DEGRADATION IN REACTOR PRESSURE VESSEL WELD SURVEILLANCE SPECIMENS

Kee-Ok Chang, Byeong-Chul Kim, and Sam-Lai Lee
Korea Atomic Energy Research Institute, P.O.Box 105, Yuseng, Daejeon, Korea

Abstract: Irradiation-induced changes in the magnetic parameters and mechanical properties of a reactor pressure vessel (RPV) Linde80 high copper weld were measured and compared to explore possible correlations between the measurements. The specimens employed in the present study were obtained from a RPV surveillance program. The material was tested in the unirradiated and two different post-irradiation conditions(Irr. I: 1.23 x 10^{19} n/cm^2, Irr. II: 3.94 x 10^{19} n/cm^2 , E>1.0 MeV, 288℃). Saturation magnetization (M_s), remanence (M_r), coercivity (H_c), and Barkhausen noise amplitude (BNA) were measured for magnetic parameters, and Vickers microhardness tensile and Charpy impact tests were performed for mechanical property parameters. After irradiation, hysteresis loops appeared to turn clockwise, resulting in an increase in H_c, and BNA appeared to decrease after irradiation while all the mechanical property changes followed the same trend as previously observed for low copper weld. Again, although limited, H_c and BNA were confirmed to be viable magnetic parameters that can be used in monitoring the mechanical parameter changes due to neutron irradiation.

Introduction: Recently, several sttempts have been made to apply magnetic methods in assessing and measuring the mechanical properties of irradiated reactor pressure vessel (RPV) steels [1-3]. Magnetic methods have been explored following the observation that the hysteresis properties and Barkhausen jumps results from the interaction of domain walls with lattice defects, and on the fact that neutron-irradiated materials acquire a high density of lattice defects (dislocations, precipitates, point defects clusters, etc.) in the reactor operational environment. Actually a few earlier studies were successful in correlating the magnetic measurements with the mechanical property or microstructural changes, for example, due to the heat treatment or stress [4-5]. However, a lack of data on irradiated RPV steels still prevents the application of the method for monitoring and characterizing the mechanical and microstructural changes in RPV materials during prolonged exposure to neutron irradiation.
In the present work, using the same rationale and measurement techniques as applied on low copper forging and weld RPV surveillance specimens [6], the magnetic and mechanical property parameters were obtained to explore possible correlations between these parameters and magnetic measurement data for a neutron irradiated Linde 80 high copper weld (HCW).

Experiment: The specimens used in the present study were obtained from the RPV surveillance program of a commercial reactor with a HCW in both the unirradiated condition and following irradiation to two different fluences (1.23 X 10^{19} n/cm^2, 3.94 X 10^{19} n/cm^2 , E>1.0 MeV, 288℃). These fluences correspond to 9 and 22 years of exposure at the surveillance capsule location in the reactor, respectively. All the mechanical properties data (Vickers microhardness, tensile and Charpy properties) were obtained from the RPV surveillance program. It is emphasized that all the unirradiated and irradiated specimens for magnetic measurement were fabricated from all weld specimens that had not been tested. Table 1 shows chemical compositions and heat treatment condition of the HCW from which the present specimens for magnetic measurement were obtained. It is seen that the copper content is high compared to the current ASME material specification, 0.1 wt % [7]. The microstructure of HCW is bainitic.

Table 1. Chemical composition and heat treatment conditions of Linde80 high copper weld specimens (wt.%).

C: 0.053 Mn: 1.60 P: 0.015 S:0.016 Si: 0.44 Ni: 0.55 Mo: 0.47 Cu:0.22
607±3.9 □, 20.25 h, Furnace-cooled stress relief

Magnetic hysteresis loops were obtained by using a vibrating sample magnetometer (VSM) for 3 mm diameter disk shape specimens to avoid the demagnetizing effects due to specimen shape and size. Saturation magnetization (M_s) was obtained for 5kOe, and coercivity (H_c) was measured for 2kOe. A U-shaped ferrite magnet core was used to magnetize a specimen of size 3 X 3 X 10 mm^3. The electromagnet was placed along the length of the specimen with the magnetic-field direction parallel to the length of the specimen. A magnetic field was applied to the specimen by supplying a sinusoidal current (2.3 Hz) to the electromagnet. An encircling sensing coil wound around the specimen was used to measure the magnetic induction in the specimen. BNA was measured by bandpass filtering (16-18 kHz) the magnetic induction signal.

