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The influence of helium co-implantation on ion-induced hardening of low activation ferritic steel evaluated by micro-indentation technique

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Abstract

An experimental technique to determine the dual-ion irradiation-induced hardening of solid materials by means of micro-indentation was developed and then applied to evaluate the irradiation response of low activation martensitic steel. Micro-hardness measurements were performed on a low activation Fe-9Cr-2W steel (JLF-1) which was obtained following bombardment of 4 MeV nickel ions up to 1 dpa with simultaneous deposition of helium ions at 0–100 appm He/dpa in a temperature range of 573–873 K. The obtained micro-hardness profiles corresponded to the expected hardness profiles. The increasing He/dpa ratio almost monotonically increased the micro-hardness at 673 K and higher temperatures. However, the irradiation-induced micro-hardness changes were no more than 10% except for very limited cases. Isothermal annealing at temperatures above 773 K caused softening within a thin surface layer of the specimens and only a small amount of helium prohibited the softening. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

Low activation ferritic/martensitic steels are, so far, the most promising materials for structural components of D–T fusion demonstrative devices due to their maturity as industrial materials as well as their superior irradiation resistance in physical and mechanical properties [1,2]. However, to ensure the feasibility and safety of fusion energy systems employing low activation ferritic steels, there still are several issues to be solved, which include the synergistic effects of fusion-relevant helium generation and atomic displacement damage on micro-structural stability and mechanical properties [2,3].

The combination of dual-beam charged particle irradiation using MV range accelerators and an ultra low-load indentation is a potential technique to evaluate the mechanical property changes due to the synergistic effects. In a typical dual-beam irradiation experiment in a fusion material study, the primary beam of heavy ions is used to introduce the atomic displacement damage and the additional beam is employed to inject helium ions to simulate the effect of transmutation through (n, α) reactions. An ultra low-load indentation technique is applicable to evaluate the indentation hardness of a surface layer as thin as a few hundred nanometers [4–6].

The objective of this series of studies is to develop an experimental technique to determine the hardness properties in dual-ion irradiated solid materials, which possess uneven hardness profiles within a surface layer of typically about 1 μm, by means of an ultra-low load indentation and then apply the developed technique to evaluate the irradiation-induced mechanical property changes of a low activation ferritic steel.

2. Experimental procedure

The material used was an Fe-9Cr-2W-V,Ta low activation ferritic steel (JLF-1), which had been developed through a fusion materials research activity at Japanese universities and the National Institute for Fusion Science

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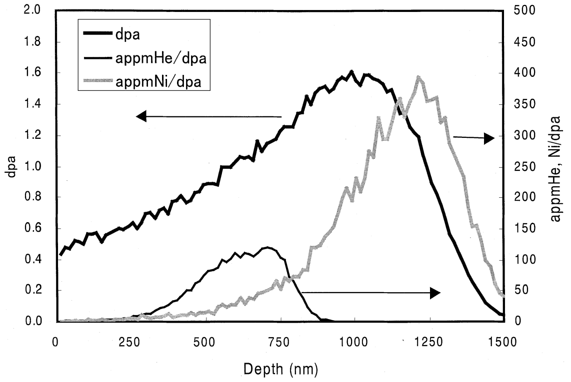

Fig. 1. The depth profile of displacement damage and deposited helium and nickel ions in the Fe-9Cr-2W steel under the 100 appmHe/dpa irradiation condition calculated by TRIM code. The average displacement threshold energy was assumed to be 40 eV.

(NIFS) and produced by Nippon Steel Corporation [6]. The chemical composition, thermo-mechanical treatment and fundamental properties were published elsewhere [7,8]. A 300 μm-thick sheet was sliced out of an as-normalized and tempered plate, and then punched into 3 mm diameter disks. The disk specimens were electrolytically finished just before the ion irradiation, following thickness reduction to approximately 250 μm by mechanical polishing.

