. However, Puigh and Hamilton pointed out in Ref. [6] that neutron irradiation eventually leads to a decrease in rupture lives for 20% CW 316SS and 10% CW D9 steel on the basis of FFTF-MOTA experiment, when compared with the in-air creep rupture data, which is contrary to his earlier indications of Ref. [5].

The reduction of the creep rupture lives due to neutron irradiation is sometimes discussed from the viewpoint of helium embrittlement [8–10]. The most important process for reduction of the creep rupture lives by helium embrittlement is the formation and growth of helium bubbles on the grain-boundaries, which eventually causes reduction of the rupture elongation as well. In our experiment, in-reactor creep rupture lives were clearly demonstrated to be reduced compared with the in-air creep rupture ones. The corresponding results of rupture strain measurement are shown in Fig. 7 as a function of Larson–Miller parameter (LMP) with a comparison between in-sodium and in-reactor tests that are only different in neutron irradiation environment. The specific form for the LMP is given by

$$\mathrm{LMP} = T[14.04 + \log(t_\mathrm{R})], \quad (1)$$

where T is temperature in K and t_R is the time to rupture in hours. The value of the constant, 14.04, was selected so as to correlate all of the data using a regression

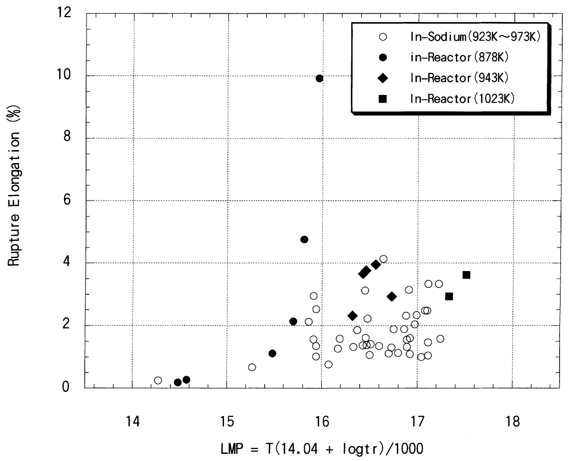

Fig. 7. Rupture elongation of in-reactor and in-sodium conditions as a function of Larson–Miller parameter.

analysis. In general, the rupture strain increases with increasing LMP, and in-reactor rupture strain appears to be greater than the in-sodium value. These results certainly suggest that helium embrittlement cannot be the dominant cause for the reduced in-reactor creep rupture lives in this experiment.

In order to directly evaluate the environmental effects on the rupture strength, in-air, in-sodium and in-reactor creep rupture data were summarized using the LMP, as shown in Fig. 8. The in-sodium creep rupture strength is similar to in-air data, but beyond approximately 16.5×10^3 for LMP the rupture lives in-sodium become relatively shorter than in the in-air condition. On the other hand, neutron irradiation causes a remarkable reduction in rupture strength throughout the LMP values from 14.5×10^3 to 17.5×10^3.

The tensile data of PNC316 were arranged with the temperature and duration exposed to the sodium and neutron irradiation, in order to investigate the reason for the reduced rupture lives under the sodium and neutron irradiation environments. Fig. 9 shows 0.2% proof strength after aging, sodium and neutron exposures at three temperature levels of 873, 923 and 973 K as a function of LMP. At 873 K, there is little difference in the 0.2% proof strength between the aged and sodium-exposed samples, whereas it is remarkably reduced after neutron exposure. This behavior surprisingly coincides with reduction of in-reactor creep rupture lives, when compared with Fig. 8. It is considered from these results that the irradiation-induced degradation of both creep rupture strength and tensile strength at 873 K results from the similar process. Such strength reduction due to neutron irradiation in 20% cold-worked modified 316SS could be mainly attributed to the earlier recovery of dislocation structure introduced by cold-working. This accelerated dislocation recovery should be owing to the jog motion of dislocations by absorbing the radiation-induced interstitials and vacancies. The coarsening

