Effects of Implanted Deuterium on *In-Situ* Irradiation Fatigue Behaviors of Cold-Worked 316 Stainless Steel at 60 and 300°C

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Stress-controlled fatigue tests were performed for 20% cold-worked 316 stainless steel under *in-situ* 10 MeV deuteron irradiation with an energy degrader at 60 and 300°C. The irradiation dose rate and the deuterium implantation rate were arranged in response to beam intensity in three conditions as (A) 1.0×10^{-7} dpa/s and 1.1×10^{-3} appm/s, (B) 5.0×10^{-7} dpa/s and 5.5×10^{-3} appm/s and (C) 1.0×10^{-6} dpa/s and 1.1×10^{-2} appm/s, respectively. The number of fatigue cycles to fracture (N_F) increased with the dose rate in order of conditions (A), (B) and (C) at 300°C, while N_F was lower in condition (C) than that in condition (B) irrespective of the higher dose rate at 60°C. Although transgranular cracking in the ductile fracture mode was indicated on fracture surface for all specimens, higher density of dimples was formed for the *in-situ* irradiation specimen in condition (C) at 60°C. Some evidence of effects of implanted deuterium on *in-situ* irradiation fatigue behavior were demonstrated in this paper. [doi:10.2320/matertrans.MBW201014]

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1. Introduction

The *in-situ* irradiation effect on fatigue behavior is one of the key issues in evaluating the feasibility of nuclear reactor materials subjected to not only severe atomic displacement damage but also simultaneous periodic external loads. In the course of extensive efforts to accumulate available experimental data on *in-situ* irradiation fatigue tests,¹⁻³⁾ the mechanism of *in-situ* irradiation effect on fatigue behavior has been proposed in terms of the interaction between radiation-induced defect (RID) clusters and moving dislocations. In addition to these two factors of *in-situ* irradiation and fatigue loading, structural materials are also exposed to hydrogen isotopes due to nuclear transmutation and permeation from light-water coolants in the reactor core environment. Although the important role of hydrogen in irradiated assisted corrosion cracking (IASCC) has been suggested in some literature,^{4,5)} the effects of hydrogen isotopes on material behaviors under *in-situ* irradiation have been little examined due to some technological difficulties to establish *in-situ* irradiation testing facilities.

In the present study, 10 MeV deuteron irradiation is adopted to produce the displacement damage and hydrogen isotopes in a small specimen. Stress-controlled fatigue tests were conducted for 20% cold-worked 316 stainless steel under *in-situ* 10 MeV deuteron irradiation with the energy degrader at 60 and 300°C. The objective of the present work was to investigate the effects of implanted deuterium on *in-situ* irradiation fatigue behavior at these temperatures.

2. Experimental

The material used in the present study was a commercial SUS316 stainless steel. Its composition is 7.82 mass% Cr, 1.98 mass% W, and Mo, Mn, V, Ta, Ni, Si and C below 1 mass% in sum total and Fe for the balance. The annealed sheets of the alloy with the grain controlled to 8 μ m in diameter were rolled to 20% reduction in thickness to