The ion irradiation experiment was carried out at the High-fluence Irradiation Facility, University of Tokyo (HIT Facility) [9]. Two sets of specimens were bombarded with 4 MeV Ni^{3+} ions up to 1 dpa (at 550 nm from the irradiated surface), with or without co-implantation of energy-degraded 1 MeV He^+ ions, at 573, 673, 773 and 873 K at a displacement rate of 1×10^{-4} dpa/s. The helium-to-dpa ratio was either 0, 1, 10 or 100 appmHe/dpa. The profiles of displacement damage and deposited Ni and He ions were calculated with TRIM code, assuming a 40 eV of average displacement threshold energy [10], and are presented in Fig. 1. One set of irradiated specimens was examined with a JEOL JEM-2010 transmission electron microscope (TEM) following sectioning and back-thinning. Ultra-low load indentation testing was performed on another set of irradiated specimens, using a triangular pyramidal diamond indenter with a 68° semi-apex angle at a maximum load in the range of 0.98–98 mN, on an Akashi MZT-3 instrumented micro-indentation device.

3. Method to obtain micro-hardness profiles

Micro-hardness profiles were obtained by a contact pressure evaluation method based on the procedure proposed by Oliver, et al. [5], where hardness, H, is defined as

$$H = \frac{P}{A(h_c)}, \tag{1}$$

and P is the indentation load and A, as a function of contact depth (h_c), is the projected area of contact between the indenter and the specimen. In the current study, it was confirmed that sufficiently reproducible indentation hardness data could be obtained through this procedure for a fixed indentation depth as small as 50 nm. However, in this procedure, h_c was determined based on the load–displacement (P–h) property during the unloading process, and this prohibited the acquisition of a continuous hardness profile from a single indentation test. The displacement and helium deposition profiles shown in Fig. 1 may result in a fairly complicated hardness profile due to contributions by at least four kinds of layers, i.e., (1) the surface layer where the surface effect and the displacement effect dominate, (2) the second layer where the displacement and the helium deposition are effective, (3) the third layer where only the displacement influences, and (4) the substrate which is free from irradiation effects. To evaluate the hardness of the layer (2), continuous hardness profile acquisition was attempted in the following manner.

The P–h property during the unloading process is determined by a combination of (a) the effective elastic modulus of the material, (b) the contact depth h_c and (c) the projected area $A = F(h_c)$. Therefore, if the irradiation-induced changes in elastic properties are negligible, the unloading P–h curve for a particular material using an identical indenter can be expressed as a function of h_c alone; $P = F_{UL}(h_c, h)$. In this work, F_{UL} was expressed simply by a polynomial function fitted to unloading P–h curves obtained from an unirradiated specimen. It was confirmed that the unloading P–h curves obtained from irradiated specimens did not significantly deviate from F_{UL}. Therefore, the micro-hardness profile $H(h_c)$ is given as follows:

$$H(h_c) = \frac{P^*}{A(h_c)}. \tag{2}$$

In the above equation, P^* is the load at which the calibrated unloading function $P = F_{UL}(h_c, h)$ intersects the loading curve $P = F_L(h)$. In this series of studies, this method to obtain micro-hardness profiles was very effective to evaluate hardness changes in a variety of metallic materials induced by MeV range ion irradiation. A typical example of $H(h_c)$ obtained by a single indentation test on an unirradiated specimen after an electrolytical surface polishing is presented in Fig. 2. It shows that the micro-hardness obtained by this method is fairly constant at $h_c > 50$ nm in spite of a minor oscillation being observed when h_c is smaller than 200 nm.