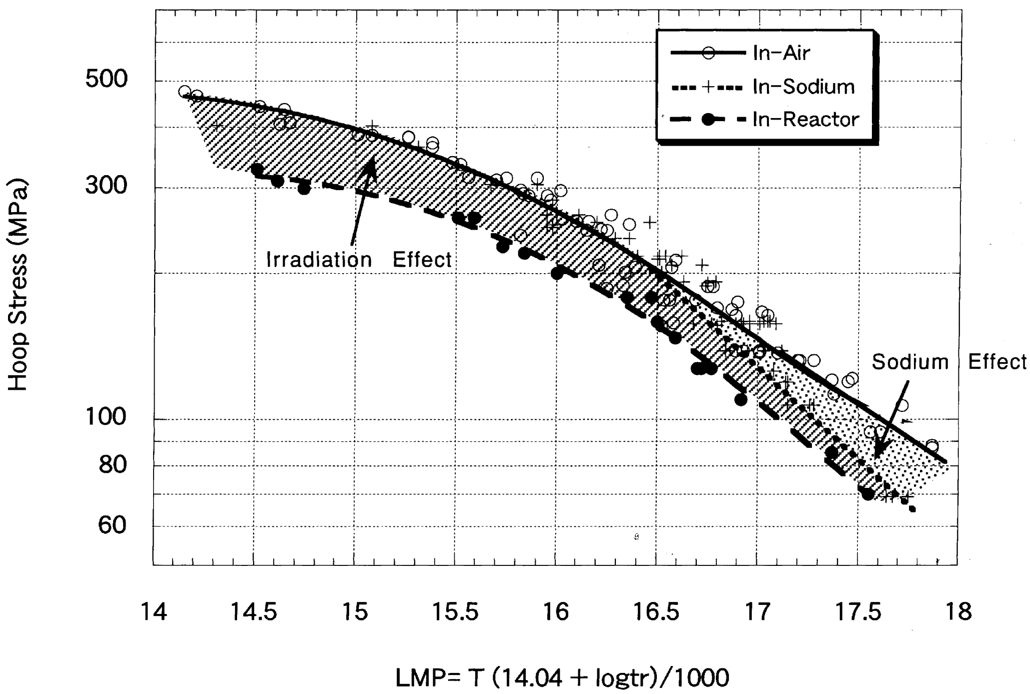

Fig. 8. Creep rupture curves for in-air, in-sodium and in-reactor conditions as a function of Larson–Miller parameter.

