Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Nuclear Materials 329–333 (2004) 643–647
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Effects of silicon, carbon and molybdenum additions on IASCC of neutron irradiated austenitic stainless steels

J. Nakano *, Y. Miwa, T. Kohya, T. Tsukada

Tokai Research Establishment, Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken 319-1195, Japan

Abstract

To study the effects of minor elements on irradiation assisted stress corrosion cracking (IASCC), high purity type 304 and 316 stainless steels (SSs) were fabricated and minor elements, Si or C were added. After neutron irradiation to 3.5×10^{25} n/m^2 ($E > 1$ MeV), slow strain rate tests (SSRTs) of irradiated specimens were conducted in oxygenated high purity water at 561 K. Specimen fractured surfaces were examined using a scanning electron microscope (SEM) after the SSRTs. The fraction of intergranular stress corrosion cracking (IGSCC) on the fractured surface after the SSRTs increased with neutron fluence. In high purity SS with added C, the fraction of IGSCC was the smallest in the all SSs, although irradiation hardening level was the largest of all the SSs. Addition of C suppressed the susceptibility to IGSCC.

1. Introduction

One of the specific concerns for application of austenitic stainless steel (SS) in the international thermonuclear experimental reactor (ITER) is irradiation assisted stress corrosion cracking (IASCC) [1]. IASCC is known as a degradation phenomenon that is caused by synergistic effects of neutron/gamma irradiation, stress/strain and high temperature water on structural materials. To evaluate the effects of minor alloying elements in SSs on IASCC, high purity type 304 and 316 SSs were fabricated and minor elements were added. Results of slow strain rate tests (SSRTs) in high temperature water and examination of the fractured surface after neutron irradiation to 6.7×10^{24} n/m^2 ($E > 1$ MeV) were reported already [2–4]. More than 20% elongation was observed in the high purity SSs with Si or Mo, and no intergranular stress corrosion cracking (IGSCC) was observed in high purity SSs with Mo. In the present study, stress–strain behavior in a SSRTs in high temperature water and susceptibility to IASCC of the same SSs were investigated to evaluate effects of additional elements, i.e., Si, C and Mo, after neutron irradiation to 3.5×10^{25} n/m^2 ($E > 1$ MeV).

2. Experimental

2.1. Materials

High purity type 304 and 316 SSs were fabricated and Si or C were added. The chemical compositions and notation of the SSs, i.e., HP304, HP304/Si, HP304/C and HP316, are shown in Table 1. After solution heat treatment at 1373 K for 0.5 h, round bar type specimens with 24 mm in gage length and 4 mm in diameter were machined.

2.2. Irradiation

An irradiation capsule (RGM-03H) was fabricated for this study and loaded into the Japan Research Reactor No. 3 (JRR-3M) at the Japan Atomic Energy Research Institute (JAERI). The specimens were irradiated at 543 ± 20 K in a helium gas atmosphere for

* Corresponding author. Tel.: +81-29 282 5348; fax: +81-29 282 5349.
E-mail address: lantis@popsvr.tokai.jaeri.go.jp (J. Nakano).

0022-3115/$ - see front matter
doi:10.1016/j.jnucmat.2004.04.098

Table 1
Chemical compositions of specimen materials (mass%)

Alloy-ID	C	Si	Mn	P	S	Cr	Ni	Mo	Ti	Al	N	Fe
HP304	0.003	0.01	1.36	0.001	0.0014	18.17	12.27	–	0.01	0.16	0.0014	Bal.
HP304/Si	0.003	0.69	1.36	0.001	0.0014	18.01	12.24	–	<0.01	0.10	0.0014	Bal.
HP304/C	0.098	0.03	1.39	0.001	0.0020	18.30	12.50	–	<0.01	0.11	0.0016	Bal.
HP316	0.004	0.02	1.40	<0.001	0.001	17.21	13.50	2.50	0.01	0.10	0.0020	Bal.

Bal. = Balance.

