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A New Approach to Evaluate Irradiation Hardening of Ion-Irradiated Ferritic Alloys by Nano-Indentation Techniques

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The present work investigates the irradiation hardening of Fe-based model ferritic alloys after Fe-ion irradiation experiments in order to deduce mechanistically-based nominal hardness from the nano-indentation tests on the ion-irradiated surface. Ion-irradiation experiments were carried out at 290 °C with 6.4 MeV Fe^{3+} ions. The constant stiffness measurement (CSM) was used to obtain the depth-profile of hardness. The results has been analyzed and discussed based on the Nix-Gao model and an extended film/substrate system hardness model. The depth-sensing nano-indentation techniques with CSM revealed that the hardness gradient of the unirradiated Fe-based model alloy can be explained through the indentation size effect (ISE). On the other hand, the gradient of ion-irradiated surface of these samples includes not only the ISE but also softer substrate effect (SSE). We propose a new approach to evaluate a nominal hardness, which may connect to the bulk hardness, from experimentally-obtained nano-hardness depth profile data.

Keywords: Ferritic alloys, irradiation hardening, ion-irradiation, nano-indentation

1. Introduction

Irradiation hardening/embrittlement is one of the essential degradation issues of Fe based ferritic alloys for fusion reactor structural materials, such as reduced-activation ferritic (RAF) steels and oxide dispersion strengthened (ODS) ferritic steels. Heavy ion irradiation techniques using MeV accelerators have many advantages to investigate the irradiation effects on the fusion reactor materials: the short irradiation time, no induced-radioactivity, and co-implantation of helium/hydrogen [1]. In order to evaluate irradiation hardening in the ion-irradiated metallic materials, an instrumented indentation test [2], which is generally called as "nano-indentation test", has been used because the irradiation damage is limited in the surface up to a few micron meters [3-16].

In 1986, Zinkle and Oliver published the first paper concerning nano-indentation test on the ion-irradiated material [3]. Using a cross-sectional specimen obtained from an ion-irradiated Cu-Zr alloy, the depth-profile of hardness was evaluated. In contrast, the recent papers directly measured the hardness from the ion-irradiated surface. Such method can obtain the hardness from complex microstructure in RAF steels and ODS steels and can avoid the complicated procedure for cross-sectional specimen preparation. Katoh et al. measured the depth profile of hardness in ion-irradiated RAF steels using nano-indentation tests [5,10]. In their works, a polynomial function fitting to load-depth relationship during unloading process was applied to obtain continuous hardness profile in the ion-irradiated specimens. The results showed no significant depth-dependence in the hardness at depth of 50 nm to 400 nm in the as-received (non-irradiated) specimens. On the other hand, the present authors observed clear decreases in hardness of RAF steels with increasing the depth by a repeated load-unloading method [16]. Ullmaier and Camus investigated the hardness depth-profile in a RAF steel after dual-beam irradiations with 300 keV Fe^{+} ions and simultaneous implantation with 15 keV He^{+} ions by the continuous stiffness measurement (CSM) [4], which has been developed and used to obtain continuously the hardness depth profile by one indent [17]. The obtained results showed the depth-dependence of irradiation hardening.

Thus, in spite of the importance of nano-indentation tests on the ion-irradiated materials for fusion reactors, little attention was given to the hardness depth-profile obtained from the ion-irradiated surface. A clear understanding of the hardness depth profile of ion-irradiated material and its connection to the "bulk" mechanical properties is important for further application of the nano-indentation techniques. In the present work, we investigates the irradiation hardening behavior of the Fe-based model ferritic alloys after Fe-ion irradiations using nano-indentation tests and suggest a model to describe a nominal hardness for the ion-irradiated materials which may connect to the bulk hardness.

2. Experimental procedure

2.1 Materials

Materials used in the present study are Fe-based binary ferritic alloys: Fe-1Cu and Fe-1.4Mn in wt.%. These alloys are made by arc-melting in an Ar atmosphere. Disk (3mm diameter × 0.3mm thickness) specimens were punched out and were recrystallized at 825 ºC followed by quenching in iced water.

2.2 Ion-irradiation

Dual-ion irradiation experimental test facility, DuET in Kyoto University, has been used for the present single-beam irradiations of 6.4 MeV Fe^{3+} ions from a 1.7 MV tandem accelerator. A typical depth-profile of displacement damage calculated from TRIM-98 code

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[18] is shown in Fig. 1. The nominal displacement damage rate and total displacement damages were 1 × 10^{-4} dpa/s and up to 1 dpa, respectively. The “nominal” damage rate and damage stand for the values at around 600 nm depth from the irradiated surface, as indicated in Fig.1. The irradiation temperature has been well controlled at 290 °C within the error of ± 10 °C. The temperature of specimen surfaces during irradiations was monitored with an infrared thermal vision.

