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Journal of Nuclear Materials 283–287 (2000) 263–267

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Microstructural changes of austenitic steels caused by proton irradiation under various conditions

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Abstract

In austenitic steels used for light water reactors (LWRs), neutron irradiation induces many kinds of degradation. For example, irradiation assisted stress corrosion cracking (IASCC) and swelling are two forms of this degradation. Although there are a great number of studies on radiation induced segregation (RIS) and void swelling at high temperatures (>400° C) corresponding to fast and fusion reactor conditions, up to now there have been few irradiation studies at low temperatures. This paper presents microchemical and microstructural changes in type 347 and 310 + Nb stainless steels due to light ion irradiation. These samples were implanted with He^+ and irradiated with 2 MeV H_2^+ at 300°C, 350°C and 400°C. This simulated generation of transmutant He in a fusion environment. In addition, at 300°C test pieces stressed close to the yield stress were also irradiated with the same ions. The irradiation tests were carried out using the Dynamitron accelerator at Tohoku University. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Austenitic alloys used in a neutron irradiation environment are generally known to suffer many kinds of degradation. Although there are many studies on radiation induced segregation (RIS) and void swelling at high temperatures (>400°C), corresponding to fast and fusion reactor conditions, there are few irradiation studies at low temperatures. Thus, it seems useful to obtain data for low temperature irradiation in order to fully understand RIS of austenitic alloys.

According to previous literature regarding irradiation damage in austenitic alloys under fast reactor conditions, the mobility of point defects increases at higher temperatures, which results in suppressing defect recombination [1]. Accordingly, since the mobility of the defects decreases at low temperature, irradiation induced degradation may be attenuated below 400°C. However, the degradation below 400°C has not been clarified. This study was therefore focused on microchemical and microstructural changes of high purity SUS347 and higher carbon content SUS310 + Nb steels at 300°C, 350°C and 400°C using a light ion accelerator to simulate neutron irradiation. In addition at 300°C test pieces stressed closed to yielding were also irradiated with the same ions.

2. Experimental procedure

The alloys consisted of high purity SUS347 and SUS310 + Nb with higher carbon content. Results of chemical analysis of the steels are shown in Table 1. Seventy five percent cold-worked alloy sheets of 0.25 mm in thickness were recrystallization annealed at 1000°C for 5 min. The grain sizes of these steel sheets were controlled to be under 10 μm, because larger grain size provides a limited number of grain boundaries for transmission electron microscopy (TEM) observation. The irradiation samples were punched disks with a 3 mm diameter or machined micro tensile specimens. Electro-

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0022-3115/00/$ - see front matter
PII: S0022-3115(00)00356-1

Table 1
Chemical composition of the alloys (wt%)

Alloy type	C	Si	Mn	P	S	Ni	Cr	Mo	Nb	B	N	V	Fe
SUS347	0.027	0.010	0.01	0.001	0.0015	10.30	17.65	<0.02	0.420	0.000	0.001	0.005	Bal
SUS310+Nb	0.025	0.250	0.69	0.010	0.0006	20.45	25.30	0.150	0.170	0.001	0.002	0.013	Bal

chemical polishing removed the surface layer of the disks to 0.15 mm in thickness.

Irradiation was carried out using 3 MeV He^- ions at room temperature to simulate the effect of transmutant He generation, and 2 MeV H_2^+ ions at 300°C to produce displacement damage of 1 dpa at a dose rate of approximately 4×10^{-5} dpa s^{-1}. 15 appm He ions were injected prior to H_2^- ion irradiation, corresponding to the amount of He generation from Ni (n,α) reaction. The depth distribution of preinjected He and displacement damage were estimated using the TRIM-96 code. During H_2^+ irradiation, the target temperature was controlled to 300°C through beam heating and a heat sink placed at the backside of the disk, and monitored by an infrared pyrometer. The irradiation beam probe was controlled to be 3 mm in diameter using a collimator. An energy degrader was used to make the distribution of injected He atoms uniform at depths ranging from 1.5 to 3.5 μm. Applied stress irradiations were carried out using a constant load apparatus. Irradiated samples were thinned through electro-chemical polishing by South Bay Technology model 550D single jet thinning apparatus. The electrolytic solution comprised 90% acetic acid and 10% perchloric acid solution at 15–25°C. Samples were thinned from the irradiated side until reaching the depth of observation and then polished on the back side until the thickness became suitable for TEM observation.