23 819 h. The fast neutron fluence was estimated from gamma spectrometry of fluence monitor materials after irradiation. The fast neutron fluence was estimated to be 3.5×10^{25} n/m^2 ($E > 1$ MeV).

2.3. SSRT

To study susceptibility to stress corrosion cracking (SCC), SSRTs of the irradiated specimens was conducted in oxygenated high purity water at 561 K in 9.0 MPa at a facility for post irradiation examination at JAERI. The applied strain rate was 1.7×10^{-7} s^{-1}. Dissolved oxygen (DO) concentration was controlled at 8 ppm. Electrical conductivity of the test water was monitored at the inlet and outlet of the autoclave and maintained below 0.06 μS/cm at inlet and 0.10 μS/cm at outlet. The flow rate of water was maintained at 3×10^{-2} m^3/h. The fraction of SCC on the specimen fractured surfaces was examined using a scanning electron microscope (SEM) following the SSRTs.

3. Results and discussion

SSRTs data are summarized in Table 2. Stress–strain curves from the SSRTs in this study are compared in Fig. 1 with the results of SSRTs and tensile tests for SSs irradiated to 6.7×10^{24} n/m^2 ($E > 1$ MeV) [2,3] and tensile tests for unirradiated SSs [4]. SEM microphotograph of fractured surface of HP304/C after the SSRT is shown in Fig. 2. The fraction of SCC for the SSs irradiated to 3.5×10^{25} n/m^2 ($E > 1$ MeV) and 6.7×10^{24} n/m^2 [2–4] are compared in Fig. 3, and the yield stress in the high purity SSs is plotted in Fig. 4 as a function of neutron fluence [2,3]. The fraction of IGSCC (%IGSCC) of the high purity SSs from this study are compared with data from Refs. [2,3,5–10] as a function of neutron fluence in Fig. 5.

3.1. Effect of silicon

In HP304/Si, elongation decreased, ultimate tensile strength increased, and %IGSCC reached more than 90% as the neutron fluence increased from 6.7×10^{24} to 3.5×10^{25} n/m^2. In HP304/Si irradiated to 6.7×10^{24} n/m^2, the number density of Frank loops was decreased by addition of Si, and irradiation hardening was suppressed [11]. It was thought that the incubation period for SCC fracture in the HP304/Si was longer than that for the other high purity SSs [2–4]. Chung et al. [5] reported that a Si concentration of 0.8–1.5 wt% was beneficial in delaying the onset of or suppressing the susceptibility to IASCC. However, the beneficial effect of Si was not observed at 3.5×10^{25} n/m^2. When comparing HP304 and HP304/Si in Figs. 1(a) and (b) and 3, it appears that the beneficial effect of Si disappeared or saturated.

3.2. Effect of carbon

In HP304/C, as the neutron fluence increased to 3.5×10^{25} n/m^2, the %IGSCC increased to 31% (see Fig. 3). Transgranular stress corrosion cracking (TGSCC) was observed on the fractured surface of the specimen (see Fig. 2). At 6.7×10^{24} n/m^2, the %IGSCC of HP304 was higher than that of HP304/C while the fraction of TGSCC (%TGSCC) of HP304 was lower than that of HP304/C. This indicated that the %TGSCC seemed to increase with C concentration [2]. Chung et al. [12] reported that higher C content seemed to suppress the susceptibility to IGSCC and that type 304L SSs ap-

Table 2
Results of SSRTs after neutron irradiation to 3.5×10^{25} n/m^2 ($E > 1$ MeV)

	Maximum stress (MPa)	Total elongation (%)	IGSCC (%)	TGSCC (%)
HP304	584	9.0	81.9	0
HP304/Si	710	7.0	91.2	0
HP304/C	860	5.2	31.0	64.5
HP316	622	6.8	67.0	17.9

TGSCC = Transgranular stress corrosion cracking.

