E-Journal of Advanced Maintenance Vol.9-2 (2017) 72-77
Japan Society of Maintenology

Evaluation of Hardening under Ion Irradiation in Reactor Pressure Vessel Model Alloys by Nano-indentation Techniques

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ABSTRACT

The neutron irradiation embrittlement of reactor pressure vessel (RPV) steels needs to be properly predicted and managed for the safe long-term operation of light water reactors. To investigate the effects of the solute elements Ni, Mn and Si on the irradiation hardening at high dose levels, pertinent to long-term operation, in RPV materials without Cu, ion irradiation for Fe-1.0Ni-0.2Si, Fe-1.0Ni-1.4Mn and Fe-1.0Ni-1.4Mn-0.2Si alloys was carried out at 290 ^{o}C up to 5.0 dpa by Fe-ions with the energy of 2.8 MeV. Nano-indentation techniques were applied to evaluate the ion irradiation hardening. The results show that Mn significantly accelerates the irradiation hardening and addition of Si reduces the irradiation hardening. The depth dependence of irradiation hardening was discussed, and the irradiation hardening was the most significant at the indentation depth of about 120 nm in all alloys. The hardening behavior in deeper region was slightly different from that observed at about 120 nm depth, which may be caused by the interactions between solutes and defects.

KEYWORDS

reactor pressure vessel, long-term operation, irradiation embrittlement, irradiation hardening, ion irradiation, nano-indentation

ARTICLE INFORMATION

Article history:
Received 7 November 2016
Accepted 27 April 2017

1. Introduction

Long-term operation, or life extension of light water reactors is being considered by many utilities in their plant life management programs. The reactor pressure vessel (RPV) is the key safety-related component to confine radioactive materials, and is considered irreplaceable. Nano-features caused by neutron irradiation, including solute clusters and matrix damage, obstruct the dislocation movement, leading to hardening and accompanying embrittlement in RPV steels [1]. Long-term operation requires demonstration that RPVs operate within safety margins, including ensuring that the irradiation embrittlement of RPV steels during life extension period can be properly predicted and managed.

The response of RPV materials to neutron irradiation has been improved, especially by reducing the amount of Cu content since the 1970s when the role of Cu was identified [2]. It is recognized that Cu atoms cluster rapidly even at low neutron fluences due to its low solubility in Fe matrix and Ni, Mn and Si commonly segregate to the cluster-matrix interface [3]. But even for the low Cu steels (< 0.07%Cu), there is still a knowledge gap on how the microstructure and resultant hardening develop at high neutron fluences, pertinent to long-term operation, since the formation conditions of Ni-Mn-Si-rich clusters and their contribution to the hardening have not been well understood.

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This work is to investigate the effects of the elements Ni, Mn and Si, which are the main constituents of Ni-Mn-Si-rich clusters, on the irradiation hardening at high dose levels in RPV materials without Cu. This will also provide the knowledge base for our study of the microstructures using the same set of materials, which will not be described in this paper. For this purpose, high-dose ion irradiation experiments up to 5.0 dpa were carried out using three kinds of alloys prepared by removing Mn and Si from the alloy Fe-1.0wt.%Ni-1.4wt.%Mn-0.2wt.%Si. Hardness measurements by nano-indentation techniques are then presented.

2. Experimental

2.1. Materials and irradiation conditions

Three kinds of RPV model alloys (Fe-1.0Ni-0.2Si, Fe-1.0Ni-1.4Mn and Fe-1.0Ni-1.4Mn-0.2Si) were used. The sample surface was treated by mechanical polishing. This was then followed by electrochemical polishing to remove the work hardened layer. Ion irradiation with 2.8 MeV Fe-ions was carried out using a tandem-type accelerator installed at the High Fluence Irradiation Facility, The University of Tokyo (HIT) [4]. The irradiation temperature was 290 ^{o}C and the maximum ion fluence was 4.65×10^{15} ions/cm^2. The damage distribution associated with the ion irradiation was determined by using SRIM-2008 [5], as shown in Fig. 1. The calculation was conducted with a displacement energy of 40 eV and to be most consistent with the standard NRT dpa [6]. The displacement damage peak appeared at 800 nm and the calculated doses (at the depth of damage peak) in this work were 0.25 dpa, 2.5 dpa and 5.0 dpa. In this study, the dose rates of all the irradiation were well controlled at the same level in order to facilitate studying the dose dependent behavior in each alloy and comparing the results in different alloys. This is because the dose rate is recognized to affect the hardening [7] as well as the formation of Ni-Mn-Si-rich clusters [8] in RPV materials.

