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The changes of the structural, magnetic and mechanical properties in a RPV steel neutron irradiated at 70℃

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Abstract

The irradiation embrittlement of reactor pressure vessel (RPV) steel has been one of the main safety concern in the nuclear power plant. In the present study a SA508-3 RPV steel was irradiated by neutron with various fluence up to 10^{18} n/cm^2 (E > 1 MeV) at a temperature of approximately 70 ℃. The irradiation responses of the structural, magnetic and mechanical properties of RPV steel were investigated by means of X-ray diffraction, Moessbauer spectra, magnetic Barkhausen noise and micro Vickers hardness measurements. The transition of all of these parameters occurred above the neutron dose of 10^{16} n/cm^2. The results of X-ray and Moessbauer experiments revealed that neutron irradiation leads to the possibility of the partial amorphization in the investigated RPV steel. The changes of physical and mechanical properties can be explained in terms of irradiation induced cascade damage of crystalline materials.

1. Introduction

Recently, non-destructive evaluation methods such as measurements of magnetic properties and Moesbauer spectra have a growing interest as an evaluating techniques of the radiation damage of nuclear reactor pressure vessel (RPV). It has shown that the degradation of RPV steel is simultaneously accompanied by embrittlement, hardening, and decrease in ductility [1,2], and this degradation increases with increasing neutron dose. The irradiation embrittlement is a limiting factor of nuclear power plants lifetime, so better understanding of the relationship between the microstructure and the observed magnetic and mechanical properties is needed.

The most important microstructural change under neutron irradiation is the atomic displacement creating of defects : a self-interstitial and a vacancy clusters having the order of several nanometer [3]. These defects have a strong interaction with the magnetic domain walls [4]. However, the underlying mechanism for the effects of neutron irradiation on the magnetic characteristics is not understood yet.

Since the above mentioned effects are characteristics in the bombardments of high energy particles in amorphous as well as in crystalline alloys like RPV steel, it is interesting to evaluate the response of material parameters to the correlated microstructural changes and compare them in the both structural states [5]. For radiation damage problems, nuclear hyperfine interactions can be used to characterize the static and dynamic local configurations of both intrinsic and extrinsic defects. In view of this fact, the present paper is focused on the investigation of the irradiation induced changes in structure, magnetic and mechanical properties of RPV steel as a function of neutron doses.

2. Experimental

The sample was a SA508-3 forging steel for nuclear pressure vessels, produced by Korea Heavy Industries and Construction Co (HANJUNG). The samples of 18mm x 23mm with 1mm thickness were irradiated by neutron for various numbers of cycles at full power (1.5 MW) at a temperature of approximately 70 ℃. The accumulated dose was 10^{12} to 10^{18} n/cm^2 ($E > 1$ MeV) where iron wire was employed to determine the neutron dose.

The structural state of the sample due to the difference of neutron dose was investigated using X-ray diffraction and Moessbauer spectroscopy. X-ray diffraction measurements were made in Philips PW-1710 diffractometer using CuK$_\alpha$. The Fe^{57} Moessbauer spectra were collected in transmission geometry at room temperature by using a standard constant acceleration spectrometer with a ^{57}Co(Rh) source.

The magnetic Barkhausen noise (BN) was measured by specially designed magnetizing core using a sinusoidal current of 0.5～5 Hz. The detected BN signal by the same pick-up coil was amplified and passed through a wide-band filter of flat response between 500 Hz - 20 kHz. The B-H loop and the BN signal including Barkhausen noise energy (BNE) and Barkhausen noise amplitude (BNA), where BNE is defined as the time integration of the squared BN voltage for a magnetizing cycle, were measured as a function of the neutron fluence.

3. Results and discussion

The X-ray diffractogram of the un-irradiated and neutron irradiated samples as a function of neutron dose is shown in Fig. 1. Significant change of X-ray diffraction profile occurred above the neutron dose of 10^{16} n/cm^2. The X-ray diffraction patterns represented by Miller indices show that this samples have the typical bcc structure. With an increase of neutron dose some crystalline peak was disappeared in the corresponding X-ray diffractogram, revealing the possibility of the partial amorphization in the investigated RPV steel, which results in a drastic deterioration of mechanical properties. However, it may be a result of photoelectric effect due to the replacement of low energy X-ray photon by gamma ray photon [6].

In order to throw more light on the up to now unclear character of irradiation induced structural changes in the neutron irradiated RPV steel, the Moessbauer experiment were performed.

