Journal of Nuclear Materials 487 (2017) 343–347

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

A novel way to estimate the nanoindentation hardness of only-irradiated layer and its application to ion irradiated Fe-12Cr alloy

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ARTICLE INFO

Article history:
Received 3 January 2017
Received in revised form
26 January 2017
Accepted 18 February 2017
Available online 20 February 2017

Keywords:
Ion irradiation
Nanoindentation
Hardness
Fe-12Cr

ABSTRACT

While nanoindentation is a very useful tool to examine the mechanical properties of ion irradiated materials, there are some issues that should be considered in evaluating the properties of irradiated layer. In this study, in order to properly extract the hardness of only-irradiated layer from nanoindentation data, a new procedure is suggested in consideration of the geometry of indentation-induced plastic zone. By applying the procedure to an ion irradiated Fe-12Cr alloy, the reasonable results were obtained, validating its usefulness in the investigation of practical effect of irradiation on the mechanical behavior of future nuclear materials.

Energetic particle irradiation is known to seriously degrade the mechanical properties of a material [1–3]. Thus, understanding the mechanical properties in neutron irradiation environment is essential for assessing the suitability and reliability of a candidate material for nuclear applications and much research has been carried out on the related issue [4–7].

Since there are certain difficulties in exploring the effects of the neutron irradiation (e.g., it needs the long-time taken to achieve high doses, and makes the sample radioactive and thus difficult to handle), ion irradiation has been recently used as a surrogate for the neutron irradiation. However, for evaluating the irradiation-induced property change, ion irradiation has both merit and demerit in comparison of neutron irradiation; i.e., ion irradiation can produce high damage rates without residual radioactivity, whereas typical ion-irradiated layer has a limited thickness of only several μm below the irradiated surface [8]. In this regard, small-scale mechanical testing methods, especially nanoindentation test that can easily estimate the near-surface strength, have been extensively performed for estimating the mechanical behavior of ion-irradiated sample [9–13]. While nanoindentation is an useful tool for the purpose, there are still some issues that should be considered in evaluating the properties of only-irradiated layer through nanoindentation experiments, which will be introduced later. With this in mind, here we suggest a novel way to estimate the nanoindentation hardness of only-irradiated layer, and apply it to the analysis of ion irradiated Fe-12Cr alloy that is base for ferritic/martensitic steels considered as candidate materials for future reactor [7].

Fe-12Cr alloy having a chemical composition of Fe-11.9Cr-0.007C-0.021O-0.0003N (in wt%) was prepared by a vacuum induction melting using electrolytic metals. The ingot was homogenized at 1473 K for 24 h, forged, and then cold-rolled down to a thickness of 1 mm. The specimens were heat treated at a rate of 5 K/s to a recrystallization temperature for 3–5 h and then water quenched. Ion irradiation experiments were performed with a multipurpose Tandem ion accelerator at the Korea Institute of Geoscience & Mineral Resources. Before irradiation, specimens were polished electro-chemically at 18 V for 30 s in a mixture of 7% hypochlorous acid and 93% methanol for a removal of any surface damage. The specimens were irradiated with Fe^{4+} ions to three

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http://dx.doi.org/10.1016/j.jnucmat.2017.02.028
0022-3115/

different fluencies of 5.04×10^{14}, 2.80×10^{15}, and 6.72×10^{15} ions/cm^2 at room temperature (RT), and the used ion energy and beam current were 8 MeV and 200 nA, respectively. During irradiation, the backside of the aluminum sample holder was air cooled to prevent excessive heating and to keep the sample temperature below 313 K. The depth profiles of the displacement damage were calculated with the SRIM-2013 [14] based on the assumption that the value of threshold displacement energy is 40 eV [15]. The results are shown in Fig. 1 in which the maximum depth of the displacement damage is ~2.3 μm from the surface and the peak dose for the samples is 0.54, 2.69 and 6.45 displacement per atom (dpa), respectively. Hereinafter, each sample is named after its peak dose. Nanoindentation tests were performed on the ion-irradiated surfaces using a Nanoindenter-XP (formerly MTS; now Keysight, Santa Rosa, CA) with a typical Berkovich indenter. The sample is loaded to the peak load of 250 mN at a constant strain rate of 0.025 s^{-1}.