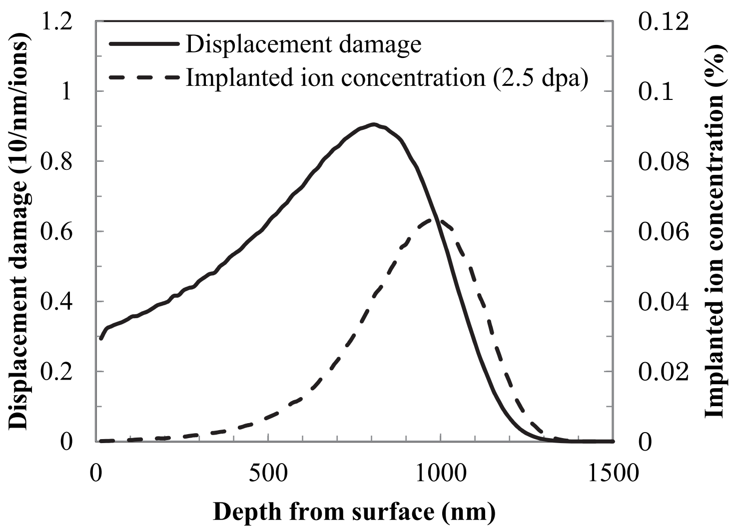

Fig. 1. Depth distribution of displacement damage and implanted ion concentration by the SRIM code

2.2. Nano-indentation tests

Nano-indentation tests were performed using two methods: the constant depth method by DUH-211S (Shimazu) and the constant stiffness measurement (CSM) method by G200 (Agilent), both with a Berkovich type indentation tip. Calibration and confirmation for the roundness of the indentation tip is based on the Oliver-Pharr method [9].

An indentation depth of 200 nm was set for the constant depth method. In order to calculate the hardness, we must know the values of contact stiffness. The constant depth method estimates the contact stiffness from the slope of fitted unloading curve. Such a calculation enables to determine contact stiffness, and thus hardness at the maximum indentation depth. In this work, the hardness was measured at approximately 400 indents per specimen and the distance between two indents was at least 20 μm [10]. The scattering of the hardness obtained by approximately 400 indents was less than 9% in unirradiated alloys.

The database provided by the constant depth method helped design the strategy for CSM tests in this work, including the shape of indentation area, indent distance and number of indents per specimen. Accordingly, the CSM tests were performed with 60 indents per specimen and an indent distance of at least 50 μm. Different from the constant depth method calculating the contact stiffness only at the maximum indentation depth, the CSM method is able to estimate the contact stiffness continuously during loading and then to give the depth profile of the hardness. This is accomplished by superimposing a small oscillation on the primary loading signal and analyzing the response of the system based on harmonic oscillator modeling [11].

3. Results and discussion

3.1. Dose dependence of hardness increase by constant depth method

Fig. 2 shows the dose dependence of hardness increase in these alloys under ion irradiation, given by the constant depth method. The hardness increase of Fe-Ni-Mn and Fe-Ni-Mn-Si is similar, and between them Fe-Ni-Mn shows higher irradiation hardening. Fe-Ni-Si shows the lowest irradiation hardening among the alloys, and its hardening difference with Fe-Ni-Mn-Si is the largest at 2.5 dpa. These facts indicate that Mn significantly accelerates the irradiation hardening. Phenomenologically, the addition of Si to Fe-Ni-Mn reduces the irradiation hardening.