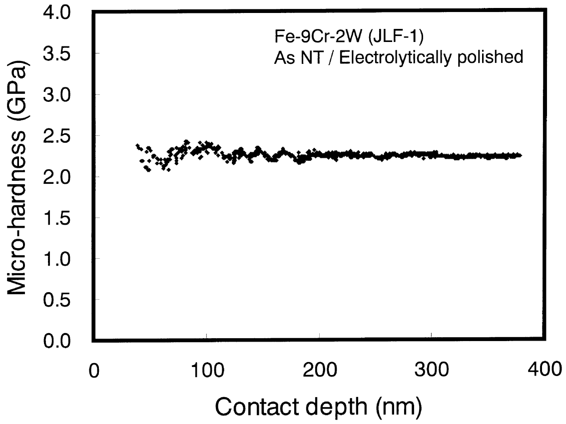

Fig. 2. An example of micro-hardness of the Fe-9Cr-2W martensitic steel (JLF-1) plotted against contact depth. The specimen was surface finished by electrolysis after the standard heat treatment.

4. Results and discussion

The micro-hardness for unirradiated specimens was 2.24 GPa on an average. This micro-hardness corresponds to a Vickers hardness number of 206, which agrees well with the Vickers hardness, 213–222, measured with the load of 98 N on the same and similar materials [8,11]. The pre-irradiation electrolysis removed the surface layer nominally by 10 μm for the 573 K-irradiated specimens, which resulted in a higher average value of 2.37 GPa in unirradiated condition, compared to the rest of the specimens with surfaces removed by 20 μm. The point-to-point data scatter range was approximately ±0.2 GPa, which was significantly larger than typical micro-hardness scatter in annealed austenitic alloys, probably due to the influences of inhomogeneous dislocation micro-structure, crystallographic orientation and anisotropic martensitic lath structures. In this work, therefore, at least five indentation measurements were made in distinct prior austenitic grains within a single specimen and the average was taken as the specimen's micro-hardness for a given h_c range. In addition, the amount of irradiation-induced hardening was determined by comparing the micro-hardness corresponding to the irradiated layer to that of the substrate within the same prior austenitic grain. The amount of scatter in the hardening was significant and so averaging was again necessary for a trend analysis.

Typical results of hardness measurement on ion irradiated specimens are provided in Fig. 3(a)–(d). Fig. 3(a) and (b) are the micro-hardness profiles $H(h_c)$ obtained from specimens which are single (0 appmHe/dpa)- and dual (100 appmHe/dpa)-ion irradiated, respectively, at 673 K. The plateau between approximately 50 and 200 nm of h_c in Fig. 3(a) corresponds to the irradiation-hardening layer of roughly 1 μm-thick. The single-ion irradiation, or displacement damage alone, caused only very minor hardening at this damage level. In the dual-ion irradiated specimen, a hardness profile consists of several stages instead of a distinct plateau. In Fig. 3(b), the flat part at approximately 90–130 nm of h_c corresponds to a layer hardened by dual-ion irradiation. The apparent rapid hardening toward the surface within $\sim$ 80 nm range has probably been caused by an uneven surface, since this kind of disturbances in $H(h_c)$ plots are observed frequently regardless of material classes. The hardening by displacement damage alone is also detectable in Fig. 3(b), in spite of disturbing influences from the dual-ion irradiation-hardening layer.

The micro-structural examination by TEM was an important part of this work. But it is not simple to determine the influences of irradiation condition on micro-structure because of its inhomogeneity. In specimens irradiated at 673 K, irradiation-produced dislocation loops were observed. The characteristics of dislocation loops were greatly affected by the pre-existing dislocations, which were spatially inhomogeneous even in a single martensitic lath as well as over the lath structures. However, as a general trend, it was confirmed that the number density of the dislocation loops increased and their mean size decreased with increasing helium concentration. Such a correlation between the helium injection rate and the dislocation loop characteristics was much weaker than that reported for austenitic stainless alloys irradiated with dual-ions at the same accelerator facility [12,13].