Fig. 2(a) provides representative nanoindentation load-displacement (*P*-*h*) curves of an unirradiated and three irradiated samples. In the figure, from the data set obtained under continuous stiffness measurement (CSM) module, which allows to get hardness (*H*) values continuously as a function of *h* from single nanoindentation [16], the selected *H* values for *h* of 250–2500 nm with an interval of 250 nm are exhibited. It is evident that unirradiated sample exhibits a larger peak-load displacement (h_{max}) than irradiated ones, and the h_{max} decreases as the dose increases. From the curves, hardness (*H*) values were estimated according to the Oliver-Pharr method [16], and the obtained *H* values are given as a function of *h* in Fig. 2(b). Only the *H* values taken at $h > 200$ nm are considered here, in order to avoid possible artifacts rising from the imperfect indenter tip geometry and surface roughness. Fig. 2(b) demonstrates that there is a hardening behavior after ion irradiation and it becomes more pronounced with increasing the maximum dose. The increase in *H* of irradiated samples above that of unirradiated one is more clearly visible at a lower *h*.

Despite the obvious irradiation-induced hardening phenomenon in Fig. 2(b), one should be careful in interpreting the nanoindentation results in a quantitative way since there are several issues to be considered for more accurate analysis, as Hosemann et al. [10] pointed out. One of the crucial issues is related with the plastic zone developed underneath the indenter. Since the volume of the plastic zone is in general much larger than the indented volume, considerable portion of the plastic zone volume may correspond to the unirradiated material located below irradiation layer, leading to a reasonable expectation that *H* values from nanoindentation of irradiated surfaces can be seriously affected by unirradiated material. Efforts were made to consider this and thus to estimate the *H* of only-irradiated region (H_{irr}). For example, Hosemann et al. [10] suggested a solid approach based on the rule-of-mixture to correct the errors rising from the presence of unirradiated material within the plastic zone. Assuming that plastic zone is a hemisphere with a radius of five times the indentation depth (i.e., $r_p = 5h$ where r_p is the plastic zone radius), the volume fraction of irradiated layer in the plastic zone (V_{irr}) at a given *h* was determined by simply considering the thickness of irradiated layer (that can be estimated by SRIM; e.g., ~2.3 μm in Fig. 1). Then, the H_{irr} could be estimated by a simple rule-of-mixture with the calculated volume fraction of irradiated layer within the plastic zone. Although the procedure is reasonable and appropriate H_{irr} was obtained [10], the results could be somewhat semi-quantitative estimates due to too simplified assumptions (such as $r_p = 5h$) without detailed consideration of inhomogeneous property change within the irradiated layer.

In order to complement the simplification and thus to estimate more accurate H_{irr}, a modified way is suggested as follows. It should be noted that this approach is valid only in the case that a few data of yield strength, σ_y, at different dose are available, as to be introduced. First of all, for calculating the volume of indentation-induced plastic zone, we adopted the Johnson's expanding cavity model [17] in which plastic zone produced by conical indentation is assumed to be hemispherical with a radius r_p that is given by:

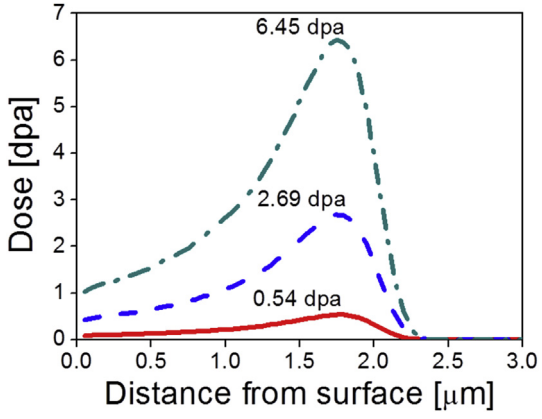
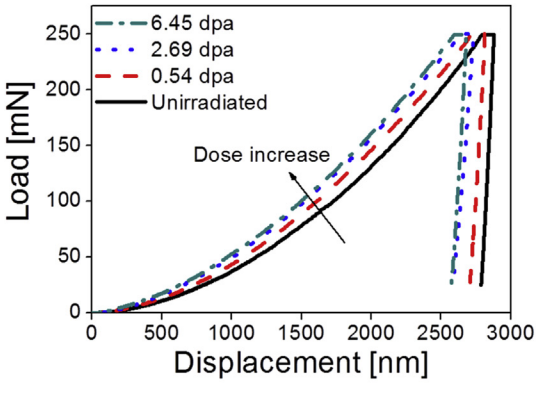
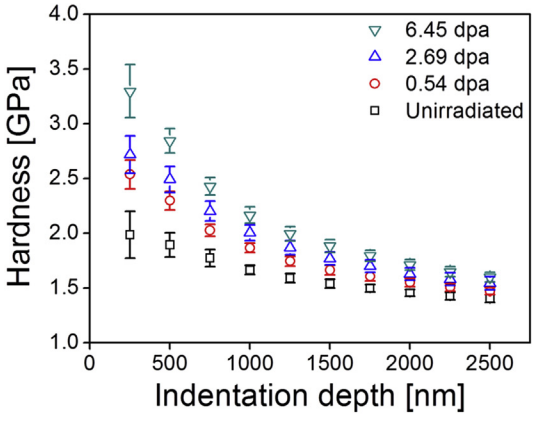


Fig. 1. Calculated results of irradiation damage vs. distance from the sample surface.



(a)



(b)

Fig. 2. Nanoindentation test results of unirradiated and irradiated samples; (a) load-displacement (*P*-*h*) curves, (b) depth-dependent hardness change.

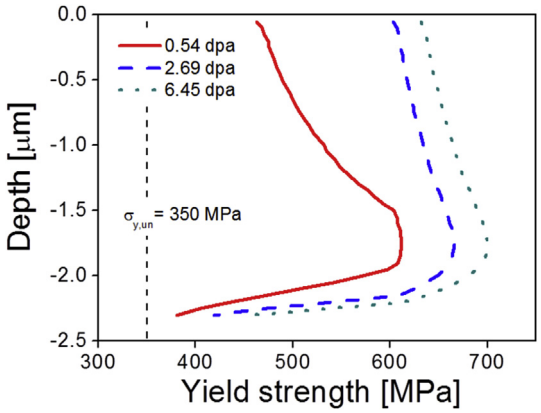


Fig. 3. Estimated yield strength change within ion irradiated layer.

$$r_p = a_c\left[\frac{1}{6(1-\nu)}\left\{\frac{E}{\sigma_y}\tan\beta + 4(1-2\nu)\right\}\right]^{1/3} \quad (1)$$

where a_c is the contact radius, and ν, E, and σ_y are the Poisson's ratio, elastic modulus, and yield strength, respectively. The β is the angle between sample surface and conical indenter. For a cone that would displace the same indented volume as a Berkovich does at a given h, β is 19.7°. The value of a_c can be calculated by putting the contact depth h_c (given by $h_c = h - \varepsilon(P/S)$ where P is load, S is contact stiffness, and ε is a geometric constant of 0.75 for a Berkovich indenter [16]) into the contact geometry equation $a_c = h_c \cot\beta$. Eq. (1) shows that r_p is not just function of h, but also depends on material properties, ν, E, and σ_y. It is known that the elastic properties, ν and E, can be irradiation-sensitive only if there is void swelling at elevated temperature [13,18]. In this study, ion irradiation was conducted at RT, and hence it is reasonable to expect there is no serious irradiation-induced change in ν and E. However, σ_y is reported to be strongly dependent on irradiation dose [4] so that the dose-dependency of σ_y is needed to be considered for estimating the r_p by Eq. (1). Among the models for irradiation hardening, the simplest one for the dose dependence of the irradiation-induced increase in yield strength ($\Delta\sigma_y$) is in the form of the power-law expression [19]:

$$\Delta\sigma_y = k(dpa)^m \quad (2)$$

where k and m are regression coefficients. It is well accepted that for many metals, the m is approximately 0.5 at low doses, but may become smaller at higher doses because there is trend towards saturation in irradiation hardening due to cascade overlap [19–21]. Therefore, as Byun and Farrell [19] suggested, two distinct regimes for Eq. (2) can be determined by drawing each log-log plot for low- and high-dose regime. Similarly, in this study, the m values for the two regimes were estimated with the literature data [5]. Note that although the σ_y data in Ref. [5] was measured at 573 K, they can be also adopted for RT irradiation case (as in this study) because, for Fe-based alloys, 573 K can be categorized as a low temperature [19] and thus irradiation hardening is almost independent of irradiation temperature in the range of RT to 623 K [22]. Among three available $\Delta\sigma_y$ data (for 0.06, 0.6, 1.5 dpa) of Fe-12Cr alloy in Ref. [5], it is reasonable to assume that 0.06 dpa case is in low dose regime, while 0.6 and 1.5 dpa cases are in high dose regime. Note that although only one set of $\Delta\sigma_y$ vs. dpa is available for low-dose regime, the constant k of Eq. (2) can be easily obtained by putting the known m of 0.5 into the equation. For high dose regime, it is easy to determine k and m of Eq. (2) since two known sets of $\Delta\sigma_y$ vs. dpa are available. From the crossing point of the two log-log plots, a transition from low-to-high dose regime could be determined as ~0.43 dpa. Now, Eq. (2) for the two regimes can be re-described as:

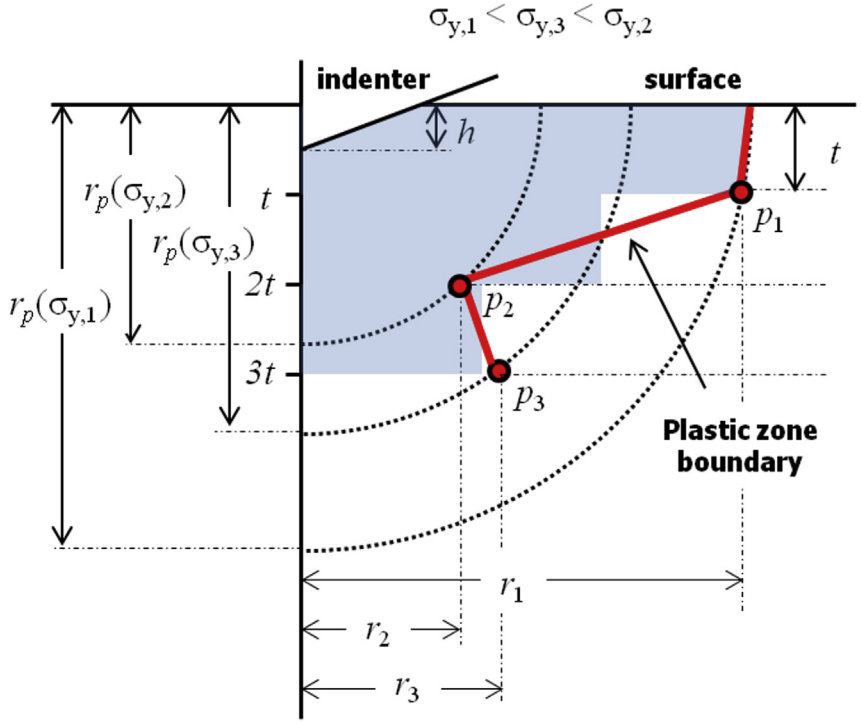


Fig. 4. Schematic illustration showing how to determine the plastic zone boundary.