Our results for the Mn and Si are consistent with the hardness data at lower doses obtained by Fujii et al. [12]. Their transmission electron microscope (TEM) observations show that the dislocation loops in Fe-Ni-Mn-Si are more developed (larger size and higher number density) than those in Fe-Ni-Si. The effect of Mn in increasing the number density of dislocation loops is also reported by Yabuuchi et al. [13]. This Mn effect can contribute to the high irradiation hardening of the alloys. Besides, the formation of solute clusters is also expected under such high dose irradiation in this work. Previous low temperature irradiation studies [14,15] conclude that the addition of Mn to pure Fe obstructs the recovery of radiation defects. It is interesting that the addition of Si to the Fe-Mn alloy is reported to mitigate this effect of Mn [16], which appears to suggest an interaction between Mn and Si. These differences in irradiation-induced defects may cause a difference in cluster formation.

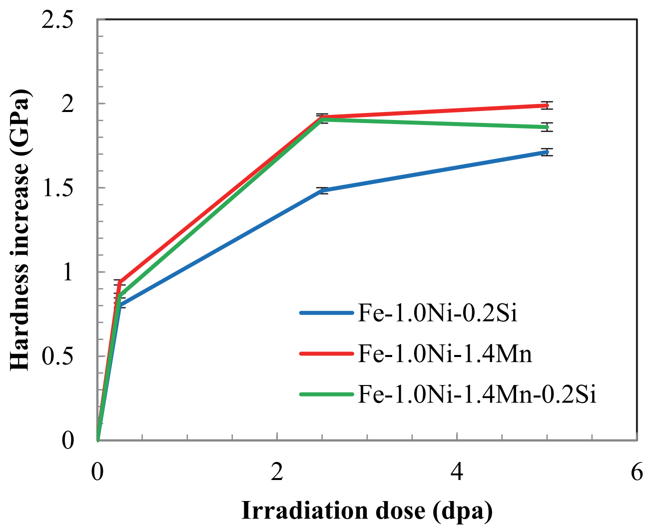

Fig. 2. Dose dependence of hardness increase in all alloys Error bars refer to the 95% confidence interval.

3.2. Depth dependence of hardness increase by CSM method

Since ion irradiation has a damage distribution along with depth direction, several methods have been applied to evaluate the depth dependence of hardness increase in previous studies [17,18]. The purpose of the CSM test in this work is to help correlate the hardening behavior of alloys to microstructure changes under ion irradiation. The data in the indentation depth less than 50 nm were considered less reliable and were ignored. Fig. 3 gives the indentation depth profile of average hardness of all indents with error bars in the alloys. It is seen that the measured value of hardness decreases with the increase of indentation depth in unirradiated alloys. This can be attributed to the indentation size effect introduced by Nix-Gao model [19].