Results and Discussion:

A. Mechanical property changes due to irradiation: Results of unirradiated and irradiated microhardness, tensile and Charpy properties measurements are summarized in Table 2. As seen, all the mechanical properties, especially the measures of transition temperature, appeared to be close to saturating at 1.2 x 10^{19} n/cm^2. Thus, for the irradiation condition II of 3.94 x 10^{19} n/cm^2, 3.2 times larger than the irradiation condition I, 1.2 x 10^{19} n/cm^2, the mechanical property changes are 0.8 % □ 75 % of the changes in irradiation condition I. In particular, the 35TT, 35 mil lateral expansion indexing temperature, seems to be saturated since the change is negligible even for an increase in fluence of 2.7 x 10^{19} n/cm^2. To examine the reliability of TT30 measurements, a comparison was made between the experimental measurements and the Wason, Wright and Oddette TTS model in NUREG/CR-6551 (MCS970501) Improved Embrittlement Correlations for Reactor Pressure Vessel Steels. Experimental measurements appeared to show a rather larger shift than the model prediction: 60 ℉ for Irr. I and 54 ℉ for Irr. II, respectively. These differences between the model prediction and experimental measurements, however, are quite acceptable if TTS residuals(model-actual) are compared with those appearing in the model calibration data, Fig. D.10. Residuals of weld data about TTS model with different Cu saturation values for Linde 80 welds versus other welds in NUREG/CR-6551. For reference, it is worth noting that the Linde80 high copper weld employed in the present study showed a rather higher increase in 30TT among 44 different Lindea80 HCWs than the respective yield strength increase after irradiation[9]. Higher hardening behavior in the HCW is seen in the yield strength change in comparison with that of the low copper weld [6].

Table 2. Unirradiated and irradiated Vickers microhardness, tensile and Charpy impact properties of the Linde 80 high copper weld.

		Tensile			Charpy		
	Microhardness (Hv)	YS[a] (MPa)	UTS[b] (MPa)	UE[c] (%)	TT30[d] (℃)	35TT[e] (℃)	USE[f] (Joule)
Unirradiated	208±4.3	482.6	593.0	15.1	-21.2	-6.7	90.2
Irr. I (1.23X10^{19} n/□)	251±12.2	580.5	670.2	10.4	94.6	128.9	63.1

Irr. II (3.94X10^{19} n/□)	262±8.2	636.4	728.1	8.7	109.5	130.1	54.8

[a]YS: Yield strength.
[b]UTS: Ultimate tensile strength.
[c]UE: Uniform elongation.
[d]TT30: 30 ft-lb(40.68J) indexing temperature.
[e]35TT: 35 mil lateral expansion indexing temperature
[f]USE: Upper-shelf energy.

B. Magnetic parameter changes due toirradiation: The changes seen in the hysteresis loop and BNA responses of the HCW appear to be the same as observed in the neutron irradiated low copper weld in the previous study. Thus, the hysteresis loop appeared to turn clockwise and BNA has decreased as the fluence increases. The hysteresis loop shape change due to irradiation is similar to the hardness effect in that the hysteresis loop becomes wider as the hardness increases due to heat treatment [8]. An example for the hardness effect due to heat treatment on hysteresis loop can be found in the work of Kwun and Burkhardt[10], where the mechanically harder specimens of AISI 410 stainless steel are also harder to magnetize. From the similarity in the hysteresis loop change between the hardening by irradiation and heat treatment, it may be concluded that some magnetic parameter change may be correlated with the hardness change.

Table 3. Magnetic parameter measurements for unirradiated and irradiated PRV Linde80 high copper weld metals.