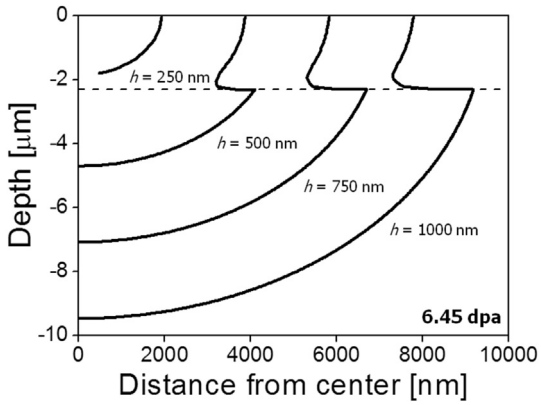


Fig. 5. An example (for 6.45 dpa) of predicted plastic zone geometry of irradiated sample.

$$\Delta\sigma_y = 387.8(dpa)^{0.5} \quad (0 < dpa \le 0.43)$$
$$\Delta\sigma_y = 281.3(dpa)^{0.12} \quad (dpa \ge 0.43)$$

These m values for the two regimes are in a good agreement with the results of previous study where Eq. (2) was applied to various metallic materials [19]. Fig. 3 shows the expected variation in σ_y along the depth from the surface with σ_y ~350 MPa of unirradiated sample [5].

With the σ_y profile in Fig. 3, one can estimate the change in r_p by Eq. (1) and then predict the plastic zone geometry in a way schematically shown in Fig. 4. First, the r_p at a given depth can be calculated by putting the corresponding σ_y into Eq. (1); e.g., in the figure, the r_p at t, $2t$, and $3t$ can be determined as $r_p(\sigma_{y,1})$, $r_p(\sigma_{y,2})$, and $r_p(\sigma_{y,3})$ by putting $\sigma_{y,1}$, $\sigma_{y,2}$, and $\sigma_{y,3}$ (that are σ_y at t, $2t$, and $3t$, respectively) into Eq. (1). Then, the point p where each circular arc of $r_p(\sigma_y)$ meets the horizontal line drawn for a given depth can be determined (e.g., in Fig. 4, p_1, p_2, and p_3 at t, $2t$, and $3t$, respectively). By connecting these p points, the boundary of plastic zone can be drawn. Fig. 5 shows the representative example (for 6.45 dpa) of the predicted plastic zone geometry (having the boundary determined as above) for various depth with an interval of 50 nm (i.e., t in Fig. 4 is 50 nm). At $h = 250$ nm, entire plastic zone is within irradiated layer (having thickness of ~2.3 μm below the surface), whereas in the cases of $h > 250$ nm, plastic zone is expanded into the underlying unirradiated region. Note that r_p of unirradiated region is unique with the assumption of unique σ_y (350 MPa [5]).

To evaluate the volume fraction of irradiated layer (f_{irr}) in whole plastic zone, it was assumed that the plastic zone within irradiated layer consists of a number of discs with the same height of 50 nm ($t = 50$ nm). As shown in Fig. 4, at a given depth, the distance from the central axis to each boundary point was denoted as r_i (e.g., in the figure, r_1 at t is the distance between the central axis and p_1), and the radius of the disc located between t and $2t$ was simply given as the average value of r_1 and r_2, ($r_{1\text{-}2}$). This makes it possible to easily calculate the volume of i th disc as:

$$V_{irr,\,i} = \pi t(r_{i-i+1})^2 \tag{3}$$

Thus, total volume of irradiated layer (V_{irr}) was calculated by summation of the volumes of the all discs within irradiated layer;

$$V_{irr} = \sum_{i=1}^{n} V_{irr,i} \tag{4}$$

where n is the maximum number of disc corresponding to the thickness of irradiated layer, i.e., in this study, n is 46 for the thickness of ~2.3 μm. Since the plastic zone of unirradiated region is developed underneath the irradiated layer (having thickness I ~2.3 μm), the plastic zone volume of unirradiated region (V_{un}) with a radius of $r_{p,un}$ is given by:

$$V_{un} = \frac{\pi(r_{p,un} - I)^2\left\{3r_{p,un}^2 - (r_{p,un} - I)\right\}}{3} \tag{5}$$