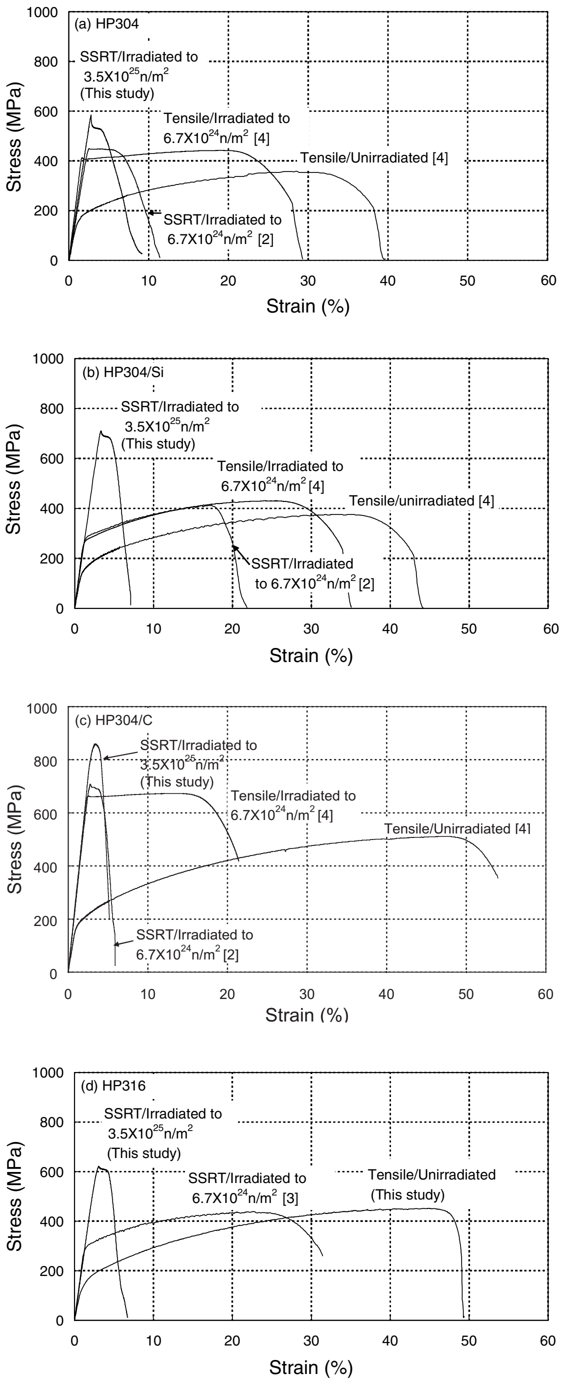

Fig. 1. Stress–strain curves of specimens after SSRTs in oxygenated high purity water at 561 K: (a) HP304, (b) HP304/Si, (c) HP304/C, and (d) HP316 [2–4].

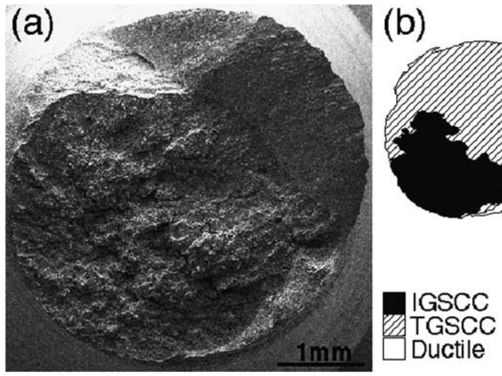

Fig. 2. SEM microphotograph of HP304/C after SSRT in oxygenated high purity water at 561 K: (a) is the fractured surface and (b) is schematic illustration of (a).

