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Journal of Nuclear Materials 307–311 (2002) 308–311

journal of nuclear materials

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Evaluation of radiation hardening in Fe alloys under heavy ion irradiation by micro-indentation technique

Naoto Sekimura *, Toru Kamada, Yohei Wakasugi, Taira Okita, Yoshio Arai

Department of Quantum Engineering and Systems Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

Abstract

To correlate micro-structural evolution near the surface under ion irradiation with macroscopic mechanical properties, micro-indentation tests were applied to heavy ion-irradiated model alloys of ferritic steels. The derivative of the load–displacement ratio with respect to the displacement, $d(L/D)/dD$, was estimated to evaluate the depth dependent formation of hardening features in the model alloys irradiated with 12 MeV Ni ions at 300 °C between 8 and 25 dpa. The depth distribution of radiation-enhanced formation of Cu precipitate in Fe–0.3Cu was found to change with dose, because of over-aging of the precipitates near the peak damage depth. Hardening near the peak damage depth region in Fe–0.3Cu–0.7Ni alloy increases with dose, indicating that the addition of Ni suppresses the growth of Cu-rich precipitates.

1. Introduction

Heavy ion irradiation has been widely used to study micro-structural sensitivities in materials on irradiation parameters such as dose, dose rate and temperature. Multiple beam irradiation techniques have also been extensively applied to study synergistic effects of displacement damage and transmutation atoms under fusion irradiation conditions [1]. A low loads, the micro-Vickers hardness test is one of the most useful methods to correlate micro-structural evolution near the surface under ion irradiation with macroscopic mechanical properties [2–5]. However, it is important to consider depth distributions of micro-structural changes and implanted ions to apply the obtained detailed information to bulk irradiation effects under neutron irradiation.

In ferritic steels containing Cu impurities, radiation-enhanced formation of Cu precipitates is the most important hardening mechanism in addition to the formation of defect clusters [6,7]. Because Cu atoms are known to diffuse only with vacancies, the hardening behavior of Fe–Cu alloys irradiated with heavy ions can be strongly affected by diffusion of vacancies to the sinks such as grain boundaries and surfaces. Ni addition to these alloys is expected to change the precipitation behavior of Cu-rich clusters. In this study, a micro-indentation technique is applied to understand depth-dependent micro-structural evolution up to high dose in ferritic model alloys under heavy ion irradiation.

2. Experimental

Materials used in this study are 99.9% pure Fe, Fe–0.3Cu and Fe–0.3Cu–0.7Ni alloys with minor interstitial impurities of 180 and 80 wt ppm O. These alloys were rolled to sheets of 0.2 mm in thickness from which 3 mm disks were punched-out and annealed at 950 °C for 1 h and 850 °C for 2 h followed by furnace-cooling.

These model alloys were irradiated with 12 MeV Ni^{3+} ions in the TIARA (Takasaki Ion Accelerators for Advanced Radiation Application) facility in JAERI (Japan Atomic Energy Research Institute). The peak displacement damage depth by 12 MeV Ni^{3+} ions in Fe was estimated to be 2.1 μm from the specimen surface by the TRIM code. Maximum irradiation dose at this damage

* Corresponding author. Tel.: +81-3 5841 6986; fax: +81-3 5841 8614.

E-mail address: sekimura@q.t.u-tokyo.ac.jp (N. Sekimura).

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PII: S0022-3115(02)01079-6

peak depth was about 25 dpa. Irradiation temperature was 300 °C, and the ion flux was fixed at 1.0×10^{-3} dpa/s, which corresponds to the ion flux of 1.21×10^{16} ions/m^2/s.

Micro-indentation tests were performed on a Shimazu DUH-201S. In this study, the indentation load was not fixed, but the indentation depth was fixed at 1.7 μm. A slower loading rate is required compared to normal micro-Vickers tests to evaluate depth dependent radiation hardening by heavy ions. Uncertainty in the indenter displacement increases at larger load speed especially at the beginning stage of its contact on the surface of the specimen, while vibration of the indentation machine can affect load–displacement results at lower speed. In this study, the load speed was set at 0.27 gf/s. The deformed regions were carefully chosen to avoid any effects of grain boundaries.