At higher temperatures, or above 773 K, isothermal annealing for 10^4 s caused significant softening which is limited to near surface regions. The average micro-hardness of thermal control specimens for the 773 and 873 K experiments were 1.97 and 1.87 GPa, respectively, at $h_c = 200$ nm. Single-ion irradiation at 873 K, as shown in Fig. 3(c), did not apparently cause hardening but rather suppressed the thermal softening [14]. Irradiation-induced or -assisted softening was not observed at 873 K at a displacement rate of 1×10^{-4} dpa/s. The micro-structural examination of the single-ion irradiated specimen exhibited dislocation network structures which were less inhomogeneous than the typical pre-existing dislocation structures. This observation suggests that the reactions between pre-existing and irradiation-induced dislocations might generate a dislocation network through irradiation-enhanced dislocation migration and the formation of network might work against a quick recovery of the pre-existing dislocation structures.

At 873 K, dual-ion irradiation resulted in significant hardening, as represented by Fig. 3(d). The amount of hardening, or the step height that appeared in the micro-hardness profiles, varied more than ±50%. Micro-structural examination revealed that the specimen irradiated at 100 appmHe/dpa contained a ⟨1 0 0⟩ type

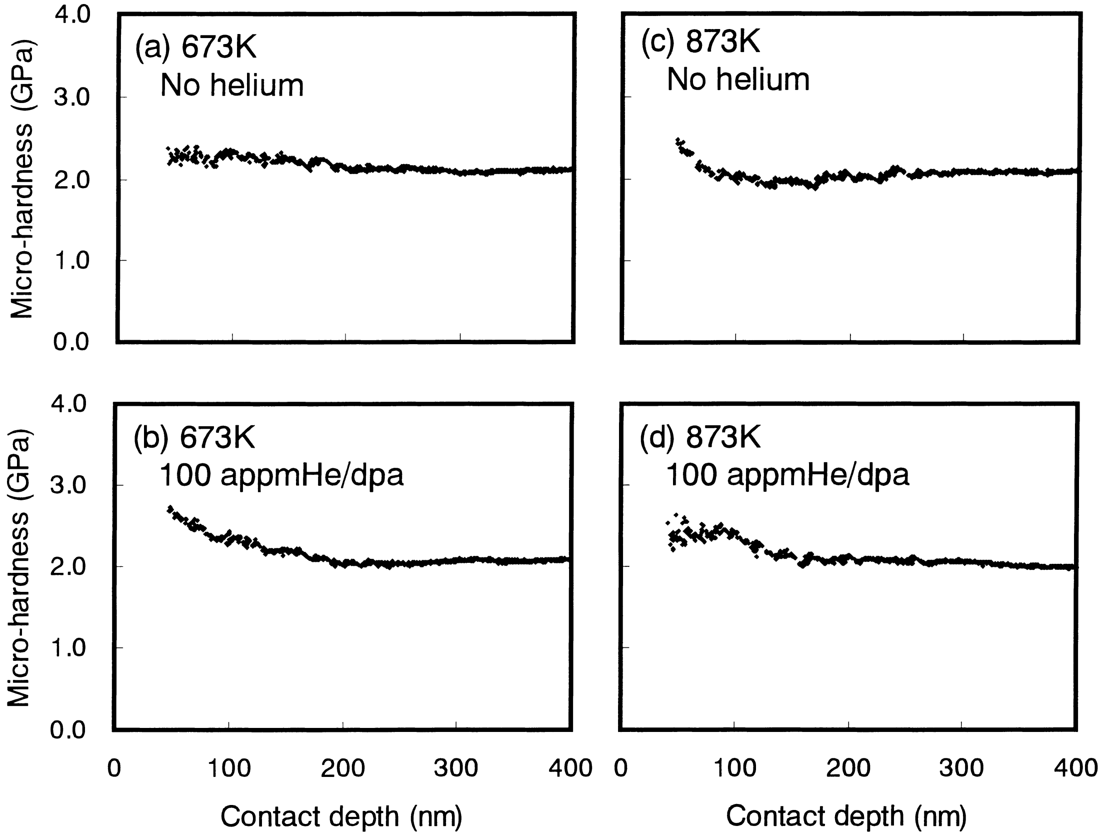

Fig. 3. Typical examples of micro-hardness profiles of the Fe-9Cr-2W martensitic steel irradiated by single- and dual-ions to a dose level of 1 dpa at a damage rate of 1×10^{-4} dpa/s.