produce the cold-work sheets. The cold-worked sheet of 90 μ m in thickness was punched out into the specimen type shown in Fig. 1. A fatigue starter side-notch was introduced into the gauge of the specimen by means of spark erosion, as presented in Fig. 1. Fatigue loading mode was tension-tension in stress control under a constant loading rate of 50 MPa/s with 416.9 $\leftrightarrow$ 183.6 MPa and 381.6 $\leftrightarrow$ 167.9 MPa at 60 and 300°C, respectively. In the present 10 MeV deuteron irradiation, an energy degrader was used to maximize and homogenize the rate of deuterium implantation into the 90 μ m-thick-specimen. In response to beam intensity, the irradiation dose rate (R_{dpa}) and the deuterium implantation rate (R_{imp}) were arranged in three conditions; (A) 1.0×10^{-7} dpa/s and 1.1×10^{-3} appm/s, (B) 5.0×10^{-7} dpa/s and 5.5×10^{-3} appm/s and (C) 1.0×10^{-6} dpa/s and 1.1×10^{-2} appm/s, respectively. The ratio of R_{imp} to R_{dpa} was a constant of 1.1×10^4 appm/dpa for all irradiation conditions. Figure 2 shows the damage and implantation profiles with depth (D_p) from the irradiated surface in condition (C) calculated by SRIM 2006 code. The specimen was cooled by circulating helium gas in order to compensate for beam heating. The specimen temperature was controlled at $60 \pm 3^\circ\text{C}$ and $300 \pm 5^\circ\text{C}$ by adjusting the output of the helium gas heater during the fatigue tests. Experimental details of the *in-situ* irradiation fatigue testing machine are described elsewhere.¹⁾

During the fatigue tests, the relative displacement of specimen gauge (Δd) was plotted as a function of the number of fatigue cycles (N) for all the fatigue tests. The relative displacement is the difference of the actual displacement at the upper stress between the N th cycle and the first cycle. The scattering of the number of cycles to fracture (N_F) is estimated to be within $\pm 15\%$ as a result of numerous function tests for fatigue specimens shown in Fig. 1. After the fatigue tests, the fracture surface was examined by a scanning electron microscope (SEM, JSM-5310) for all specimens. Tensile tests were also performed at 60 and 300°C for the irradiated specimens without a side-notch at the respective

Fig. 1 Dimensions of specimen for fatigue test.

Fig. 2 The damage and implantation profile calculated by SRIM 2006 code in irradiation condition (C).

temperature. The changes in tensile properties were measured from the pre-irradiation dose levels, ranging up to 0.138 dpa.

Although the measurement of thermal desorption spectroscopy (TDS) could not be performed for the irradiated specimen in the present experiments, the equilibrium deuterium concentration in specimen under irradiation at steady state was roughly estimated by a simple diffusion equation. Figure 3 shows the schematic deuterium concentration in the specimen. A simple diffusion equation is described as below,

$$du/dt = D \times d^2u/dx^2 + J_0 \quad (1)$$

where $u(x, t)$, D and J_0 are deuterium concentration (m^{-3}), deuterium diffusivity (m^2/s) and deuterium implantation rate (m^{-3}/s), respectively. At steady state, $du/dt = 0$, a solution of eq. (1) is represented as

$$u(x) = J_0/2D \times (l-x)x + u_0 \quad (2)$$

In eq. (2), l and u_0 are specimen thickness (90 μm) and deuterium concentration at the specimen surfaces ($x = 0, l$), respectively. When the deuterium recombination coefficient, K_0 (m^4/s), is assumed to be similar at both surfaces, boundary condition at specimen surface for the solution is

$$\phi/2 = K_0 \times u_0^2 \quad (3)$$

Fig. 3 Schematic deuterium concentration profile in simple diffusion model.

Table 1 The estimated value of equilibrium deuterium concentration at 60°C and 300°C.

Temperature	Irrad. Cond.	u_0 (appm)	u_p (appm)
60°C	A	3.0	1.5×10^2
	B	6.6	7.2×10^2
	C	9.4	1.4×10^3
300°C	A	1.7	2.0
	B	3.9	5.1
	C	5.5	7.9

where ϕ is the equilibrium deuterium flux ($= J_0 \times l$). The value of ϕ is $8.42 \times 10^{16} (\text{m}^{-2}/\text{s})$ in irradiation condition (C) in the present experiment. The concentration $u(x)$ at steady state is derived from eqs. (2) and (3) in applying the experimental results of D and K_0 at T (K) for 304 stainless steel⁶⁾ expressed as