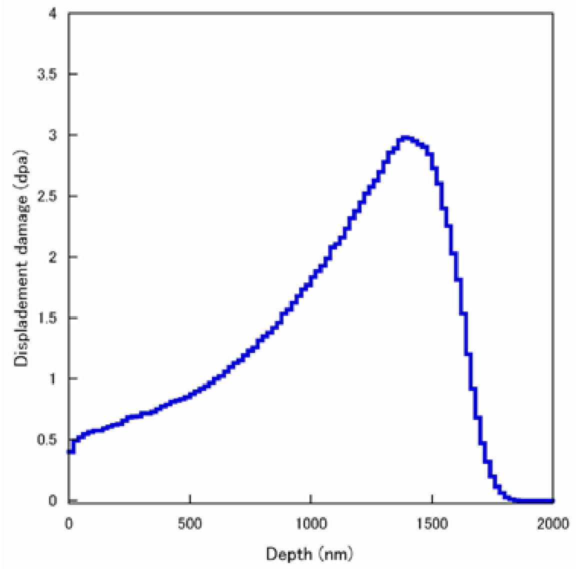

Fig.1 Depth profile of displacement damage (dpa) in the 6.4 MeV Fe^{+} ion irradiation.

2.3 Nano-indentation tests

Nano-indentation micro-hardness was measured by using the Agilent Technologies, Inc. Model Nano Indentor G200 with a Berkovich type indentation tip. The CSM were carried out with nm oscillation to obtain depth (h) -profile of nano-hardness (H). Calibration of blunting of the indentation tip and calculation of hardness is based on the Oliver-Pharr method [17].

3. Results and Discussion

Fig. 2 shows depth profiles of hardness in a) Fe-1Cu and b) Fe-1.4Mn before and after the ion-irradiation. For the unirradiated samples, the decrease in hardness with increasing indent depth was observed at the indentation depth of h > 50 nm. Such depth-dependent hardness behavior has been noticed as an indentation size effect (ISE) [19]. In contrast, for h < 50 nm, the increases in hardness with depth, known as a reverse ISE, was observed. Because the reverse ISE are usually attributed to testing artifacts, we don’t use the hardness data at h < 100 nm for further discussion in this study. In order to explain the normal ISE, Nix and Gao developed a model based on a concept of geometrically necessary dislocation [20]. Using the Nix-Gao model, the hardness depth profile is given as follows:

$$H = H_0\,(1+h^*/h)^{0.5} \qquad (1)$$

where H_0 is the hardness at infinite depth (i.e., macroscopic hardness), h^* is a characteristic length which depends on the material and the shape of indenter tip. In Fig. 2, the hardness data was plotted as H^2 versus $1/h$ to compare with the Nix-Gao model. The unirradiated specimens show good linearity in the range of h > 100 nm. Table 1 includes H_0 and h^* for the unirradiated samples which were calculated by the least square fitting of hardness data in the range of 100 nm < h< 500 nm to the equation (1). As expected, the H_0 values for unirradiated Fe-1Cu and Fe-1.4Mn are quite similar.

Comparing to the unirradiated samples, the ion-irradiated samples clearly showed increases of hardness in the depth investigated in the present work. Such irradiation hardening behavior of the ion-irradiated materials also can be seen in the literatures [4,5,10,14-16]. Note that the plot of ion-irradiated samples in Fig.2 and 3 has an inflection at a critical indentation depth (h_c) of ~250 nm for Fe-1Cu and ~330 nm for Fe-1.4Mn. Over the critical indentation depth, a contribution of softer unirradiated region (substrate) beyond the harder ion-irradiated surface to the measured hardness cannot be neglected with increasing the indent depth because the substrate will begin to plastic deformation before the indenter tip reaches the substrate. This kind of the softer substrate effect (SSE) can be seen in the various systems of hard thin film on soft substrate. Manika and Maniks investigated the effect of the ratio of the film/substrate hardness (H_f/H_s) on the ratio of the ritical indentation depth to the film thickness (h_c/t) for various film/substrate systems, where the H_f/H_s was in the range from 0.01 to 20, and revealed that the h_c/t strongly depends on the H_f/H_s [21]. Here, to apply the film/substrate model for the ion-irradiated materials, two assumptions are introduced as follows: Firstly, H_0^{irr} and H_0^{unirr} are assumed to be corresponding to H_f and H_s, respectively. Here, H_0^{irr} was calculated from the least square fitting for 100 nm < h < h_c to the equation (1). Secondary, the ion-irradiation damage range of 1.5 µm is assumed as t. After these assumptions, the relationship between h_c/t and H_0^{irr}/H_0^{unirr} for Fe-1Cu and Fe-1.4Mn exactly follows the results of Manika and Maniks for the hard film on soft substrate. Indeed, the h_c depends on the irradiation hardening level of ion-irradiated surface. This fact is a critical issue when the irradiation hardening of ion-irradiated samples was evaluated by nano-indentation tests with a constant depth method or a constant load method. With a constant depth method, for example, it is possible to underestimate the irradiation hardening when the H_0^{irr}/H_0^{unirr} is very high as to decrease significantly the h_c. However, the present method to derive the H_0^{irr} is believed to be useful to evaluate the irradiation hardening in the ion-irradiated materials because the mechanistically-based H_0^{irr} may connect to the bulk mechanical properties. In the present method, the corresponding damage level to the obtained H_0^{irr} should be set to the h_c.