Fig. 2 shows the dose dependence of the Moesbauer spectra of neutron irradiated sample. In order to evaluate the corresponding hyperfine parameters, the measured spectra were fitted with three sets of six Lorentzians using a least-squares computer program, and the results are shown in Table 1. The hyperfine parameters nearly constant up to 10^{16} n/cm^2, followed by a rapid increase with neutron dose. The Moesbauer spectra do not show large variation up to neutron dose of 10^{16} n/cm^2, but significant changes of Moesbauer spectra were observed in the samples with neutron dose of 10^{17} n/cm^2 and 10^{18} n/cm^2. The high energy neutron bombardment changes the energy state of the constitute atoms associated wth nuclear or electron charge distribution, resulting in the corresponding hyperfine parameters. The resonance absorption area of A, B sites representing the Zeeman splitting [7] were decreased with increasing neutron dose, but no variation of peak position was observed. The trapped defect produces an electric field gradient at the site of the radioactive probe atom, which leads to a level splitting of the excited nuclear state. Therefore, the Zeeman splitting is attributed to the defect clusters attached on the ^{57}Fe isotope. However, it is not possible to directly identify a trapped defects by its characteristic parameters determined via the hyperfine interaction at the radioactive probe, since until now the theoretical models have not been able to reproduce these values with sufficient accuracy.

Fig. 3 shows the change of BNE, BNA and hardness as a function of neutron fluence. These parameters are characterized by three stages with respect to neutron dose. The rapid decrease of BNE developed at the early stage of irradiation and did not show a considerable change up to the neutron fluence 10^{16} n/cm^2, followed by a rapid decrease of 38% at 10^{18} n/cm^2, compared with unirradiated sample. The change of BNA also showed similar trend but the rate of change is less than that of BNE. These stages seem to be related with the behavior of irradiation induced defects on radiation damage under increasing irradiation dose. The profound effects of irradiation on BN signifies the change of domain wall motion from the dose of 10^{16} n/cm^2, in qualitative agreement with the results on hardness in the same figure. The decrease of BN is attributed to domain wall pinning and the increase of hardness due to the hindrance of dislocation motion by irradiation induced defect clusters [3]. Although the dimensional difference between dislocation and domain wall, it was anticipated that increased density of defect clusters with irradiation dose leading to greater hardness would also cause lower magnetic Barkhausen emissions because of increased number of defect pinning centers which impede both the movement of magnetic domain walls and dislocations.

The hardness difference between unirradiated and 10^{18} n/cm^2 irradiated specimen, ΔHv = 80, is very large considering the fluence level compared with that of 15 in the SA508-3 steel with dose of 9 x 10^{18} n/cm^2 (E>1 MeV) at 290 ℃ [8]. It is known that the hardness decreases with increasing irradiation temperature [9] and the SA508-3 steel was fairly insensitive to higher temperature [10]. The present study suggests that the degradation is more pronounced for irradiation at lower temperature.

The relationship between hardness and BN characteristic including the BNE and BNA giving some insight into the relation between mechanical and magnetic properties is shown in Fig. 4. The observed BNE and BNA showed linear relationship with hardness in the magnetization region, and similar relationship between Barkhausen parameter and hardness obtained by others [11,12]. Radiation hardening is due to the dislocation pinning by small defect cluster, whereas the BN associated with the domain-wall pinning by defect clusters, thus good correlation is found. The relation suggests that the radiation hardening can be evaluated non-destructively by using Barkhausen noise parameter.

4. Conclusion

The effects of irradiation on structural, magnetic and mechanical properties were investigated by means of X-ray, Moessbauer, magnetic Barkhausen noise and micro hardness measurements in the neutron irradiated RPV steel of both as-received, and irradiated with dose up to 10^{18} n/cm^2 (E>1 MeV). The results of all of these experiments showed similar characteristic trends with neutron dose : near constant up to a neutron dose 10^{16} n/cm^2, and rapid change above this dose. The BNE and BNA decreased and hardness increased with neutron dose, and all of these parameters showed great change above the neutron fluence 10^{16} n/cm^2. This dose level seems to correspond to the condition where the density of radiation induced defects reaches an appreciable fraction. From the sensitive change of BN and hardness, it seems that the defect clusters impede both the movement of magnetic domain walls and dislocation. The linear relation between hardness and BN parameters suggests that the degradation of mechanical properties by neutron irradiation could be evaluated well by the measurements of BNE and BNA.