Chemical compositions at grain boundaries and microstructure were examined using a field emission gun transmission electron microscope (FETEM), Hitachi HF-2000 and energy dispersive X-ray analysis (EDX), Kevex. The thickness of the TEM observation area was evaluated by electron energy loss spectroscopy (EELS). Quantitative analysis of the chemical composition was performed with a probe under 1 nm diameter.

3. Experimental results

3.1. Grain boundary segregation

Significant changes in compositions were observed within 5 nm of the grain boundary in each irradiated sample. Elements analyzed were Fe, Ni, Cr, Si, P, S, Mn and Mo. Fig. 1 shows the Cr concentration profile across a grain boundary for various irradiation temperatures. In SUS310 + Nb, Cr was depleted near the grain boundary after irradiation and the depletion became wider and larger with increasing irradiation temperature. However, a different trend was observed in SUS347. Chromium depletion was the largest at 350°C irradiation in SUS347.

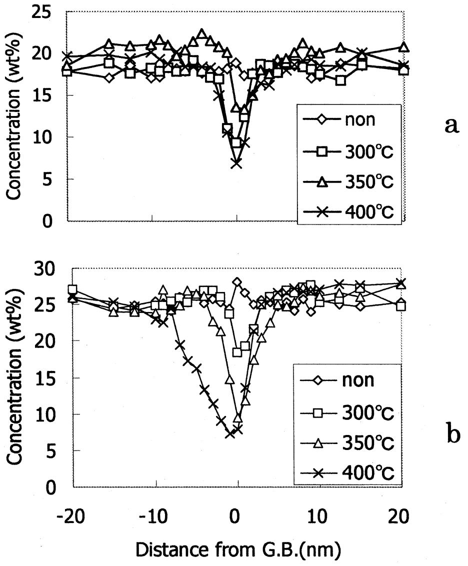

Fig. 1. Cr concentration profile across grain boundaries of (a) SUS347 and (b) SUS310+Nb irradiated at various temperatures.

3.2. Microstructural change

Results of TEM examination of microstructures and analysis are summarized in Table 2, Swelling S_w was calculated for TEM observations by

$$S_w = \Delta V/(V_{irr} - \Delta V), \tag{1}$$

where ΔV is the increased volume after irradiation and V_{irr} is total volume after irradiation. Fig. 2 shows the results of TEM observations for SUS347 and SUS310 + Nb irradiated at 300°C, 350°C and 400°C. Fig. 3 shows the results of the variation of swelling with irradiation temperature. The swelling of SUS310 + Nb shows a monotonic increase with increasing irradiation temperature. On the contrary, the swelling of SUS347 has a swelling peak at 350°C. Fig. 4 shows the results of TEM observation on samples with and without applied stress. In these pictures stress was applied in the hori-

Table 2
Summary of TEM observations and analysis[a]

Alloy type	He concentration (appm)	Dose (dpa)	Temperature (°C)	Cr bf (wt%)	Cr af (wt%)	Delta Cr (wt%)	Ni bf (wt%)	Ni af (wt%)	Delta Ni (wt%)	Cavity size (nm)	Swelling (%)
SUS 347	15	1	300	17.65	9.75	7.9	10.3	22.56	219.0	8.1	0.07
SUS 347	15	1	350	17.65	14.92	2.7	10.3	18.67	181.3	18.2	2.81
SUS 347	15	1	400	17.65	9.04	8.6	10.3	19.77	191.9	7.6	0.31
SUS 347 st	15	1	300	17.65	11.58	6.1	10.3	24.66	239.4	14.2	1.00
SUS 310+Nb	15	1	300	25.3	17.93	7.4	20.45	30.25	147.9	5.4	0.02
SUS 310+Nb	15	1	350	25.3	9.15	16.2	20.45	48.39	236.6	10.7	0.18
SUS 310+Nb	15	1	400	25.3	9.12	16.2	20.45	32.18	157.4	10.1	0.52
SUS 310+Nb st	15	1	300	25.3	13.97	11.3	20.45	39.24	191.9	8.9	0.32

[a] st: Irradiated with stress applied. bf: Concentration before irradiation at G.B. af: Concentration after irradiation at G.B. Delta: absolute value of concentration changing at G.B.