Table 1: Calculated H_0 and h^* based on the Nix-Gao model

Material	unirr./irr.	H_0 (GPa)	h^* (nm)
Fe-1Cu	unirr.	1.44	315
Fe-1Cu	irr.	3.16	228
Fe-1.4Mn	unirr.	1.40	600
Fe-1.4Mn	irr.	5.90	34

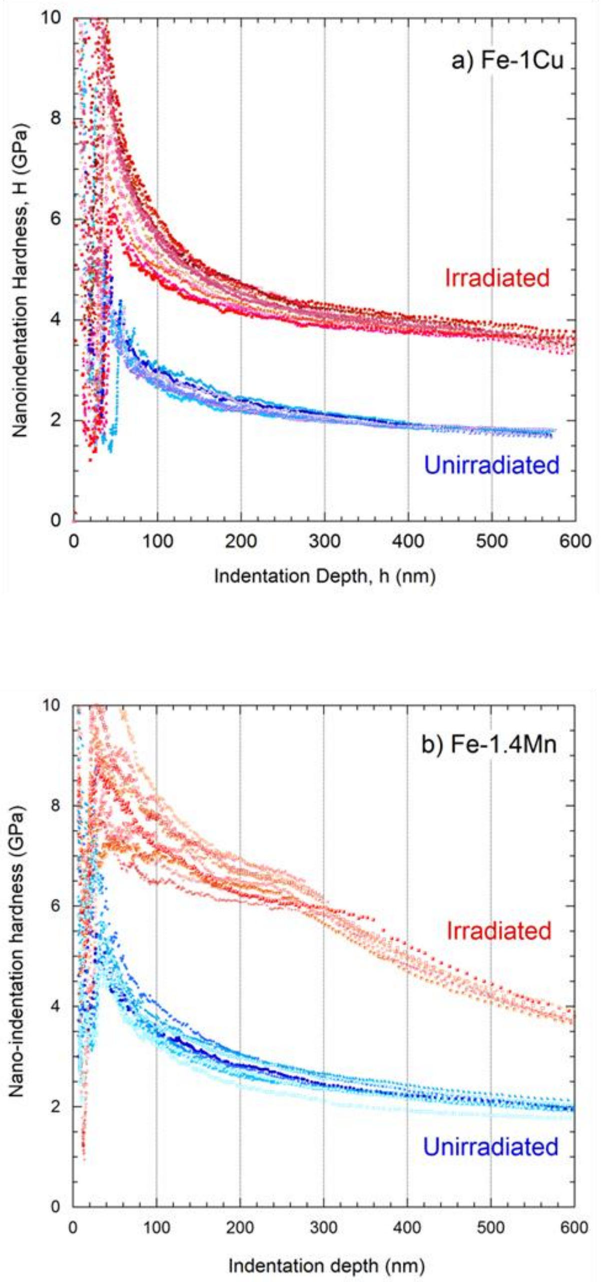

Fig.2 Depth profiles of hardness of a) Fe-1Cu and b) Fe-1.4Mn before and after 6.4 MeV Fe^{3+} ion irradiation at 290 °C.