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References

[1] M. K. Devine, D.C.Jiles, P.K.Liaw, R.D.Rishel and D.S.Drinon, Rev. of Progress in Quantitative Eval. 12, (1993), 1815

[2] J. F. Stubbins, W.-J.Shong, M.Giacobbe, A.M.Ougouag, J.G.Williams, ASTM STP 1204, (1993), 5

[3] D.R. Olander, "Fundamental Aspects of Nuclear Reactor Fuel Elements", Ch. 17 (1976)

[4] B.D. Cullity, Introduction to Magnetic materials, Ch. 4 (1972)

[5] P. Ehrhart, J. Nucl. Mat. 216 (1994) 170

[6] S. Glasstone and A. Sesonske, "Nuclear Reactor Engineering" Van Nostrand Company (1981) P.57

[7] E. Pecknagel and Th. Wichert, Nucl. Inst. Meth 182/183 (1981) 439

[8] J.T. Buswell, W.J. Phythian, R.J. McElroy, S. Dumbill and P.H.N. Ray, J. Nucl. Mater. 225 (1995) 196

[9] C.J. McHargue, Int. Met. Rev. 27, 3 (1982) 121

[10] G.R. Odette and G.E. Lucas, ASTM STP 909 (1986) 206

[11] Jae-Kyung Yi, Byung-Whi Lee and H.C. Kim, J. Mag. Mag. Mat. 130 (1994) 81

[12] Duck-Gun Park, Jun-Hwa Hong, In-Sup Kim and H.C. Kim, J. Mater. Sci. 32 (1997) 6141

Table 1. Change of (a) isomer shift, (b) quadrupole splitting, (c) magnetic hyperfine field, and (d) absorption area and line width

Fluence (n/cm^2) Parameters		10^0	10^{12}	10^{13}	10^{14}	10^{15}	10^{16}	10^{17}	10^{18}
I.S (mm/sec)	Site A	-0.01	-0.06	-0.02	-0.02	-0.02	-0.01	-0.03	-0.01
	Site B	-0.02	-0.07	-0.02	-0.02	-0.02	-0.03	0.00	-0.02
	Site C	0.21	0.18	0.21	0.18	0.21	0.17	0.32	0.33

(a)

Fluence (n/cm^2) Parameters		10^0	10^{12}	10^{13}	10^{14}	10^{15}	10^{16}	10^{17}	10^{18}
Q.S (mm/sec)	Site A	-0.09	-0.08	-0.08	-0.08	-0.07	-0.09	-0.09	-0.08
	Site B	-0.02	-0.05	-0.08	-0.07	-0.08	-0.04	-0.05	-0.04
	Site C	0.50	0.48	0.53	0.50	0.54	0.47	0.64	0.66

(b)

Fluence (n/cm^2) Parameters		10^0	10^{12}	10^{13}	10^{14}	10^{15}	10^{16}	10^{17}	10^{18}
H.F (kOe)	Site A	330.6	329.0	330.5	327.1	328.5	328.3	327.3	326.9
	Site B	306.6	304.3	308.3	305.3	306.5	307.2	303.8	304.0

(c)

Fluence (n/cm^2) Parameters		10^0	10^{12}	10^{13}	10^{14}	10^{15}	10^{16}	10^{17}	10^{18}
Area (%)	Site A	68.0	62.8	62.3	59.2	58.1	61.9	63.0	56.6
	Site B	27.4	33.2	33.4	36.5	37.2	33.9	22.0	22.1
	Site C	4.6	4.0	4.3	4.3	4.7	4.2	15.0	21.3
Line Width (mm/sec)	Site A	0.38	0.39	0.33	0.35	0.39	0.35	0.41	0.48
	Site B	0.43	0.37	0.34	0.33	0.40	0.38	0.53	0.75

(d)

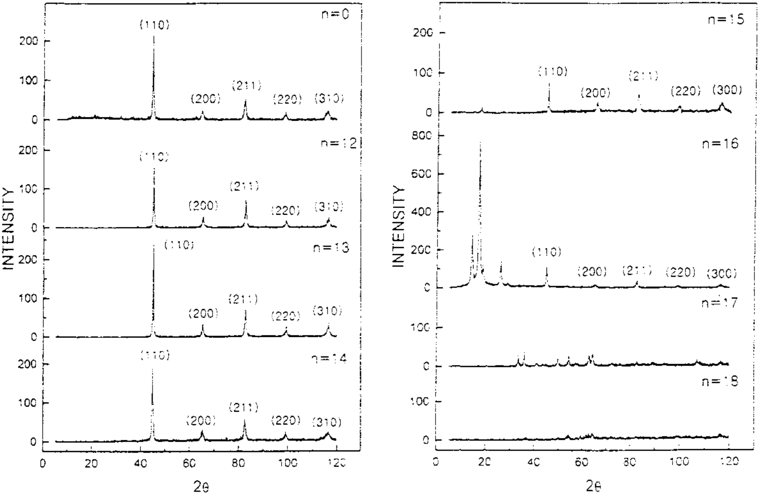

Fig. 1 X-ray diffraction pattern as a function of neutron fluence (10^n n/cm^2)