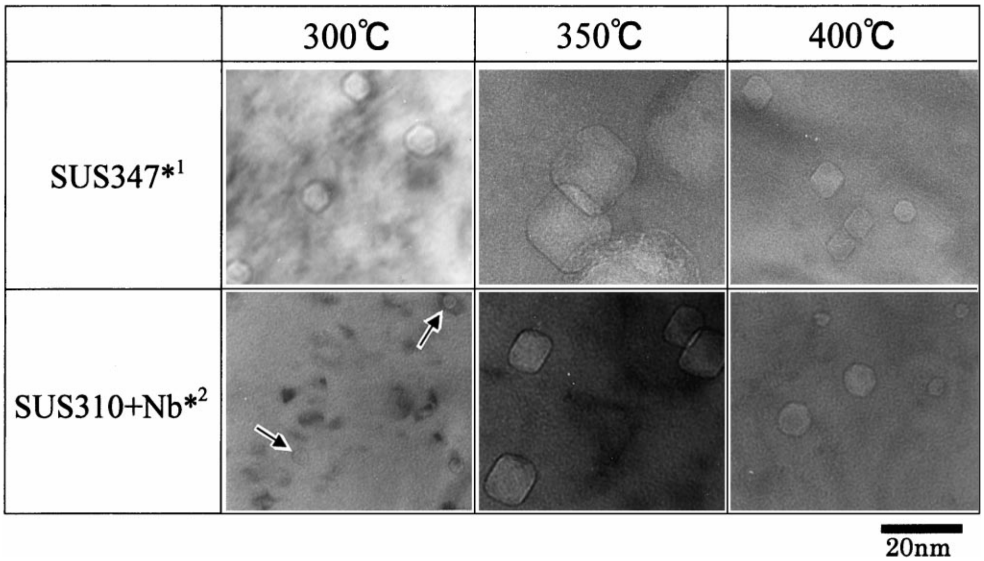

Fig. 2. TEM observation of SUS347 and SUS310+Nb irradiated at 300°C, 350°C and 400°C.

zontal direction. Although the shapes of voids in irradiated samples with and without applied stress were very similar, the void density and diameter were larger in samples with applied stress.

4. Discussion

4.1. Microchemical and microstructural changes with temperature increment

The amount of segregation increased with increasing irradiation temperature in this study. This is because vacancies and interstitials easily recombine at lower temperatures. On the other hand, at higher temperature the interstitial and vacancies move to sinks, such as surfaces and grain boundaries. As a result the amount of segregation increases. Swelling increases with increasing temperature in SUS310 + Nb, but for SUS347 swelling behavior showed a peak at 350°C.

4.2. The effect of applying stress on grain boundary segregation and microstructural change

Radiation induced Cr segregation appeared to be accelerated by a applied stress in the case of

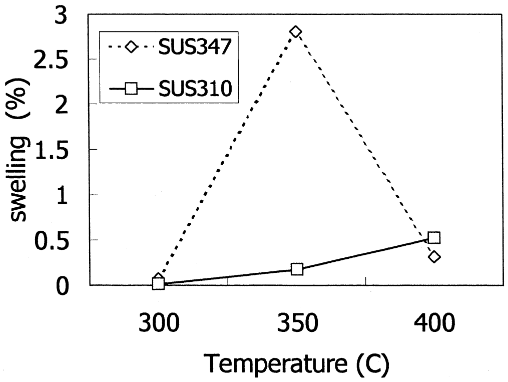

Fig. 3. Variation of swelling with irradiation temperature.

SUS310 + Nb, but applied stress had the opposite effect in SUS347. The amount of Ni segregation was increased at grain boundaries in both of the alloys under an applied stress, suggesting that the contribution of interstitial defects to segregation increased with applied stress. Once a large number of cavities form in the alloy, vacancies are used in growing these cavities. When higher helium was introduced, the vacancies seemed to be trapped by small He cavities. When the vacancy type defects are decreased, this contributes to segregation of Cr in the matrix.

Tanigawa [2] reported dislocations growing in a sample irradiated under applied stress conditions. In this report, the result showed that irradiation with an applied stress induced Flank loop unfaulting to form perfect loops. When the stress increased, the distribution of perfect loop diameters became larger [2,3]. In the same way, the applied stress seemed to induce a large size in this study. The applied stress during irradiation was kept near the yield stress in both alloys (203 MPa in SUS310 + Nb and 218 MPa in SUS347). This condition was chosen to simplify the analysis for microstructural changes due to irradiation. In this condition, dislocations are not moved by the applied stress and defect migration to voids may be enhanced by the applied stress.