$$D (\text{m}^2/\text{s}) = 3.5 \times 10^{-8} \exp(-Q_D/RT) \quad (4)$$

and

$$K_0 (\text{m}^4/\text{s}) = 8.3 \times 10^{-31} \exp(-E/RT) \quad (5)$$

where Q_D ($= 42400 \text{ J/mol}$) and E ($= 7000 \text{ J/mol}$) are activation energies of diffusion and recombination coefficients, respectively. Table 1 presents the estimated values of equilibrium deuterium concentration (appm), u_0 ($x = 0$) and u_p ($x = l/2$), in irradiation conditions (A), (B) and (C) at 60 and 300°C. Some upward revision of deuterium concentration could be expected at 60°C when the effect of deuterium trap at RID clusters is considered in the simple diffusion model. Although the present estimation only provides tentative values due to adoption of D and K_0 for different material (304 stainless steel) as well as disregard of deuterium trap, a significant difference of deuterium concentration between 60 and 300°C was indicated in each irradiation condition.

3. Results

Figure 4 shows the plots of (Δd) as a function of N for the unirradiated, *in-situ* irradiation specimens at (a) 60 and (b) 300°C. The number of cycles to fracture (N_F) was larger under *in-situ* irradiation conditions than that in unirradiated condition at both 60 and 300°C. The extension of N_F under

Fig. 4 Relative displacement in specimen gauge as a function of the number of cycles for unirradiated and *in-situ* irradiation specimens at (a) 60°C and (b) 300°C.

in-situ irradiation increased in order of conditions (A), (B) and (C) at 300°C (see Fig. 4(b)), while in order of (A), (C) and (B) at 60°C (see Fig. 4(a)). Thus, the extension of N_F was enhanced with irradiation dose rate at 300°C, whereas not in that manner at 60°C. The changes in tensile stresses with dose level are shown in Table 2. The increase of ultimate tensile stress (ΔUTS) and 0.2% offset yield stress ($\Delta 0.2\% YS$) was 270 MPa and 230 MPa at 0.138 dpa and 60°C, similarly 170 MPa and 115 MPa at 0.137 dpa and 300°C respectively. This indicates that irradiation hardening is induced by introduction of RID clusters for specimens during *in-situ* irradiation fatigue tests at both 60 and 300°C. After the fatigue tests, the fracture surface was examined by SEM for all specimens. The fractographic morphology of fracture surface was quite similar for all specimens and characterized by glide plane decohesions, fatigue striations and dimples, with an indication of transgranular cracking in the ductile fracture mode. However, for the *in-situ* irradiation specimen in condition (C) at 60°C, a higher density of dimples was detected on the fracture surface. The SEM photos of typical fracture surface for unirradiated and *in-situ* irradiation specimens in condition (C) are shown in Fig. 5 at (a) 60°C and (b) 300°C. Figure 6 presents the plotting of striation spacing (s_p) as a function of the distance from the notch tip (d_n) for each specimen at (a) 60°C and (b) 300°C. Since

Table 2 The changes in tensile stresses with dose level at 60°C and 300°C.

Temperature	Dose level (dpa)	0.2% Yield Stress (MPa)	Tensile Strength (MPa)
60°C	0	690	800
	0.0306	820	960
	0.0612	905	1025
	0.1380	960	1030
300°C	0	590	695
	0.0513	760	820
	0.1370	760	810

Table 3 Number of cycles for fatigue process in each irradiation condition at 60°C and 300°C.

Temperature	Irrad. Cond.	N_F	N_c	N_i
60°C	unirradiated	9419	4866	4553
	A	13864	5809	8055
	B	17849	6581	11268
	C	14452	5240	9212
300°C	unirradiated	6054	2561	3493
	A	9983	3963	6020
	B	11896	4798	7098
	C	15990	6791	9199

striation spacing was too small to be resolved by SEM in the vicinity of the notch tip, the measurement was conducted in the area between the distance starting at 60 μm from the notch tip up to the unstable ductile fracture point. This area is designated as the striation area in the present paper. Striation analysis^{2,3)} was conducted to calculate the number of fatigue cycles in the striation area (denoted as N_c) for all specimens. The difference between N_F and N_c is denoted as N_i , corresponding to the number of cycles for crack initiation and growth until 60 μm in length. Table 3 presents N_F , N_c and N_i for all specimens. These results indicate that the extension of N_F was attributed to the increases in both N_c and N_i under *in-situ* irradiation conditions. The increase of N_c and N_i with irradiation dose rate was quite similar in its inclination to that of N_F , namely in order of conditions (A), (B) and (C) at 300°C, whereas in order of (A), (C) and (B) at 60°C.