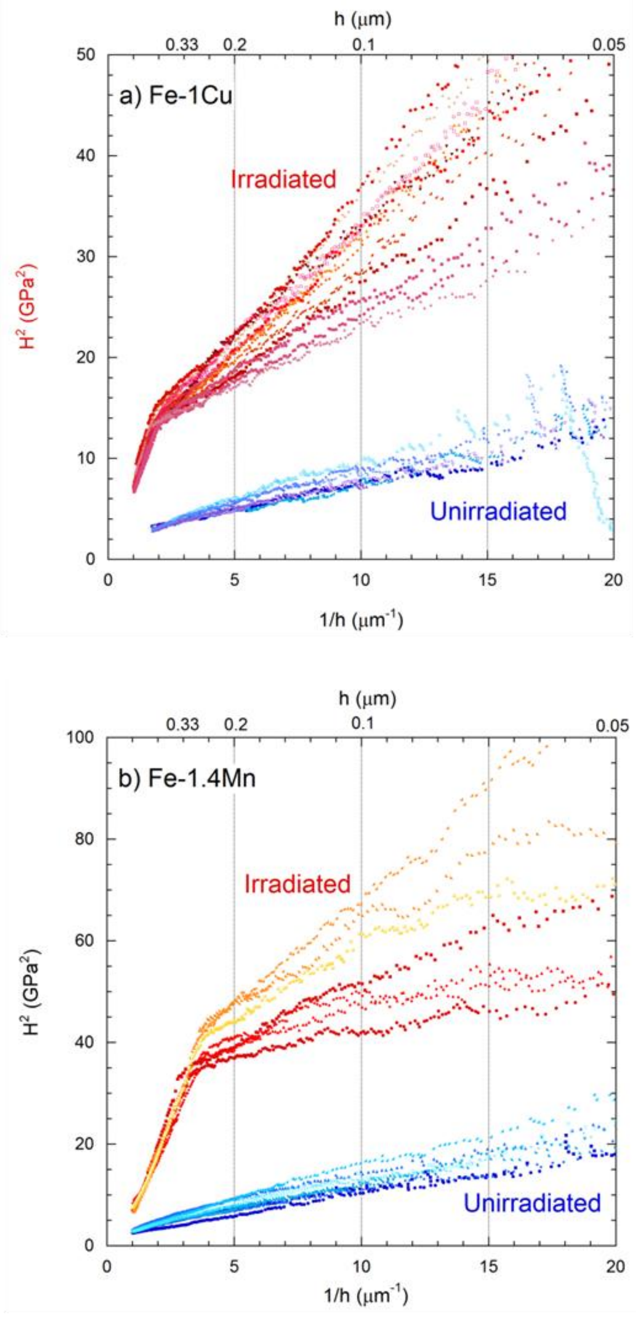

Fig.3 Plots of H_2 versus 1/h for a) Fe-1Cu and b) Fe-1.4Mn before and after 6.4 MeV Fe^{3+} ion irradiation at 290 °C.

Remained problems to model the depth-profile of hardness in the ion-irradiated materials is possible damage gradient effect (DGE) and implanted-ion effect (IIE). Shin et al. used the nano-indentation with CSM to evaluate the hardness profile of Fe-9Cr alloy after 2MeV Fe^{4+} ion irradiation [14]. The results clearly showed peak hardness at depth of approximately 250 nm, indicating that the DGE exists in this case. They also proposed a finite element model including the DGE to explain the hardness depth profile. In the present study, however, Fe-1Cu and Fe-1.4Mn have no clear DGE. This discrepancy is probably due to the difference of dose dependence of irradiation hardening between the materials [22]. Since the damage gradient in the range of $h < h_c$ is small as shown in Fig.1, a measurable DGE cannot be expected for the present Fe-1Cu and Fe-1.4Mn. The IIE may have an important effect when the ion species is not a self-ion. Taniguchi et al. reported the hardness depth profile of iron and copper after irradiations of 1.5 MeV Si ions and then 1.2 MeV C ions. They considered that the foreign element region introduced by the ion-irradiations blocks the deformation during nano-indentation [6]. Although implantation of self-ions also should give additional internal strain in the implanted layer of target materials, the effect is believed to be smaller than the effect of irradiation hardening due to displacement damage.

Although further experimental and theoretical studies to model the hardness depth profile are needed for the various materials after ion-irradiation, the proposed approach to express the nominal hardness after ion-irradiations provides a beneficial method to expand the ion-irradiation technique for evaluations of irradiation hardening in fusion reactor materials.

5. Conclusions

This paper suggested a new approach to derive a mechanistically-based nominal hardness from depth profiles of the ion-irradiated Fe-based ferritic alloys. The conclusions are as follows:

1. The hardness depth profile after self-ion irradiation could intrinsically include three different depth dependent effects: ISE, SSE, and possible DGE.
2. The Fe-1Cu and Fe-1.4Mn after the Fe-ion irradiation showed a composite depth profiles which includes ISE and SSE.
3. The nominal hardness H_0^{irr} of the ion-irradiated region can be determined by fitting the depth profile in the range of 100 nm < h < h_c to the Nix-Gao model.

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