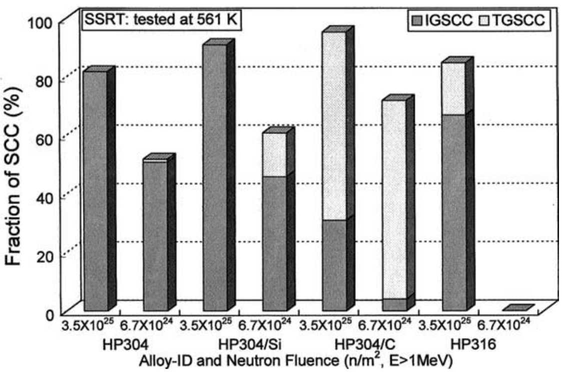

Fig. 3. Fraction of SCC at fluences of 3.5×10^{25} and 6.7×10^{24} n/m^2 [2–4].

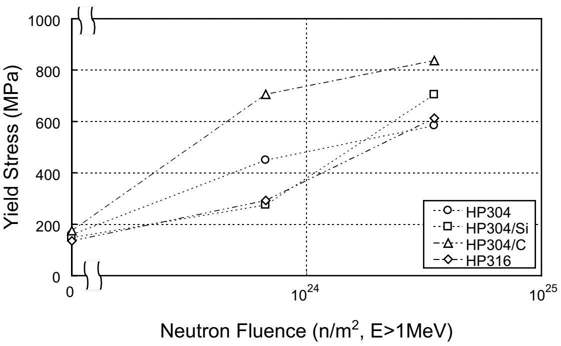

Fig. 4. Fluence dependence of yield stress for the high purity SSs.

peared to be generally more susceptible to IGSCC. The susceptibility to IGSCC increased with increasing yield stress due to irradiation hardening. Bruemmer and Simonen [13] reported that IGSCC correlated better to yield strength than to fluence. Fukuya et al. [10] indicated that harder materials were more susceptible to IGSCC. At 3.5×10^{25} n/m^2, HP304 showed IGSCC

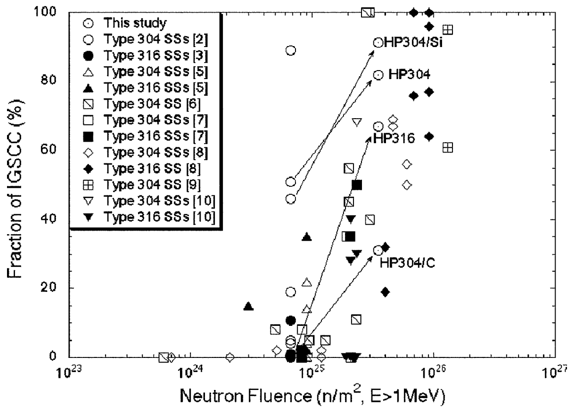

Fig. 5. Fluence dependence of the fraction of IGSCC in type 304 and 316 SSs [2,3,5–10].

alone, while HP304/C showed IGSCC with TGSCC as shown in Fig. 4. The yield stress of HP304/C was the largest in those of all the SSs due to irradiation hardening, and the susceptibility to IGSCC of HP304/C SS was the smallest in those of all the SSs in this study. Thus, it appears that effect of C was to inhibit IGSCC by changing the failure mode from IGSCC to TGSCC.

3.3. *Effect of molybdenum*

In HP316, as the neutron fluence increased to 3.5×10^{25} n/m^2, the %IGSCC increased to more than 60% (see Fig. 3). The %IGSCC of HP316 which was 0% at a fluence of 6.7×10^{24} n/m^2 remarkably increased to 3.5×10^{25} n/m^2. Addition of Mo suppressed the formation of Frank loops and delayed irradiation hardening [3,4]. Since the yield stress of irradiated HP316 increased (see Fig. 4), it appeared that the beneficial effect of Mo almost disappeared as the neutron fluence increased.

Although the susceptibility to IGSCC of HP304 and HP304/Si was higher than that of other type 304 SSs, the susceptibility to IGSCC of HP304/C was smaller than that of other type 304 SSs. This suggests that effect of Si disappeared and that higher C content had a beneficial effect of suppressing the susceptibility to IGSCC from Fig. 5.