4. Discussion

Fatigue behaviors of 20% cold-worked 316 stainless steel under *in-situ* 17 MeV proton irradiation at 60 and 300°C have been investigated in previous papers.^{2,3)} Striation analyses have revealed the prolonged fatigue processes of crack initiation and propagation under *in-situ* irradiation at both temperatures. It is well recognized that the development of inhomogeneous dislocation structures such as cells and wall-channels is closely related to the progress of fatigue processes in ductile fracture mode. Since the radiation-induced defect (RID) clusters can act as obstacles for mobile dislocations, the continuous introduction of RID clusters under *in-situ* irradiation would severely impede the rearrangement of dislocation structures. Therefore, the effect of *in-situ* irradi-

Fig. 5 SEM photos of typical fracture surface for the unirradiated and *in-situ* irradiation specimens in condition (C) at (a) 60 °C and (b) 300 °C.

Fig. 6 Plots of striation spacing with distance from notch tip for the unirradiated and *in-situ* irradiation specimens at (a) 60 °C and (b) 300 °C.

ation on fatigue behavior has been explained in terms of the interaction between RID clusters and moving dislocations.^{2,3)} On the other hand, the effect of hydrogen on the interaction between dislocations in 310S stainless steel has been examined during *in-situ* deformation experiments by transmission electron microscope (TEM).⁷⁾ It has been directly demonstrated that solute hydrogen reduced the elastic interaction between dislocations, thereby enhancing the mobility of dislocations. Another aspect of hydrogen effect

in ductile fracture of steels has been pointed out on the basis of the hydrogen-enhanced strain-induced vacancy model.^{8,9)} Enhanced formation of strain-induced vacancies due to interaction with hydrogen promotes their agglomeration, leading to nucleation of a high density of micro-voids at the cross slips of dislocations in the vicinity of the crack tip. Therefore, the presence of hydrogen can induce the localization of unstable dimple fractures, leading to rapid crack propagation.

In the present experiments, the number of cycles to fracture (N_F) was larger under *in-situ* irradiation conditions than that in unirradiated condition at both 60 and 300°C, as shown in Fig. 4. Table 2 data suggest irradiation hardening, which is induced by introduction of RID clusters for specimens during *in-situ* irradiation fatigue tests at both 60 and 300°C. Striation analysis reveals that the extension of N_F was attributed to the increases in both N_C and N_i under *in-situ* irradiation conditions (see Table 3). Furthermore, the transgranular cracking in the ductile fracture mode was indicated by SEM observation on fracture surface for all specimens. These results strongly suggest that the extension of N_F under *in-situ* irradiation can be explained by the *in-situ* irradiation effect based on the interaction between RID clusters and moving dislocations. At 300°C, the increase of N_F as well as N_C and N_i with irradiation dose rate is shown in Table 3. Because continuous introduction of RID clusters is proportionally enhanced with dose rate, the increase of dose rate would simply reflect the increase of *in-situ* irradiation effect at this temperature. However, at 60°C, Table 3 shows lower N_F as well as N_C and N_i in irradiation condition (C) than those in (B) irrespective of the higher dose rate. Furthermore, the higher density of dimples on fracture surface was detected in irradiation condition (C) at 60°C (see Fig. 5(a)). Rough estimation of deuterium concentration during *in-situ* irradiation indicated a significant increase of deuterium concentration at 60°C in particular in condition (C), as shown in Table 1. Therefore, when a large amount of deuterium is existent in the material, the implanted deuterium can play a role in reducing the effect of *in-situ* irradiation on fatigue behavior.