	Hysteresis loop				Barkhausen Noise
Materials	M_s (emu/g)	M_r (emu/g)	M_r / M_s (%)	H_c (Oe)	(rms V)
Unirradiated.	210.2	5.4	2.6	7.3	13.08
Irr. I (1.23X10^{19} n/□)	212.5	5.0	2.5	7.5	9.83
Irr. II (3.94X10^{19} n/□)	208.2	3.4	1.5	7.9	8.46

Table 3 summarizes the magnetic parameter changes after irradiation. BNA appeared to decrease about 24.8% and 35.3%, and H_c to increase about 3 % and 8 %, respectively, from the unirradiated values after Irr. I and Irr. II, respectively. Any dependency on fluence was not observed in M_s (saturation magnetization) and M_r (remanence). The changes in M_s was small but discernible and not systematic. M_s was constant in the previous study irrespective of irradiation and materials. M_r appeared to decrease further as the fluence increased. The changes in the magnetic parameters after irradiation, especially in BNA and H_c , may be understood by the decrease in the mean free path for the domain wall movement and by the decrease in domain wall energy resulting from the interaction of domain walls with irradiation induced/enhanced defects, including, but not limited to, precipitates, interstitial or vacancy clusters and dislocations. Thus, the increase in H_c with fluence implies an increase in the strength of the interaction of the domain walls with irradiation-induced defects which will increase as the fluence increases. The decrease in the BNA may be understood from the decrease in the energy released when the domain walls are unpinned as the defect density increases.

C. Correlation between the changes in mechanical properties and magnetic parameters after irradiation: In the similar previous study on low copper forging samples obtained from a RPV surveillance program, several mechanical parameters were examined to find parameters that show viable correlation with magnetic parameter change due to irradiation[5.6]. Unfortunately, mechanical parameters related to the transition temperature shift due to irradiation did not show a viable correlation with magnetic parameters. This observation may be attributed to the difference in the physical or mechanistic processes that determine each parameters:

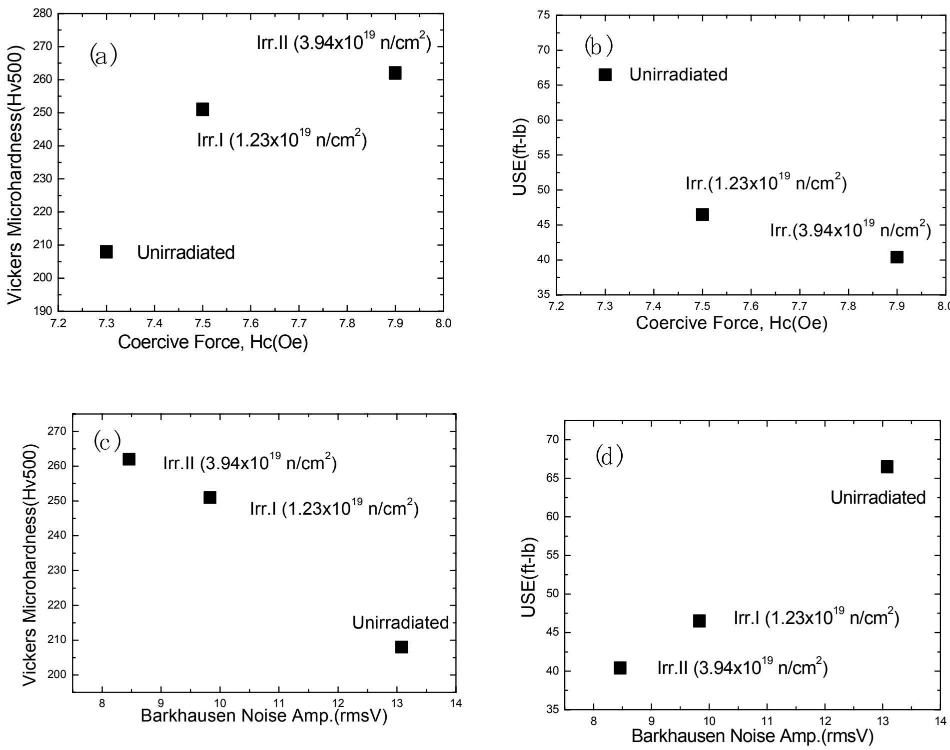

Fig.1 Correlations between mechanical property and magnetic parameters for Linde80 high copper weld metals