4. Conclusions

To study the effects of the addition of minor elements in high purity type 304 and 316 SSs on susceptibility to SCC, SSRTs were conducted in oxygenated high purity water at 561 K after irradiation to 3.5×10^{25} n/m^2. The fraction of SCC on the fractured surface of the specimens was examined using SEM. From these experiments, the following conclusions were drawn:

1. Yield stress in SSRTs and %IGSCC increased with increasing neutron fluence from 6.7×10^{24} to 3.5×10^{25} n/m^2 for all high purity SSs.
2. Suppression of irradiation hardening and an increase in the time to SCC fracture which were beneficial effects of Si or Mo, were not obviously observed at a neutron fluence of 3.5×10^{25} n/m^2.
3. Addition of C suppressed the susceptibility to IGSCC at a neutron fluence of 3.5×10^{25} n/m^2. %IGSCC in HP304/C was the smallest of all high purity SSs and the irradiation hardening level was the largest of all high purity SSs in this study.

Acknowledgements

The authors would like to thank the JAERI staff of the Tokai WASTEF and the Oarai hot laboratory, especially to S. Endo, N. Iizumi, F. Takada and Y. Kato for their extensive post-irradiation examinations.

References

[1] ITER-EDA, Final Design Report, Materials Assessment Report, 1.1-13, 1999.
[2] T. Tsukada, Y. Miwa, H. Nakajima, in: Proceedings of 7th International Symposium on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, NACE, International, Houston, TX, 1995, p. 1009.
[3] T. Tsukada, Y. Miwa, H. Nakajima, T. Kondo, in: Proceedings of 8th International Symposium on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, ANS, 1997, p. 795.
[4] T. Tsukada, JAERI-Research 98-007, JAERI, 1998 (in Japanese).
[5] H.M. Chung, W.E. Ruther, R.V. Strain, W.J. Shack, NUREG/CR-6687, ANL-00/21 (ANL, 2000).
[6] W.L. Clarke, A.J. Jacobs, in: Proceedings of 1st International Symposium on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, NACE, International, Houston, TX, 1983, p. 451.
[7] A.J. Jacobs, G.P. Wozadlo, K. Nakata, T. Yoshida, I. Masaoka, in: G.J. Theus, J.R. Weeks (Eds.), Proceedings of 3rd International Symposium on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, The Metallurgical Society, Warrendale, PA, 1988, p. 673.
[8] M. Kodama, S. Nishimura, J. Morisawa, S. Suzuki, S. Shima, M. Yamamoto, in: D. Cubicciotti, E.P. Simonen, R. Gold (Eds.), Proceedings of 5th International Symposium on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, American Nuclear Society, La Grange Park, IL, 1992, p. 948.
[9] M. Kodama, R. Katsura, J. Morisawa, S. Nishimura, S. Suzuki, K. Asano, K. Fukuya, K. Nakata, in: R.E. Gold, E.P. Simonen (Eds.), Proceedings of 6th International Symposium on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, The Minerals,

Metals, and Materials Society, Warrendale, PA, 1993, p. 583.
[10] K. Fukuya, S. Shima, K. Nakata, S. Kasahara, A.J. Jacobs, G.P. Wozadlo, S. Suzuki, M. Kitamura, in: R.E. Gold, E.P. Simonen (Eds.), Proceedings of 6th International Symposium on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, The Minerals, Metals, and Materials Society, Warrendale, PA, 1993, p. 5856.
[11] T. Miwa, T. Tsukada, S. Jitsukawa, S. Kita, S. Hamada, Y. Matsui, M. Shindo, J. Nucl. Mater. 233 (1996) 1393.
[12] H.M. Chung, W.E. Ruther, J.E. Sanecki, A. Hins, N.J. Zaluzec, T.F. Kassner, J. Nucl. Mater. 239 (1996) 61.
[13] S.M. Bruemmer, E.P. Simonen, Corros. Sci. 50 (1994) 940.