3. Results and discussion

Fig. 1(a) shows a typical load (L)–displacement or depth (D) curve in Fe–0.3Cu specimens irradiated with 12 MeV Ni^{3+} ions at 300 °C to 9.0 dpa at the peak depth. Depth dependence of the load–displacement ratio, L/D, is also shown in Fig. 1(b). Vicker's hardness increases from 109 to 147 for this irradiation condition. As shown in Fig. 1(b), irradiation hardening by 12 MeV Ni^{3+} ions is most clearly observed for the initial part of the $L/D(D)$ curve up to about 0.4–0.5 μm.

The correlation between indentation load and depth during indentation in uniform specimens can be expressed as follows [8];

$$L = AD + (B\mathrm{p} + B\mathrm{e})D^2, \tag{1}$$

where A is a constant depending on the shape of indenter, and Bp and Be can be taken as characterizing plastic and elastic contributions to deformation, respectively. It should be noted that Bp depends on the depth in the ion-irradiated samples with non-uniform formation and evolution of defects.

L/D, can be written by the following equation,

$$L/D = A + (B\mathrm{p} + B\mathrm{e})D. \tag{2}$$

The parameter A in Eq. (2) can be estimated by extrapolating the L/D vs. D to $D = 0$. No effect of irradiation on this parameter is detected for all the tested materials as the ratio of load–depth. All the estimated values of the A were found to fall within 10% of one another in all the tested samples, when load–displacement data up to 150 nm is neglected.

Fig. 2 shows the indentation depth dependence of $\mathrm{d}(L/D)/\mathrm{d}D$, which is the derivative of the load–depth ratio with respect to the depth. The depth distribution of the displacement damage estimated by the TRIM code is also indicated in this figure. The peak in $\mathrm{d}(L/D)/\mathrm{d}D$ is clearly observed at an indenter depth between 200 and 250 nm, which is (1/10)–(1/8) of the calculated displacement damage peak depth which exists at 2.1 μm from the surface.

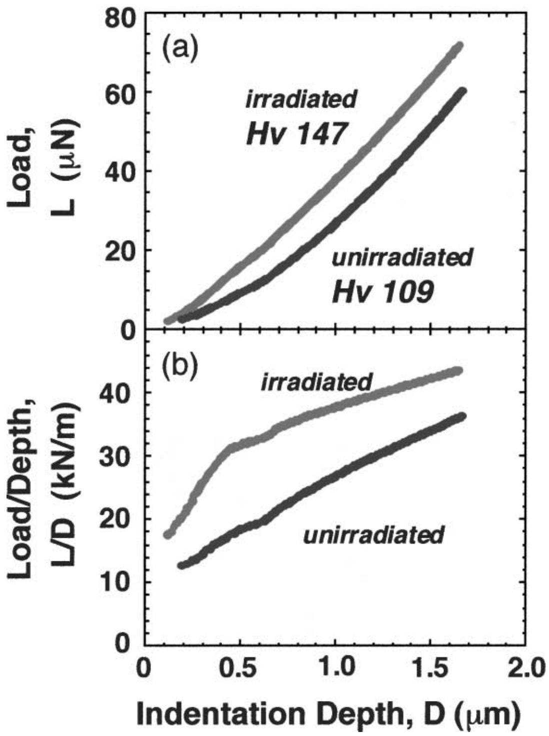

Fig. 1. (a) Load (L) vs. indentation depth (D) and (b) L/D vs. D curves in an Fe–0.3Cu alloy both un-irradiated and irradiated with 12 MeV Ni^{3+} ions at 300 °C to 9.0 dpa.

