Journal of ELECTRICAL ENGINEERING, VOL 61. NO 7/s, 2010, 119-122

A COMPREHENSIVE STUDY OF RELATIONSHIP BETWEEN MAGNETIC HYSTERESIS PROPERTIES AND IRRADIATION EMBRITTLEMENT IN NUCLEAR REACTOR PRESSURE VESSEL STEELS

Satoru Kobayashi* ─ Hiroaki Kikuchi ─ Seiki Takahashi ─ Yasuhiro Kamada

We report results of measurements of magnetic minor hysteresis loops for neutron-irradiated A533B-type nuclear reactor pressure vessel (RPV) steels with varying alloy composition and irradiation condition. A minor-loop coefficient, which is obtained from a scaling power law between minor-loop parameters shows a steep decrease just after irradiation, followed by a maximum in the intermediate fluence regime for most investigated alloys. This behavior is largely different from that obtained previously for RPV steels with a different initial microstructure. The results were explained by a model analysis assuming Avrami-type growth for Cu-rich precipitates and an empirical logarithmic law for relaxation of internal stress.

Keywords: neutron irradiation embrittlement, magnetic hysteresis, Cu precipitate, scaling analysis, pressure vessel steel

1 INTRODUCTION

Long-term irradiation with high-energy neutrons of reactor pressure vessel (RPV) steels leads to the formation of nanometer scale defects and makes the ductility lower and susceptible to rupture [1]. Currently, the irradiation embrittlement is evaluated by a ductile-brittle transition temperature (DBTT) shift obtained by Charpy impact tests. However, the diminishing stock of Charpy specimens preinstalled in the reactors has become an urgent issue and an establishment of a reliable non-destructive testing method is required.

The magnetic method using hysteresis loops is one of useful non-destructive evaluation methods to investigate the formation of microstructural defects in ferromagnetic materials. Since magnetic domain walls interact with lattice defects through magnetostatic, magnetoelastic couplings *etc*, and their movement is largely disturbed by the defects, which is reflected in a shape of magnetic hysteresis loops. Our recent studies on neutron-irradiated simple model alloys [2, 3] and RPV steels [4] revealed systematic variations of coercivity with the content of Cu, Ni, and Mn, which are key elements for irradiation embrittlement. However, the coercivity for RPV steels and model alloys showed an opposite dependence with fluence; the coercivity for the RPV steels with moderate Cu and Ni contents steeply increases at the early stage of irradiation, followed by a slow decrease with fluence, whereas that for the model alloys exhibited a decrease with fluence. In addition, even in RPV steels with a similar content of key elements, the trend just after the irradiation can vary depending on a pre-heat treatment condition. These observations imply that the coercivity change is governed by at least two mechanisms; the formation of nanoscale defects (Cu-rich precipitates, Ni-Mn-Si-rich precipitates, solute-vacancy clusters etc.) and the stress relaxation, which lead to an increase and decrease in coercivity, respectively. These results are in contrast to those of mechanical property measurements, where a yield stress change is simply related to precipitate volume fraction.

In this paper, we review our recent results of the comprehensive investigation of the relationship between magnetic hysteresis properties and irradiation embrittlement, and present model analysis results for an understanding of the magnetic property changes in neutron-irradiated RPV steels.

2 EXPERIMENTAL

2.1 Samples and irradiation condition

We examined two series of RPV steels listed in Tables 1 and 2; LV-series: A533B-type RPV steels with systematic variations of Cu and Ni contents, CWP-series: commercial or program steels. The baseline heat treatment for the LV alloys was as follows: austenitize at 900°C for 1 h, air cool, temper at 664°C for 4 h, air cool, stress relieve at 600°C for 40 h, and air cool. The heat treatment of the CWP alloys is dependent on the type of alloy and the detail is given in literature [5]. Chemical compositions of the LV-series alloys is similar to that of the CM-series alloys studied previously [4]; however, because of a high cooling rate after stress relief for the LV-series alloys, they have smaller grain size of 15 μm comparing with 50 μm for the CM-series alloys

