Journal of Nuclear Materials 443 (2013) 378–385

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Concomitant formation of different nature clusters and hardening in reactor pressure vessel steels irradiated by heavy ions

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ARTICLE INFO

Article history:
Received 4 September 2012
Accepted 26 July 2013
Available online 2 August 2013

ABSTRACT

Specimens of A533B steels containing 0.04, 0.09 and 0.21 wt%Cu were irradiated at 290 °C to 3 dpa with 3 MeV Fe ions and subjected to atom probe analyses, transmission electron microscopy observations and hardness measurements. The atom probe analysis results showed that two types of solute clusters were formed: Cu-enriched clusters containing Mn, Ni and Si atoms as irradiation-enhanced solute atom clusters and Mn/Ni/Si-enriched clusters as irradiation-induced solute atom clusters. Both cluster types occurred in the highest Cu-content steel and the ratio of Mn/Ni/Si-enriched clusters to Cu-enriched clusters increased with irradiation doses. It was confirmed that the cluster formation was a key factor in the microstructure evolution until the high dose irradiation was reached even in the low Cu content steels though the dislocation loops with much lower density than that of the clusters were observed as matrix damage. The difference in the hardening efficiency due to the difference in the nature of the clusters was small. The irradiation-induced clustering of undersized Si atoms suggested that a clustering driving force other than vacancy-driven diffusion, probably an interstitial mechanism, may become important at higher dose rates.

1. Introduction

Radiation embrittlement of reactor pressure vessel (RPV) steels is known to be sensitive to various material variables and irradiation conditions. Understanding details of the microstructural evolution in RPV steels under irradiation is indispensable to predict and to evaluate radiation embrittlement. Many investigations have been carried out to understand the effects of material variables and irradiation conditions on the microstructural evolutions. Several types of microstructural features have been proposed which commonly belong to two groups: solute atom clusters and matrix damage [1–3]. Cu-rich precipitates or Cu-enriched solute atom clusters are the main contributors to hardening-induced embrittlement in Cu-containing steels. Radiation-induced defects other than Cu-enriched solute atom clusters, so-called matrix damage, provide an additional contribution to embrittlement in high Cu-content steels and become a main contributor to embrittlement in low Cu-content steels. The other concern is the so-called "late blooming phase," which consists of Ni and Mn, without Cu [4]. This phase is expected to form in low Cu-content, high Ni-content steels irradiated to high doses. The formation of Cu-free or very low Cu solute atom clusters consisting of Mn, Ni and Si atoms in very low Cu-content steels and interstitial dislocation loops has been confirmed from low doses [5–15].

Providing the recent understanding of embrittlement mechanisms, mechanism-guided embrittlement correlation methods have been developed [16–18]. The modern method presented by Soneda et al. [18] adopted an approach to predict the microstructural changes in materials during irradiation using a set of rate equations describing the time evolution of the number densities of irradiated-induced and irradiated-enhanced solute atom clusters and matrix damage. In this model, the physical process of irradiation-induced clustering was assumed to be segregation of solute atoms to matrix damage features by irradiation. This assumption may be very useful for the modeling of the experimental observation of solute atom clusters with little or no Cu atoms in low Cu-content steels irradiated at low flux of light water reactors [18] and at high flux of material test reactors [5,8,9]. However, few experimental studies have been carried out on the role of matrix damage features as the nucleation site of solute atom clusters, since the nature of matrix damage is not identified excluding dislocation loops. In regard to the formation of different population of solute atom clusters, which are correlated with irradiation-induced and irradiation-enhanced clusters, not only Cu-enriched clusters with Cu centers but also clusters without Cu atoms have been observed in Cu-containing A533B steels irradiated by heavy ions at high doses [19]. In addition, some of the solute atom clusters in low-Cu surveillance materials have been found to include

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0022-3115/$ - see front matter
http://dx.doi.org/10.1016/j.jnucmat.2013.07.056

no Cu atoms [18]. However, quantitative analyses of the distinguished clusters have not been carried out. For model alloys, two groups of solute atom clusters have been distinguished in Fe–0.09Cu–1.1Mn–0.7Ni (at.%) alloy irradiated in the BR2 test reactor [11]. The former was mainly enriched in Cu atoms, and the latter was enriched in Mn and Ni atoms or in Mn atoms only and detected at higher fluences. However, experimental studies on the concomitant formation of different nature clusters are limited. For better understanding the role of matrix damage as a cluster nucleus and the concomitant formation of different nature clusters, more experimental studies with high resolution techniques such as atom probe tomography (APT) and transmission electron microscopy (TEM) are needed.