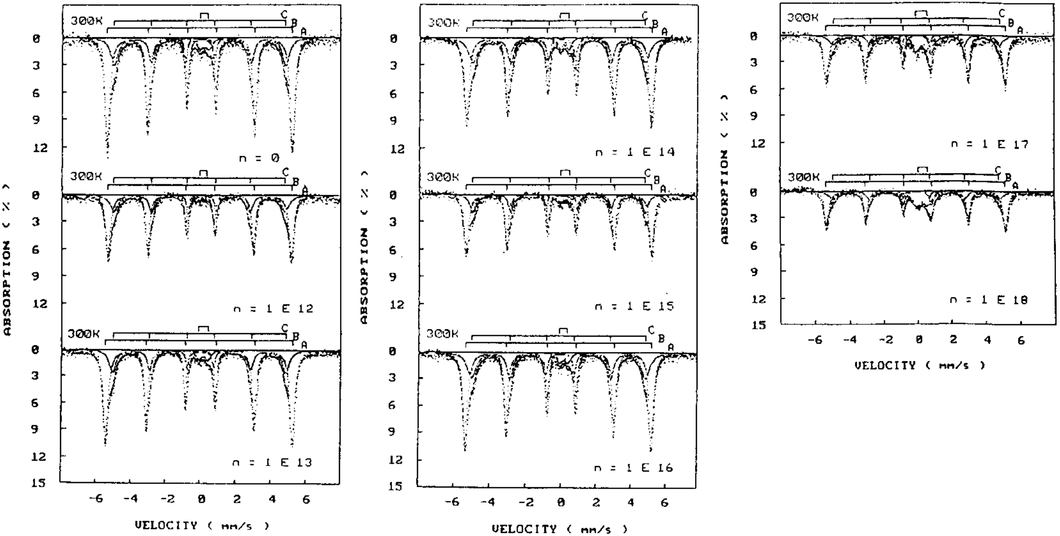

Fig. 2 Relative change of Moessbauer spectra as a function of neutron fluence (10^n n/cm^2)

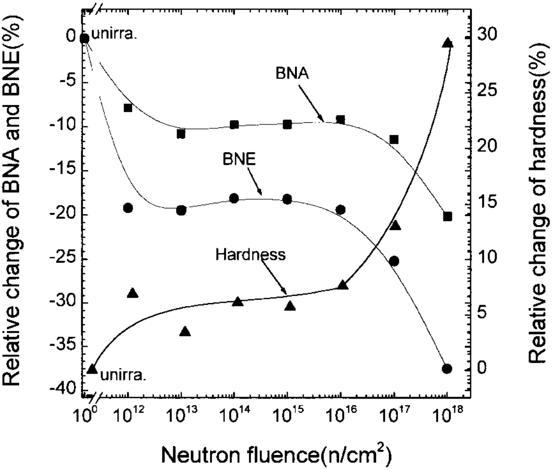

Fig. 3. Relative change of BNA,BNE and hardness as a function of neutron fluence(n/cm^2)

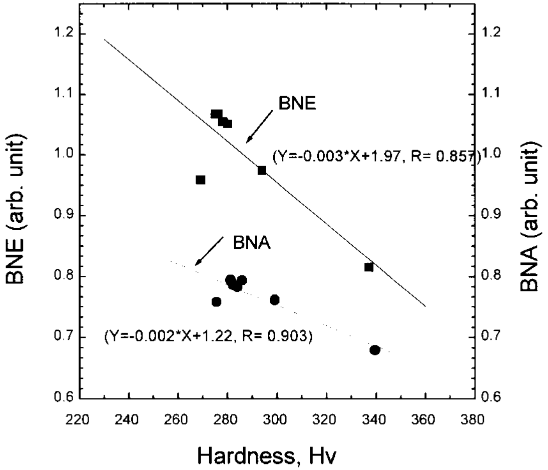

Fig. 4. The linear relation between BNA,BNE and Hardness.