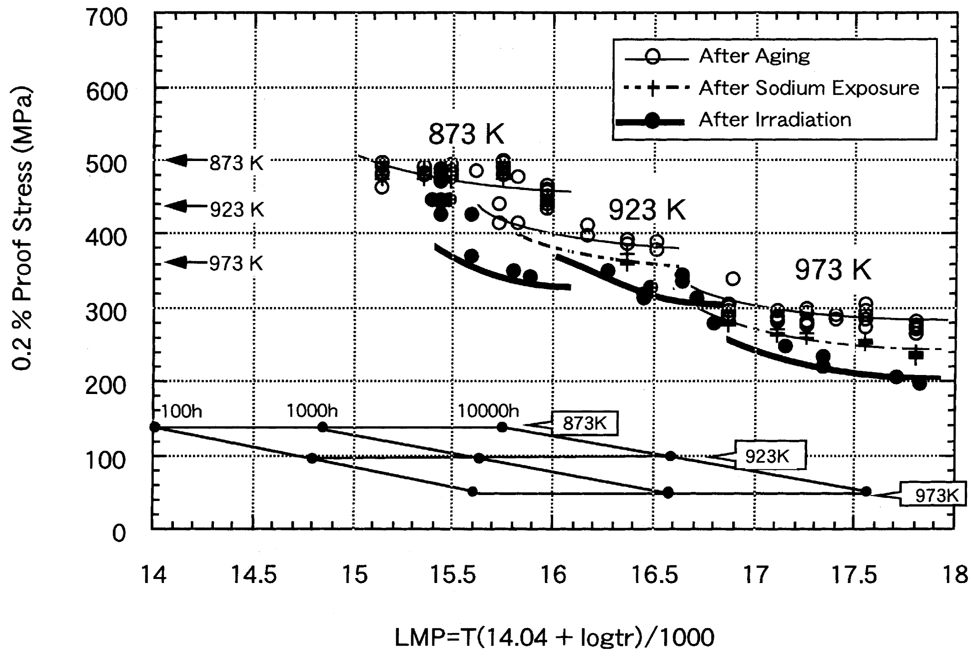

Fig. 9. 0.2% proof stress after aging, sodium and irradiation exposures as a function of Larson–Miller parameter.

of MC precipitates by the radiation-induced solute segregation may be indirectly associated with the early dislocation recovery [8]. The accelerated in-reactor creep strain and earlier onset of tertiary creep as shown in Figs. 4–6 seem to be also related with the enhanced dislocation recovery during irradiation.

At 923 and 973 K in Fig. 9, 0.2% proof strength after sodium exposure shows tendency toward more decrease than that after aging with increasing LMP above approximately 16.5×10^3, which also seems to agree completely well with the in-sodium creep rupture lives in Fig. 8. Concerning the sodium environmental effect on the tensile and creep rupture properties, extensive works have been conducted using sodium loop at Oarai Engineering Center. In PNC316, 300 ppm phosphorus as well as 35 ppm boron are contained to stabilize the MC precipitates [1,2]. It was considered that sodium environment enhanced dissolution of phosphorus and boron into the sodium, and mechanical property degradation after sodium exposure could be caused by earlier coarsening of MC precipitates [11].

0.2% proof strength after neutron irradiation at 923 and 973 K tends to decrease more than after sodium exposure as shown in Fig. 9. These tensile behavior seems to be similar to the in-reactor creep rupture trend of Fig. 8. In Fig. 8, the in-air, in-sodium and in-reactor creep rupture properties are represented by the regression curves. The degradation of in-reactor creep rupture lives compared with the in-air test should be adequately expressed in terms of neutron irradiation and sodium exposure effects. The neutron irradiation makes the enhanced dislocation recovery and its effect is dominant in the low LMP region. At increasing LMP above approximately 16.5×10^3, which corresponds to the high temperature of 943 and 1023 K, the effect of neutron irradiation is relatively weakened due to an enhancing thermal effect. On the other hand, the shortened lives of creep rupture due to sodium exposure appear at increasing LMP above approximately 16.5×10^3, which should be dominated by the thermal activation process with the solute elements dissolution into sodium.

5. Conclusion

The following conclusions are derived from this work.

1. In-reactor creep rupture lives are shortened in 20% cold worked modified 316 stainless steel, as compared with the results of out-of-reactor creep rupture tests.
2. In-reactor creep also appears to show earlier onset of tertiary stage, and it is significantly accelerated. The resulted larger rupture strains in the in-reactor creep tests support that the reduced rupture lives are not ascribable to the helium embrittlement.
3. From the similar trend of 0.2% proof strength after neutron irradiation, sodium exposure or aging to that of the creep rupture properties, it is concluded that the degraded lives of in-reactor creep are adequately expressed in terms of enhanced dislocation recovery

due to neutron irradiation as well as solute elements dissolution into sodium.

Acknowledgements

Execution of this experimental program required the active cooperation of many people in JNC and US, and we wish to gratefully acknowledge their support and encouragement. In particular, we thank Mr S. Shikakura, Mr I. Shibahara and members of Fuels and Materials Division in Oarai Engineering Center as well as Mr D. Watrous and Dr R.J. Puigh of Hanford in the US side. The authors are pleased to thank Dr K. Iwata for discussions and reviewing of this manuscript.

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