Irradiation with energetic particles is widely used to study fundamental processes in microstructural and microchemical evolution in materials under irradiation. However, the dose rate of heavy ion irradiation is usually much higher than that of neutron irradiation. The effects of high dose rate on evolution of solute atom clusters and dislocation loops have been investigated by the comparison of microstructure in RPV steels under neutron and heavy ion irradiations [19,20]. The formation of the same nature clusters and dislocation loops have been reported under both irradiations. However, it has been presented that the number density and diameter of clusters in the material irradiated by heavy ions were higher and smaller, respectively, than those in the neutron-irradiated material. Therefore, ten times dose or more of heavy ion irradiation was necessary to change the same cluster volume fraction and hardness as neutron irradiation. These results indicate that high dose and high flux ion irradiation is the only way to model neutron irradiations, with differences but to understand basic mechanism it is a useful tool.

In this study, APT and TEM observations were made to examine the continuousness of the microstructural evolution in RPV A533B steels irradiated to high doses by high energy heavy ions. Solute atom clusters of different natures were separated by a devised analytical method for cluster identification, and the concomitant formation of the solute atom clusters of different natures and the dose dependence of the ratio of those clusters were demonstrated.

2. Experimental procedures

2.1. Materials and irradiation conditions

The materials used in this study were commercial A533B steels. The chemical compositions are shown in Table 1 (hereafter, all weight percentages of Cu are shown simply as %). The Cu concentrations of the steels ranged from a low level to a medium level within RPV steels used in Japan. Heavy ion irradiations were conducted with 3 MeV Fe ions using the tandem ion accelerator in the High Fluence Irradiation Facility of the University of Tokyo. The irradiation temperature was 290 ± 5 °C. The SRIM-2006 code was used for an irradiation damage calculation [21]. The corresponding dose calculation was done in pure Fe with the displacement energyE_d = 40 eV. The peak damage depth was 760 nm. The dose and dose rate at a depth of 300 nm were used for damage parameters, since post-irradiation microanalyses were performed at that depth. The applied dose rate was 1.0×10^{-4} dpa/s, and the doses were 0.25, 0.5, 1, 2 and 3 dpa.

Table 1
Chemical composition of steels (wt%).

	C	Si	Mn	P	S	Ni	Cr	Mo	Cu	Fe
0.04%Cu steel	0.19	0.22	1.50	0.003	0.006	0.62	0.11	0.51	0.04	Balance
0.09%Cu steel	0.19	0.26	1.38	0.007	0.008	0.62	–	0.48	0.09	Balance
0.21%Cu steel	0.24	0.17	1.55	0.009	0.018	0.63	0.14	0.52	0.21	Balance

2.2. Measurements

Hardness at room temperature was measured using an ultra-micro hardness tester (Elionix ENT1100) with a Berkovich diamond indenter tip. In the present study, the indentation depth was kept at 150 nm, thus average hardness in the damaged region up to about the damage peak depth was measured. The hardness values were calculated from the maximum load and maximum displacement. The average hardness was determined by averaging over more than 40 indents.

Needle-shaped specimens for 3DAP analyses were prepared using a focused ion beam (FIB) system (Hitachi FB-2000A). The probe was prepared parallel to the irradiation surface and the probe tip was set at the depth of 300 nm. 3DAP analyses were carried out at a specimen temperature of 50 K using a voltage-pulse mode of an atom probe system (CAMECA LEAP3000XSi). Although Ga atoms which were used in FIB micro-processing were detected in the measured atom probe data, the count of Ga atoms was similar to that of background noise, which was negligibly low. No clustering induced by the FIB micro-processing of the 3DAP specimen preparation was confirmed in unirradiated material measurements.

Thin foils for TEM observation were prepared so that the foil was ~300 nm from the irradiated surface using the same Hitachi FB-2000A FIB system. To remove defects appeared under Ga implantation at the FIB preparation, the specimens were finished by low-energy ion sputtering of Ar ions accelerated at 150 V using an ultralow-energy Ar ion beam sputtering system (Technorg-Linda GentleMill IV). Observations were carried out using a TEM (Hitachi HF-3000) equipped with a field emission gun of 300 kV. Analysis of radiation-induced defects was carried out at a depth of 300 nm in the specimens using a weak-beam technique; details have been described previously [6].

3. Results

3.1. Hardness measurements

Fig. 1 shows dose dependence of radiation-induced hardening calculated by subtracting average hardness of unirradiated material from that of the irradiated one. The standard deviation of average hardness was less than 50 for over forty measurements. Hardness increased with increasing dose in all steels. The

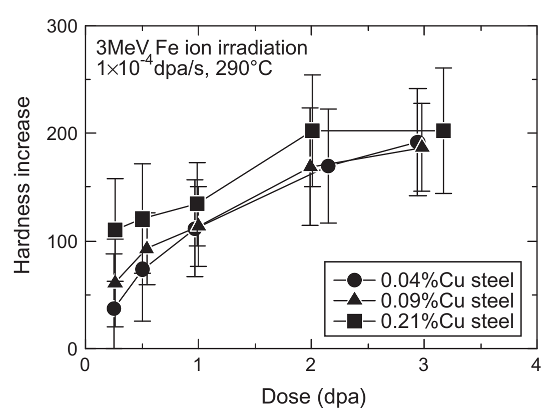

Fig. 1. Dose dependence of irradiation-induced hardening.