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Journal of Nuclear Materials 274 (1999) 341–344

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Letter to the Editors

An Auger electron spectroscope analysis of thermally-sensitized type 304 stainless steels irradiated to low neutron fluences

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Received 2 March 1999; accepted 23 April 1999

Abstract

An Auger electron spectroscopy was used to examine grain boundary chromium depletion of the thermally-sensitized type 304 stainless steel (SS) irradiated to low neutron fluences. The chromium depletion was greater for 3×10^{23} n/m^2 ($E > 1$ MeV) and moreover for 1×10^{24} n/m^2 than that of the unirradiated SS. It is suggested that the thermally-sensitized SS is prone to be more sensitive to radiation induced segregation than the non-sensitized austenitic SS.

PACS: 61.82.Bg; 81.40.Gh; 61.80.Hg; 82.80.Pv

1. Introduction

Intergranular stress corrosion cracking (IGSCC) of the thermally-sensitized austenitic stainless steel (SS) is an important degradation mechanism for the component materials of light water reactor (LWR) [1]. It is known that the susceptibility to IGSCC is closely related to the grain boundary chromium concentration of the sensitized materials [2], because precipitation of chromium carbides at grain boundaries during welding results in the formation of the chromium depletion. Since in some cases IGSCC incidents happen in the irradiation environment of the systems, we need to determine to what extent neutron irradiation affects IGSCC performances. In this respect a series of IGSCC and mechanical property studies on the irradiated thermally-sensitized type 304 SSs have been conducted by Hide and his co-workers [3–5], who showed that the IGSCC susceptibility tended to increase with neutron fluences in the range of up to 1.1×10^{24} n/m^2 ($E > 1$ MeV) and mechanical factors played a role without evidencing any irradiation-induced changes in chromium depletion at the grain boundary. However, no grain boundary chemistry data of the neutron irradiated thermally-sensitized austenitic SSs have been reported yet.

Concerning analytical techniques, field emission gun scanning transmission electron microscopy (FEG-STEM) and Auger electron spectroscopy (AES) have been most commonly used for studying grain boundary microchemistry, although they have disadvantages as well as advantages [6]. In this work we used an AES technique to examine grain boundary chromium concentrations of thermally-sensitized SSs irradiated to low neutron fluences.

Two different heats of thermally-sensitized type 304 SSs, designated as Materials X and V, were used. Conditions of sensitization heat treatment were that the solution-annealed materials were heat-treated for 100 min at 750°C, followed by aging for 24 h at 500°C and then by air cooling. The purpose of this two-step aging treatment was to provide the specimens with hypothesized worst-case sensitization conditions. Table 1 shows the chemical compositions of the two different materials. It is seen that the compositions of the two materials are almost equal to each other and the contents of alloying elements such as chromium, nickel and iron were vir-

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0022-3115/99/$ – see front matter
PII: S0022-3115(99)00118-X

Table 1
Chemical compositions of the test specimen (wt%)

	C	Si	Mn	P	S	Ni	Cr	Co	N	Fe
Material X	0.063	0.49	0.95	0.026	0.016	9.93	18.49	<0.01	0.032	Bal.
Material V	0.060	0.49	0.98	0.028	0.016	9.92	18.39	0.01	0.046	Bal.

tually identical. The chromium contents were approximately 18.5 and 18.4 wt% for Materials X and V, respectively.

Material X was irradiated to a neutron fluence of 3×10^{23} n/m^2 at a typical BWR temperature of 290°C and Material V was to 1×10^{24} n/m^2 at a representative PWR temperature of 340°C in the Japan Material Test Reactor. The Auger electron spectroscope which we used in this work was named as shielded scanning Auger microscope (SAM) in Argonne National Laboratory (ANL). A description of the shielded SAM and a procedure of the analyses were given elsewhere [7]. The AES analyses of intergranular fracture surfaces were made at selected positions away from precipitated chromium carbides to minimize the influence of the carbides.

Fig. 1 shows typical AES concentration profiles of major alloying elements in Material X unirradiated and irradiated to 3×10^{23} n/m^2 at 290°C. It is seen that the chromium content at a grain boundary of the unirradiated material is markedly depleted and the nickel content is enriched due to the thermal sensitization heat treatments. In the irradiated material the chromium concentration is further depleted and the nickel composition is enriched. No influence of chromium carbides is appreciably identified on the profiles.

Fig. 2 represents also typical AES composition profiles in Material V unirradiated and irradiated to 1×10^{24} n/m^2 at 340°C. Apparently the difference in chromium depletion and nickel enrichment at grain boundaries between unirradiated and irradiated materials became larger at this fluence. Again no influence of chromium carbides is discernible in the AES profiles.

Fig. 3 plots the AES chromium contents of Materials X and V against neutron fluence. It is found that the reduction in grain boundary chromium concentration of Material V irradiated to 1×10^{24} n/m^2 is much greater than that for Material X irradiated to 3×10^{23} n/m^2. We assume that the larger decrease in the AES chromium concentration of the Material V primarily be brought about by the higher neutron fluence. However, it is uncertain at this moment whether or not the 50°C higher irradiation temperature at 1×10^{24} n/m^2 affected the larger reduction in AES chromium concentration.