Table 1. Chemical compositions of LV-series alloys

Sample	Cu	Ni	Mn	Mo	P	C	Si
LA	0.40	0.00	1.37	0.55	0.005	0.14	0.22
LB	0.40	0.18	1.35	0.55	0.005	0.14	0.22
LC	0.41	0.86	1.44	0.55	0.005	0.14	0.23
LD	0.38	1.25	1.38	0.55	0.005	0.19	0.23
LG	0.00	0.74	1.37	0.55	0.005	0.16	0.22
LH	0.11	0.74	1.39	0.55	0.005	0.16	0.24
LI	0.20	0.74	1.37	0.55	0.005	0.16	0.24
LJ	0.42	0.81	1.34	0.55	0.005	0.16	0.13
LK	0.80	0.81	1.13	0.55	0.005	0.13	0.13
LO	0.41	0.86	1.44	0.55	0.005	0.14	0.23

---

* NDE&Science Research Center, Faculty of Engineering, Iwate University, 4-3-5 Ueda, Morioka 020-8551, Japan; koba@iwate-u.ac.jp

ISSN 1335-3632

Table 2. Chemical compositions of CWP series alloys

Sample	Cu	Ni	Mn	Cr	Mo	P	C	Si
JRQ	0.14	0.82	1.40	0.12	0.50	0.019	0.18	0.25
HSST02	0.14	0.67	1.55	0.04	0.53	0.009	0.23	0.20
A302B	0.14	0.20	1.20	0.24	0.60	0.015	0.21	0.28
A508	0.06	0.80	1.30			0.005		0.01

Table 3. Irradiation condition (neutron energy E > 1MeV)

Irrad. condition	Flux (10^{12} n/cm^2/s)	Fluence (10^{19} n/cm^2)
T1	0.78	0.07
T2	0.78	0.18
T3	0.78	0.34
T4	0.97	0.75
T5	0.78	1.36
T6	0.97	3.32

Tensile test samples in the form of 24 × 5 × 0.5 mm^3 coupon were neutron irradiated at the University of Michigan Ford Nuclear Reactor under the University of California Santa Barbara (UCSB) irradiation variable (IVAR) program. The irradiation condition is listed in Table 3. The maximum fluence ϕ_t was 3.32 × 10^{19} n/cm^2 and the neutron flux ϕ was 0.78 or 0.97 × 10^{12} n/cm^2/s, which is categorized as high flux regime [5]. The irradiation temperature was 290°C.

2.1 Magnetic measurements and the analysis method

Magnetic hysteresis measurements were performed at room temperature using an apparatus designed for neutron-irradiated tensile test samples [3]. The sample was fixed by upper and lower yokes made of Fe-3wt% Si steel, which forms a closed magnetic circuit. A cyclic magnetic field with a frequency of 1 Hz was applied along the long axis of the sample, by an exciting coil wound around the sample.

A set of symmetrical magnetic minor hysteresis loops with various amplitudes of a cyclic field H_a up to 6 kA/m, was measured by increasing H_a, step by step. The magnetic properties for each alloy-irradiation condition were obtained from 1 or 2 samples. An experimental accuracy of magnetic properties was within 1% for each sample and errors of magnetic properties are mainly due to scattering in magnetic properties of samples.

Our analysis [6] showed that there exist several scaling power laws between parameters of minor loops in a limited range of H_a, such as

$$W_F^* = W_F^0 \left(\frac{M_a^*}{M_s} \right)^{n_F}, \qquad (1)$$

where W_F^* and M_a^* are hysteresis loss and maximum magnetization of a minor loop, respectively, and M_s is saturation magnetization. W_F^0 is a minor-loop coefficient sensitive to internal stress due to lattice defects and an exponents n_F is approximately 1.5, being independent of temperature, stress, and kinds of lattice defects. This analysis method using scaling laws is advantageous for magnetic non-destructive testing because of its sensitivity to lattice defects and low measurement field. The relation of (1) is valid for minor loops where irreversible movement of Bloch wall mainly contributes to magnetization. W_F^0, obtained at low applied fields is proportional to internal stress and is a useful magnetic property for evaluation of material degradation. In this study, the coefficient was used to investigate changes of internal stress in the LV- and CWP-series alloys after neutron irradiation.