With the volume fraction of irradiated layer within indentation-induced plastic zone, $f_{irr} = V_{irr}/(V_{irr} + V_{un})$, and the experimental hardness values, H_{exp} (e.g., data in Fig. 2), the H_{irr} can be calculated by a rule of mixture:

$$H_{irr} = \frac{H_{exp} - H_{un}(1 - f_{irr})}{f_{irr}} \tag{6}$$

where H_{un} is the hardness of unirradiated sample. The H_{irr} values estimated in the way suggested above are summarized in Fig. 6(a), and the hardening ratio (representing how the material was hardened by irradiation), (H_{irr}-H_{un})/H_{un}, is provided as a function of h in Fig. 6(b). Irradiation-induced hardening ratio reaches up to 0.79, 1.24 and 1.72 times for 0.54, 2.69, and 6.45 dpa, respectively. Both trends in Fig. 6(a) and (b) are somewhat similar to the dose profile in Fig. 1, and the depth showing the maximum value in Figs. 1 and 6 are reasonably close to each other. This suggests that the new procedure proposed above may be useful for properly estimating the irradiation-induced hardening amount. Before

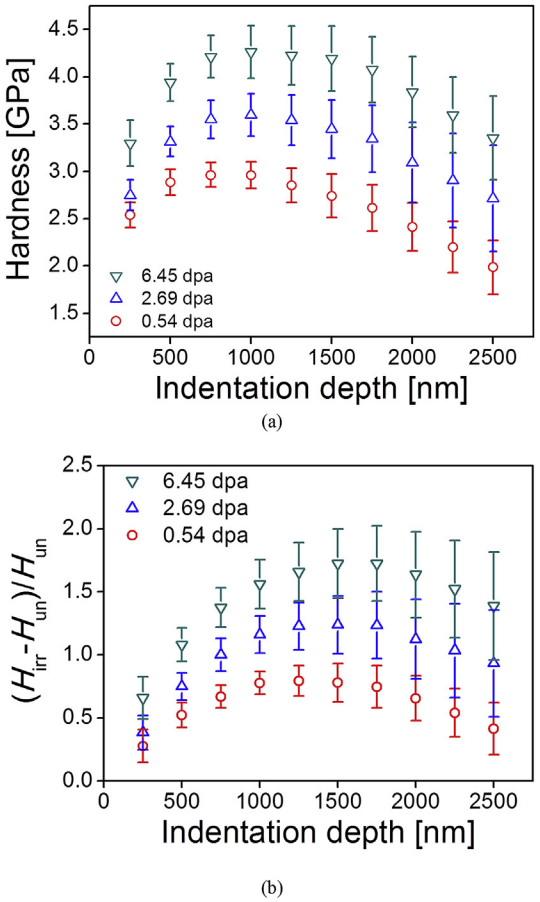


Fig. 6. (a) Estimated hardness of only-irradiated layer, and (b) depth profile of the irradiation hardening ratio.

closing, it is constructive to note two things. First, it should be reminded that this procedure requires a few (two or three) data of σ_y for different dose, which can be a limitation of its application. Second, there is non-negligible indentation size effect (ISE [10,23]) on the H_{irr} values (especially, at low h regime) of Fig. 6(a) and hence its additional consideration and correction is desirable in future study.

In summary, here we suggested a novel way to estimate the hardness of only-irradiated layer. In considerations of Johnson's expanding cavity model and the correlation between yield strength and dose, plastic zone geometry within ion-irradiated layer was estimated. Then, the hardness of only-irradiated layer was evaluated by applying a simple rule-of-mixture with the volume fraction of irradiated layer within plastic zone. The hardening behavior estimated from the new procedure show similar trend to that of dose profile, which can be an indirect evidence for the appropriateness of the proposed procedure.

This work was supported in part by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP) (No. NRF-2014M2A8A1030385), and in part by the NRF grant funded by the MSIP (No. NRF-2015R1A5A1037627).

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