Although a relative comparison of experimental results between different irradiation conditions has brought the presence of deuterium effect in condition (C) at 60°C into sharp relief, the evidence of deuterium effect in other irradiation conditions could not be detected in the present experiments. Since the balance of effects between *in-situ* irradiation and implanted deuterium would reflect fatigue behavior, the effect of deuterium can be covered by the effect of *in-situ* irradiation in the present comparative experimental methods. More extensive experiments of *in-situ* irradiation fatigue with precise arrangement of separate parameters between irradiation dose rate and deuterium implantation rate are definitely needed to clarify the threshold concentration of deuterium effects on fatigue behavior under *in-situ* irradiation.

5. Conclusions

Stress-controlled fatigue tests were performed for 20% cold-worked 316 stainless steel under *in-situ* 10 MeV deuteron irradiation with an energy degrader at 60 and 300°C. The irradiation dose rate was arranged in three conditions as (A) 1.0×10^{-7} dpa/s, (B) 5.0×10^{-7} dpa/s

and (C) 1.0×10^{-6} dpa/s under a constant deuterium implantation ratio of 1.1×10^4 appm/dpa. From the experimental results, the following conclusions can be drawn:

- (1) The number of cycles to fracture (N_F) was larger under *in-situ* irradiation conditions than that in unirradiated condition at both 60 and 300°C. The extension of N_F increased with irradiation dose rate at 300°C, while not in that manner at 60°C.
- (2) Transgranular cracking in the ductile fracture mode was indicated by SEM observation on fracture surface for all specimens. Higher density of dimples was detected on fracture surface of the *in-situ* irradiation specimens in condition (C) at 60°C.
- (3) The extension of N_F under *in-situ* irradiation can be explained by the *in-situ* irradiation effect based on the interaction between RID clusters and moving dislocations. Because continuous introduction of RID clusters is proportionally enhanced with irradiation dose rate, the increase of dose rate would simply reflect the increase of *in-situ* irradiation effect at 300°C.
- (4) Rough estimation of deuterium concentration during *in-situ* irradiation indicated a significant increase of deuterium concentration at 60°C in particular in condition (C). When a large amount of deuterium is existent in the material, the implanted deuterium can play a role in reducing the effect of *in-situ* irradiation on fatigue behavior.

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REFERENCES

- 1) Y. Murase, J. Nagakawa, N. Yamamoto and Y. Fukuzawa: *Creep-fatigue response of 20% CW 316SS under irradiation at 60°C*, (ASTM STP 1366, ASTM, West Conshohocken, 2000) pp. 713–724.
- 2) Y. Murase, J. Nagakawa and N. Yamamoto: *J. Nucl. Mater.* **302** (2002) 211–216.
- 3) Y. Murase, J. Nagakawa and N. Yamamoto: *Fusion Engin. Design* **81** (2006) 999–1003.
- 4) K. Fukuya, M. Nakano, K. Fujii and T. Torimaru: *J. Nucl. Sci. Technol.* **41** (2004) 673–681.
- 5) G. Furutani, N. Nakajima, T. Konishi and M. Kodama: *J. Nucl. Mater.* **288** (2001) 179–186.
- 6) M. Takizawa, K. Kikuchi, M. Okamoto and Y. Fujii: *J. Nucl. Mater.* **248** (1997) 15–18.
- 7) P. J. Ferreira, I. M. Robertson and H. K. Birnbaum: *Acta Mater.* **46** (1998) 1749–1757.
- 8) K. Takai, H. Shoda, H. Suzuki and M. Nagumo: *Acta Mater.* **56** (2008) 5158–5167.
- 9) M. Nagumo: *Mater. Sci. Technol.* **20** (2004) 940–950.