3 RESULTS AND DISCUSSION

A set of minor loops with various field amplitudes H_a was analyzed according to (1). Figure 1(a) shows a typical example of a set of symmetrical minor loops, where the data for LD alloy before irradiation is shown. For each minor loop measured in various alloy-irradiation conditions, parameters were obtained according to the definition shown in Fig. 1(b), and their relations were examined in detail.

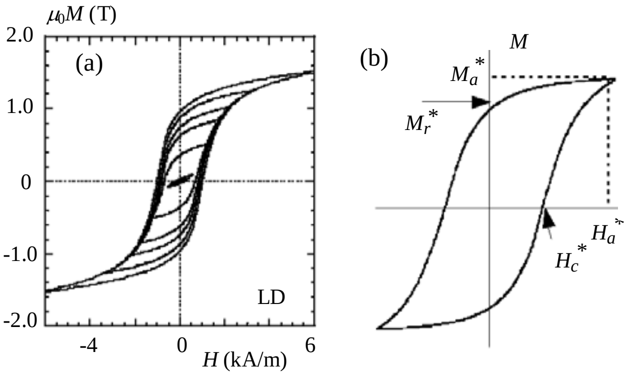

Fig. 1. (a) - A typical example of a set of minor hysteresis loops with various field amplitudes (LD). (b) - Parameters of a minor hysteresis loop

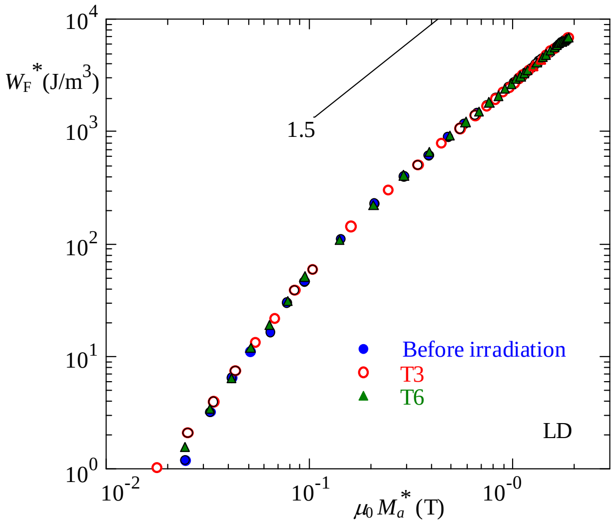

Fig. 2. Double logarithmic plots of the relation between hysteresis loss and maximum magnetization of minor loops for LD alloy before and after irradiation

Figure 2 shows the double logarithmic plots of the relation between W_F^* and M_a^* for LD alloy before and after irradiation. The W_F^*-M_a^* curves show straight lines in a limited range of M_a^* above 0.2 T and their slopes are almost the same before and after the irradiation. This M_a^* range corresponds to the field range where the irreversible movement of Bloch wall dominates in magnetization process and M_a^* has a steep slope in the H_a dependence. From least-squares fits of the data to (1), both an exponent n_F and minor-loop coefficient W_F^0 were determined for all alloy-irradiation condition. Here, minor loops with $\mu_0 M_a^*$ = 0.3-1.2 T were used for the fits and $n_F = 1.58 \pm 0.02$.

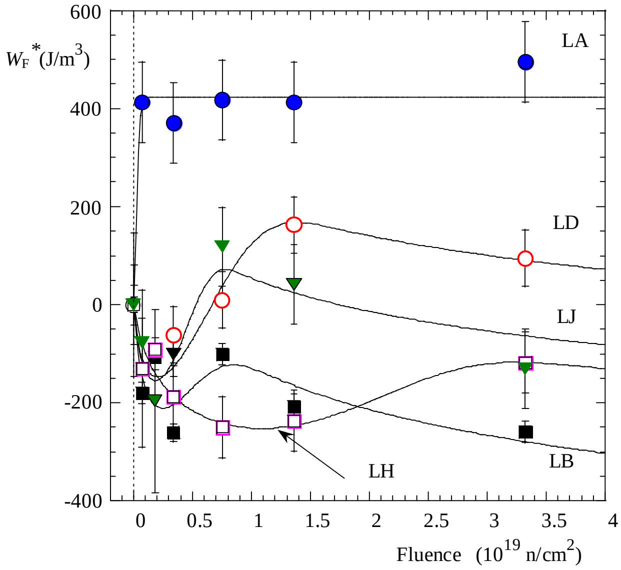

Fig. 3. Changes of minor-loop coefficient, ΔW_F^0, as a function of neutron fluence for some LV-series alloys. The solid lines through the data show least-squares fits to (2)