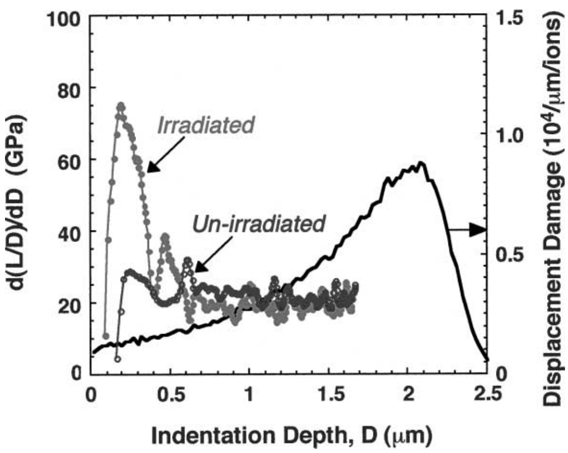

Fig. 2. Derivative value of (L/D) as a function of indentation depth (D) in an Fe–0.3Cu alloy both un-irradiated and irradiated with 12 MeV Ni^{3+} ions at 300 °C to 9.0 dpa. The calculated depth distribution of displacement damage by the TRIM code is also indicated.

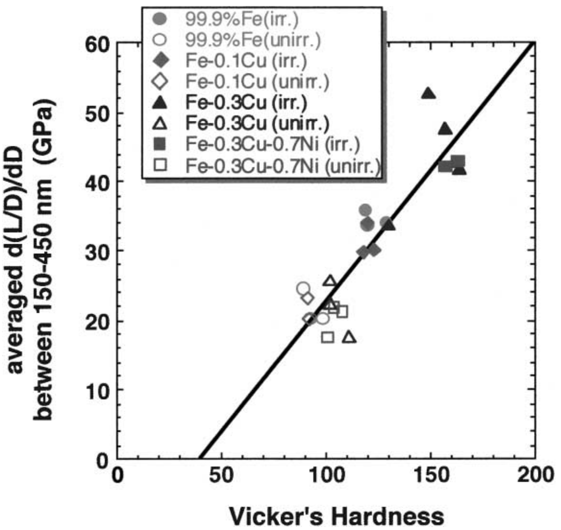

Fig. 3. Correlation between Vicker's hardness by the fixed indentation depth at 1.7 μm and micro-indentation data in unirradiated and irradiated specimens.

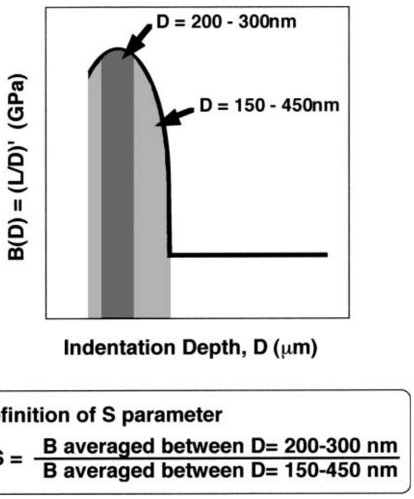

Fig. 4. Schematic illustration of the definition of the S parameter to measure the depth distribution of hardening species.

We found that the value of $\mathrm{d}(L/D)/\mathrm{d}D$ can be a good measure of the formation of hardening species which are dependent on depth distribution of displacement damage by heavy ion irradiation and resultant defect interactions. As evidence of this relationship between measured Vicker's hardness by the fixed indentation depth of 1.7 μm and this value between $D = 150$ and 450 nm is shown in Fig. 3. There is a good correlation between these data points; however, the trend line indicates that the effect of elastic deformation which is the offset at $\mathrm{d}(L/D)/\mathrm{d}D = 0$ should also be considered in estimating very low load indentation testing.