4.3. Correlation between swelling and segregation

One of the purposes of this study was to investigate the correlation between RIS and swelling. The SUS310 + Nb samples showed a positive correlation between swelling and segregation with increasing temperature. On the other hand, SUS347 irradiated at 350°C showed larger swelling but a lower amount of segregation. Comparing the result of irradiation for SUS347 at 400°C with that at 350°C, indicates significant difference in Cr segregation. However, the increase in Ni segregation was insignificant as shown in Fig. 5. This result suggests that voids nucleated with a higher density, and that radiation induced vacancies were absorbed by voids while interstitials of undersized atoms such as Ni were absorbed at grain boundaries causing segregation.

4.4. Swelling behavior high purity alloy

Comparing the result of SUS347 commercial purity alloy with high purity SUS347 [4], swelling increased when the concentrations of Si, Mn and P are decreased.

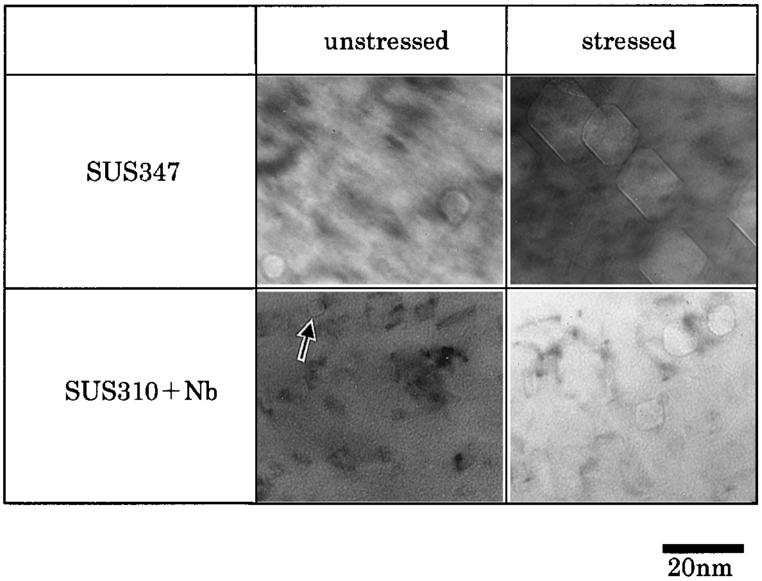

Fig. 4. TEM observation of SUS347 and SUS310+Nb irradiated with and without applied stress.

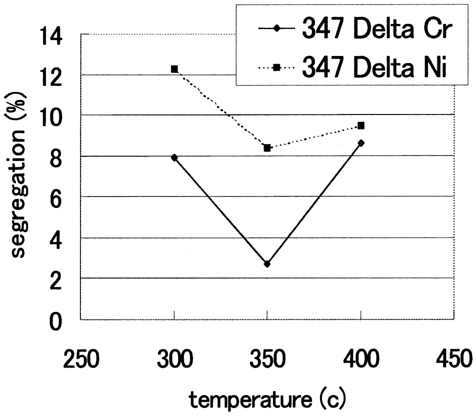

Fig. 5. Change of Cr and Ni concentrations at grain boundaries as a function of irradiation temperature.

Generally speaking silicon and phosphorus are effective in suppressing the swelling in fast breeder reactor conditions [5]. The same tendency was observed in SUS347 commercial alloy and SUS347 high purity alloy at <400°C in this study.

It is well known that IASCC occurs by grain boundary Cr depletion or Si enrichment [6]. In this study, SUS347 high purity alloy was aimed at reducing SCC susceptibility. Silicon was enriched at grain boundaries in commercial purity SUS347, however, in high purity SUS347 no segregation was detected at grain boundaries. However, a reduction in silicon concentration induced a larger amount of swelling in this study. These results suggest that there is a suitable level of silicon additions to suppress the swelling at low temperatures, similar to the FBR conditions reported in previous studies [7].

5. Conclusions

The following conclusions were found:

1. Swelling and RIS increased as the temperature of irradiation was raised.
2. The swelling was increased when stress was applied during irradiation.
3. The void shape was independent of the stress direction.
4. The reduction of minor element addition such as Si, Mn, P, S increased swelling.

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