Literature data [8–11] of grain boundary chromium contents of the non-sensitized SSs irradiated in BWR and in PWR by FEGSTEM and AES were also included in Fig. 3 as a function of neutron fluence. Apparently the FEGSTEM data are in accord with each other regardless of BWR and PWR irradiation. In addition, a theoretical calculation of radiation-induced segregation was also made [12], indicating no explicit effect of 50°C difference in irradiation temperature on the grain boundary chromium depletion of irradiated solution annealed and thermally-sensitized SSs. It is inferred from these results that the higher irradiation tempera-

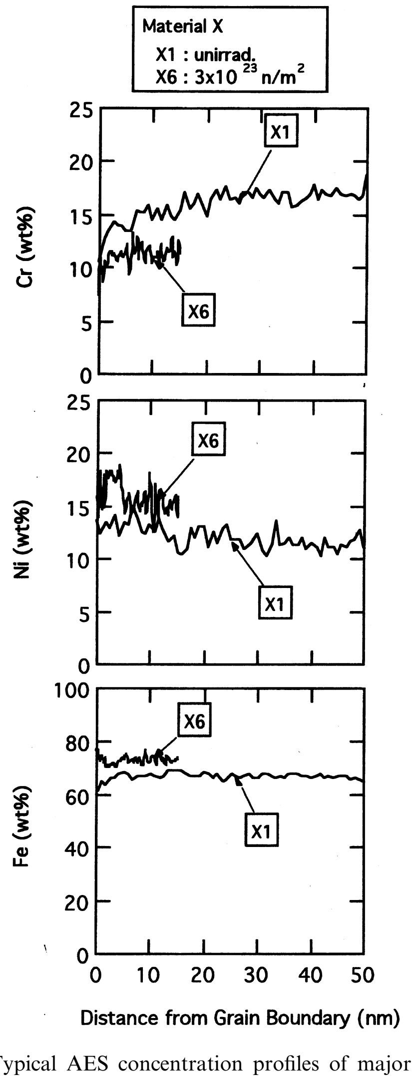

Fig. 1. Typical AES concentration profiles of major alloying elements for Material X unirradiated and irradiated to 3×10^{23} n/m^2.

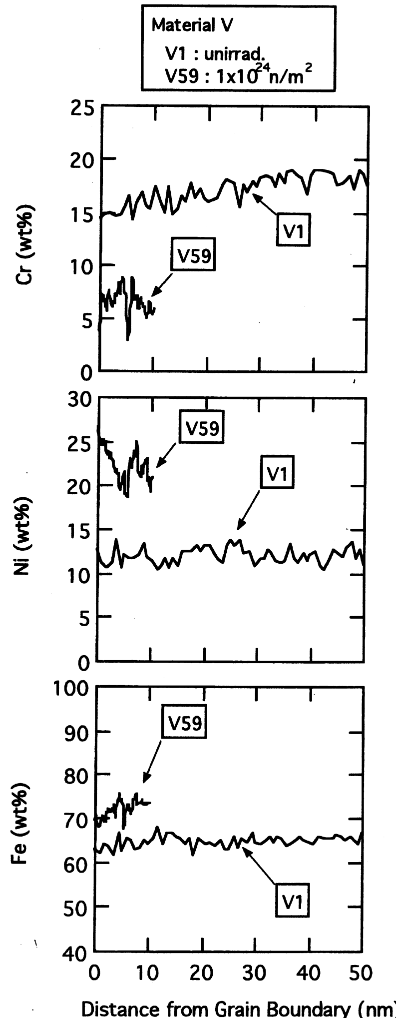

Fig. 2. Typical AES concentration profiles of major alloying elements for Material V unirradiated and irradiated to 1×10^{24} n/m^2.

ture by 50°C at 1×10^{24} n/m^2 than at 3×10^{23} n/m^2 in this work would not affect the grain boundary chromium depletion.

The AES and FEGSTEM data at $\sim 10^{25}$ and $\sim 10^{26}$ n/m^2 in Fig. 3 were acquired from the same non-sensitized SSs irradiated in BWR. No significantly large difference in grain boundary chromium concentrations is found at each fluence between AES and FEGSTEM, although the AES data are slightly smaller than those for the FEGSTEM analysis. From the fact that the amount of chromium depletion in the thermally-sensitized SS irradiated to 1×10^{24} n/m^2 in the present work is comparable to that for the non-sensitized austenitic SSs irradiated to 1×10^{26} n/m^2, as seen in Fig. 3, it is suggested that the thermally-sensitized SSs is prone to be

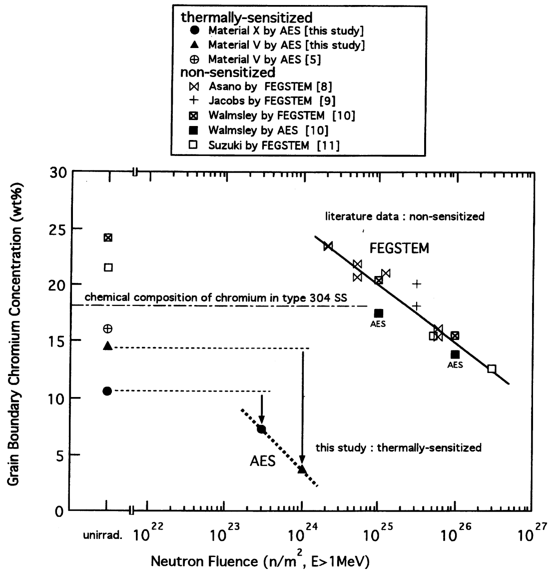

Fig. 3. Grain boundary chromium concentrations against neutron fluence. The AES data of the thermally-sensitized type 304 SSs from the present work, and the FEGSTEM data of non-sensitized austenitic SSs from [8–11].

more sensitive to radiation induced segregation than the non-sensitized austenitic SSs.

More grain boundary chemistry data by AES as well as by FEGSTEM are required to discuss sensitivity of thermally-sensitized SSs to radiation induced segregation in a quantitative manner.

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