Deformation barriers formed around the peak damage depth region can be clearly detected at the indentation depth, D, of around 250 nm. In order to estimate the dose dependence of nucleation and growth kinetics of hardening features in ion-irradiated samples, we define an S value by the following equation,

$$S = \frac{\mathrm{d}(L/D)/\mathrm{d}D \text{ averaged between 200 and 300 nm}}{\mathrm{d}(L/D)/\mathrm{d}D \text{ averaged between150 and 450 nm}}. \tag{3}$$

This value measures the importance of formation of hardening species at the peak hardening depth region by 12 MeV Ni ion irradiation as schematically indicated by Fig. 4.

Fig. 5 shows the dose dependence of the S parameter in irradiated 99.9% Fe, Fe–0.3Cu and Fe–0.3Cu–0.7Ni alloys. In pure Fe, interstitial clusters are the major hardening species, which form by nucleation and growth. No clear dose dependence of the S parameter is observed, indicating that the depth distribution of the clusters does not change from 8 to 20 dpa. In the case of

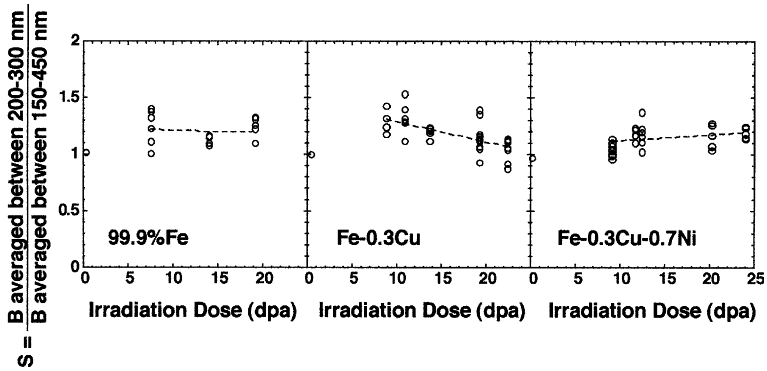

Fig. 5. Dose dependence of the S parameter in 99.9% Fe, Fe–0.3Cu and Fe–0.3Cu–0.7Ni irradiated with 12 MeV Ni^{3+} ions between 8 and 25 dpa at 300 °C.

Fe–0.3Cu, it is clearly observed that S decreases with dose. At low dose, Cu clusters are nucleated at the peak hardening depth region. Cu clusters are considered to grow larger under irradiation and coalesce around the peak displacement damage depth region at higher dose, which leads to the saturation or decrease of hardening. It has been clarified that dose dependence of ion-irradiation hardening in Fe–Cu alloys measured by micro-Vicker's machine shows radiation-enhanced over-aging around 10 dpa at 290 and 400 °C [9]. Therefore, the S value which measures hardening features around the damage peak depth region is not as large as those in Fe at 10 dpa or higher dose. Cu precipitates are considered to be nucleated also near the surface even at higher dose. This can explain the decrease of S parameter with dose in Fe–0.3Cu alloy.

In Fe–0.3Cu–0.7Ni ternary alloy, however, the S parameter seems to increase slightly with dose up to 25 dpa. This suggests that Cu-rich precipitates continuously nucleate around the damage peak depth region, while hardening in Fe–0.3Cu alloy saturates or even decreases at higher dose. Ni addition is considered to increase density of Cu-rich precipitates and to prevent them to grow larger.

4. Summary

The micro-indentation technique is found to be a very useful method to evaluate the response of materials under irradiation up to high dose. This is essentially a non-destructive method which does not prevent further micro-structural investigation by TEM. The differential value of load–displacement ratio, $\mathrm{d}(L/D)/\mathrm{d}D$, is estimated to evaluate the depth dependent formation of hardening features in ferritic model alloys. The depth distribution of the radiation-enhanced formation of Cu precipitates in Fe–Cu binary alloy is found to change with dose. This appears to be a result of over-aging of the precipitates near the peak hardening depth. Hardening near the peak hardening depth region in Fe–Cu–Ni ternary alloys increases with dose, indicating that the addition of Ni suppresses the growth of Cu-rich precipitates.

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