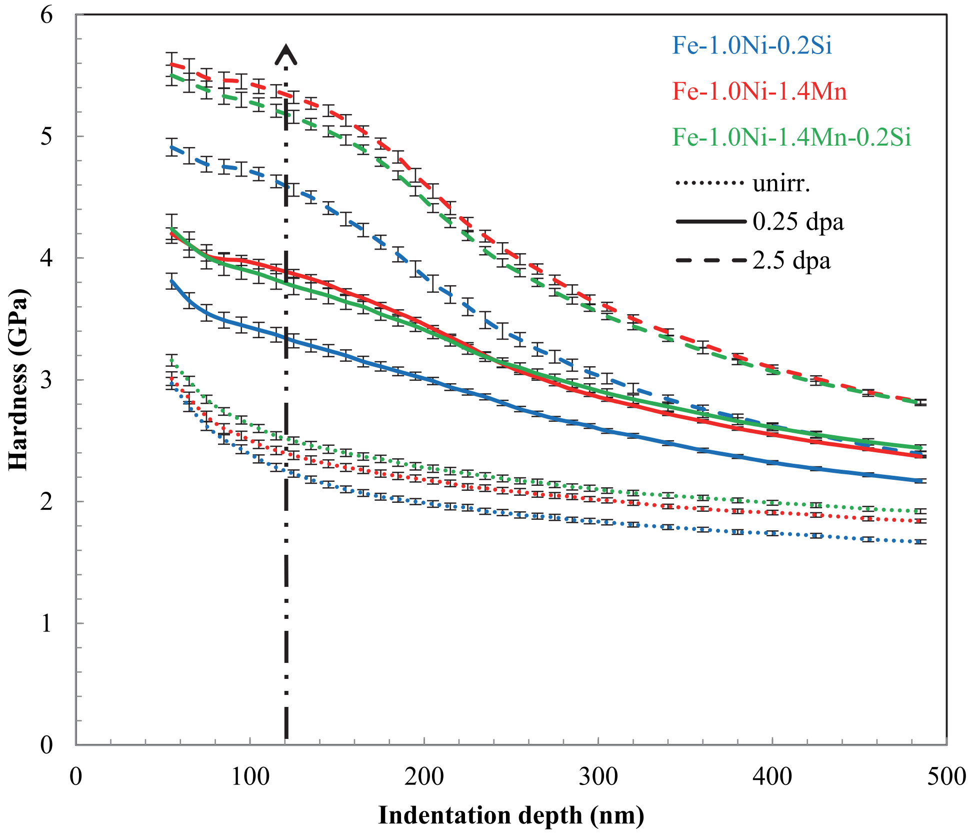

Fig. 3. Indentation depth dependence of hardness in alloys
Error bars refer to the 95% confidence interval.

In order to evaluate the ion irradiation hardening, one method is to assume simply that the indentation size effect is the same for the hardness before and after ion irradiation, and then to estimate the hardness increase ΔH_{CSM} by subtraction (see Eq. (1)), although this assumption is not based on theoretical model.

$$\Delta H_{\mathrm{CSM}} = H_{\mathrm{CSM,irr}} - H_{\mathrm{CSM,unirr}} \quad (1)$$

This simplified subtraction analysis shows that the hardening differences among these three alloys irradiated to 0.25 dpa can be observed up to 300 nm indentation depth, and these differences under 2.5 dpa can be observed until even deeper. The effects of Mn addition and Si addition on irradiation hardening observed in the shallow region agree qualitatively with the results obtained by the constant depth method. This analysis also indicates that there is a maximum value of hardness difference between irradiated and unirradiated alloys with the increase of indentation depth. The maximum difference appears at about 120 nm indentation depth for all alloys irradiated to 0.25 dpa and 2.5 dpa. At this indentation depth, the radius of volume affected by the indent reaches much deeper and may correspond to the depth of peak hardening region. The TEM studies [20,21] of deformation microstructure under nano-indentation show that the radius of volume affected by the indent can reach up to 10 times as large as the indentation depth for unirradiated pure Fe and unirradiated steel, and that this affected volume will be reduced by damage structure developed in ion irradiation.

As stated in the previous paragraph, irradiation hardening and their differences among these three alloys are still observed at deeper indentation depth, such as twice as deep as 120 nm. This is natural, because the measured value of hardening is the average hardening from the surface to the depth which corresponds to the boundary of the volume affected by the indent. In addition to this, microstructure may also form at deeper depth. While ion beam irradiation is performed, ions lose energy quickly because of high electronic energy loss, giving rise to not only a spatially non-uniform damage profile, but also a stopping of ions in the material itself. The peak of ion stopping range in this work is located at 1000 nm depth, deeper than the peak of damage profile (800 nm) , as shown in Fig. 1. As for 2.5 dpa irradiation, implanted Fe-ion concentration at this peak location reaches up to 0.06%. These implanted Fe-ions may act as effective self-interstitial atoms (SIAs) since there is no corresponding vacancy production for the recombination. Further modelling studies on migration of point defects along ion implantation direction are required, since these defects are expected to interact with solute atoms, causing different hardening behaviors along ion implantation direction in these alloys.

4. Conclusion

The hardening of RPV model alloys without Cu under high dose ion irradiation by 2.8 MeV Fe-ions at 290 ^{o}C was examined using nano-indentation techniques. The alloy without Mn showed less irradiation hardening and the alloys without Si showed more irradiation hardening. These facts indicate that Mn significantly accelerates the irradiation hardening, and addition of Si reduces the irradiation hardening. The depth dependence of irradiation hardening was evaluated, and a maximum hardness difference was observed at about 120 nm indentation depth for all alloys irradiated to 0.25 dpa and 2.5 dpa. This indicates that the radius of the volume affected by the indent at about 120 nm may correspond to the depth of peak hardening region. The hardening behavior in deeper region was slightly different from that observed at about 120 nm indentation depth. This may be caused by the interactions between solutes and defects.

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