tiny dislocation loops with radii of 2–5 nm at a number density of approximately 5×10^{21} m^{-3}. According to Orowan's simplest expression for dispersed barrier, the change in shear strength due to planar defects can be written as

$$\Delta\tau = \frac{\mu b}{\beta (Nd)^{-0.5}}, \qquad (3)$$

where μ is the shear modulus, b is the Burgers vector magnitude and β is a measure of the barrier strength [15], and $(Nd)^{-0.5}$ is the average barrier spacing where N and d are the obstacle's number density and average diameter, respectively. Taking the values of $\beta = 4$ [16], the assumptions of $\Delta\sigma_y \approx 3.1\Delta\tau$ [17] and $\Delta H \approx 3\ \Delta\sigma_y$ [18] gives $\Delta H \approx 0.33$ GPa, which is about 73% of the measured net hardening of the irradiated specimen, 0.45 GPa. Thus, the irradiation hardening might be due mainly to the contribution of tiny loops. Also, the existence of loops which contributed to the irradiation hardening should have slowed the thermal recovery of the pre-existing dislocation structures. Therefore, the hardening and the reduced softening in the specimen irradiated with dual-ions at 873 K can be attributed to the production of tiny dislocation loops. The variation of ΔH over grains may be due to the anisotropy of the barrier strength of a ⟨1 0 0⟩ dislocation loops.

The average micro-hardness data obtained from the entire specimen set are summarized in Fig. 4. The 573 K

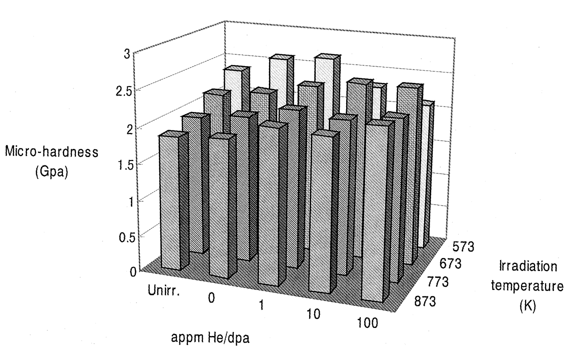

Fig. 4. Summary of the averaged micro-hardness of the Fe-9Cr-2W martensitic steel irradiated with 4 MeV nickel ions up to 1 dpa with and without helium ion co-injection. Note that the material condition for the 573 K irradiation was slightly different from that for others (see text).

data are less reliable compared to the others, because of insufficient surface finishing as described above. In all other temperature cases, hardness increased almost monotonically with the increasing He/dpa ratio. However, the amounts of hardening in most irradiation conditions were no more than 10% of the hardness in unirradiated materials. The effect of He/dpa ratio tended to be more pronounced as the irradiation temperature increased, within the range of irradiation conditions in this study.

5. Conclusions

A method for evaluating the micro-hardness profiles of dual-ion irradiated metallic materials was developed and then applied to study the synergistic effect of displacement damage and helium production on hardness properties of an Fe-9Cr-2W (JLF-1) low activation martensitic steel. The following are the important results of the present work:

(1) Increasing the He/dpa ratio almost monotonically increased the micro-hardness of materials irradiated at 673 K and higher temperatures. However, the irradiation-induced micro-hardness changes were no more than 10% except for very limited cases.

(2) The effect of He/dpa ratio was more noticeable at higher irradiation temperatures. This may be mostly because the dislocation evolution was influenced by helium more effectively at higher temperatures.

(3) Isothermal annealing at temperatures above 773 K caused softening within the surface layer of the specimens. Co-injection of only a small amount of helium prohibited the softening very effectively.

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