Thus, transition temperature shift due to irradiation is mainly determined by dynamic fracture properties and the change in magnetic parameters due to irradiation is determined by magnetic domain movement[6]. Among mechanical parameters examined, Vickers microhardness and Charpy USE showed the best correlation with magnetic parameters, i.e. H_c and BNA. Figures 1(a)-1(d) shows the correlations between mechanical property parameters (Vickers microhardness, Charpy USE) and magnetic parameters (H_c, BNA). It appears that both magnetic parameters correlate comparatively well with the mechanical property changes. These results are the same as the previous study on the base and low copper weld. As seen in Fig. 1(b), the parameter H_c shows again a viable correlation to the change in USE. The viability of parameters H_c and BNA as magnetic NDE parameters for monitoring the mechanical property change in RPV steels has already been confirmed on several occasions [1-4]. Although limited, the present data shows the additional possibility for the application of a magnetic method in monitoring the mechanical parameter change due to neutron environment. If a data base is prepared, both magnetic parameters may be used to monitor the changes in Charpy upper shelf energy and Vickers microhardness due to neutron irradiation.

Conclusion: Changes in the magnetic hysteresis loop and BNA of a RPV Linde 80 high copper weld surveillance specimen were almost the same as those of low copper weld and base forging metal RPV surveillance specimens of a reactor pressure vessel. Thus, after irradiation, BNA decreased and the hysteresis loop turned clockwise as neutron fluence increased. Both parameters show viable correlations to the changes in mechanical parameters (Vickers microhardness, USE) due to irradiation. If we consider many intrinsic and extrinsic factors that affect defect microstructures of RPV steels, comparisons of magnetic measurement data reported on irradiated RPV samples may need careful consideration of factors, including, but not limited to, irradiation temperature, flux, fluence, chemical composition, and heat treatment conditions of the specimens.

References:

1. M.R. Govindaraju, L.B. Sipahi, D.C. Jile, P. Liaw and D. Drinon, "NDE of irradiation degradation in nuclear pressure vessel steels" in Non-destructive Evaluation and Material Properties II, The Minerals, Metals & Materials Society, Warrendale, PA (1994) 121.
2. M. Blaszkiewicz, Mater. Sci. Forum **210-213**, 9, 1996.
3. M. K. Devine, D. C. Jiles, P. K. Liaw, R. D. Rishel, and D. S. Drinon, "Magnetic property changes in various structural steels due to irradiation" Rev. Prog. Quant. Nondestr. Eval. 12, 1815, 1993.
4. B. Raj, V. Moorthy and S. Vaidyanathan, "Influence of microstructural changes due to tempering at 923K and 1023K on magnetic Barkhausen Noise behavior in normalized2.25Cr-1Mo ferritic steel" Mater. Eval. **55**,81, 1997.
5. S. H. Chi, S. H. Song, D. G. Park, J. H. Hong, I. H. Kuk, and D. S. Shur, Proceedings of the Fourth Far East Conference on NDT (FENDT'97), 101-111.
6. Se-Hwan Chi, Kee-Ok Chang and Jun-Hwa Hong, "Changes in magnetic parameters of neutron irradiated SA508 Cl.3 reactor pressure vessel forging and weld surveillance specimens" Journal of Applied Physics, Vol.85, No.8, 1999.
7. ASME Boiler and Pressure vessel Code, Section II, Part A Material Specification.
8. K. Titto, Nondestr. Test Inst. **26**, 36, 1989.
9. Se-Hwan Chi, Bong-Sang Lee, Jun-Hwa Hong, iwf-lmNPP-97/2, Irradiation Effects and Mitigation. Proceedings of the IAEA Specialist Meeting held in Bladimir, Russian Federation; 15-19 September 1997. P240
10. Kwun H, Burkhardt GL. J. Appl Physics 1987 61:1576