Figure 3 shows changes of the minor-loop coefficient, ΔW_F^0 as a function of neutron fluence ϕ_t for some LV-series alloys. For LA alloy with high-Cu and no-Ni contents, ΔW_F^0 steeply increases just after irradiation and becomes almost constant above $\phi_t = 0.2 \times 10^{19}$ n/cm^2. On the other hand, for most alloys with Ni contents larger than 0.18 wt %, ΔW_F^0 steeply decreases just after irradiation, followed by a broad maximum at a higher fluence. The height of the maximum seems to increase with increasing Cu and Ni contents. A similar behavior of ΔW_F^0 was also observed for the CWP-series alloys. These observations are in contrast with those observed previously in the A533B-type model alloys with a different heat treatment prior to irradiation, where ΔW_F^0 for high-Cu and high-Ni alloys shows a steep increase just after irradiation [4].

Depending on chemical compositions, various kinds of lattice defects are formed by neutron irradiation. These defects interact with magnetic domain walls and influence their movement. According to an earlier theory for micromagnetism [7], the arrangement of magnetization is determined so as to minimize magnetic Gibbs free energy consisting of exchange energy, magnetocrystalline anisotropy energy, magnetostatic energy, and magnetoelastic energy. In ferromagnets including lattice defects, the Gibbs free energy is lowered when domain walls are located at lattice defects and the defects act as obstacles to the domain wall motion. Therefore, one will expect that the formation of irradiation defects results in a monotonous increase of ΔW_F^0 with neutron irradiation. However, for most alloys ΔW_F^0 shows a sudden decrease just after irradiation. This result indicates that another mechanism that yields a decrease in internal stress exists besides the irradiation mechanism due to the formation of irradiation defects. Three mechanisms for the decrease in internal stress would exist there: (i) a decrease of internal stress of pre-existing dislocations due to the preferential formation of irradiation defects on those dislocations, (ii) a decrease of lattice distortion due to solute atoms in a matrix, associated with the growth of precipitates, (iii) relaxation of residual stress.

Assuming that the increase of internal stress obeys the Avrami-type growth equation for Cu-rich precipitates [8] and the decrease of internal stress results from relaxation of residual stress and the relaxation process obeys an empirical logarithmic law, ΔW_F^0 would be given by

$$\Delta W_F^0 = \Delta W_F^p \{1-\exp[-(F_p \phi t)^{\beta}]\}^{1/2} - S \ln(1+F_s \phi t)\ , \qquad (2)$$

where ΔW_F^P is a saturation value of ΔW_F^0 due to Cu-rich precipitation, F_p and F_s are a scaling parameter of ϕ_t, β, which was fixed to 3 in this study, is a parameter that determines ϕ_t range for 5-95% precipitation, and S is a relaxation rate. By least-squares fitting the fluence dependence of ΔW_F^0, ΔW_F^P was obtained for all alloys as listed in Table 4.

With increasing Cu and Ni contents increases ΔW_F^P, though the reason of a high value for LA alloy is not clear. This increase of ΔW_F^P is due to the increase of the volume fraction of Cu-rich precipitates due to Cu and Ni addition [1], [5]. On the other hand, F_p increases with Cu content, whereas it decreases with Ni content. This means that the growth of Cu-rich precipitates during irradiation becomes faster by the Cu addition, while the growth slows down due to the Ni addition, being consistent with results of mechanical property measurements [5].

Table 4. Fitting parameters ΔW_F^P and F_p for selected alloys

	Cu (wt%)	Ni (wt%)	ΔW_F^P (J/m^3)	F_p
LA	0.40	0.00	424±22	22.1
LB	0.40	0.18	292±39	1.5
LD	0.38	1.25	561±26	1.1
LH	0.11	0.74	290±35	0.4
LJ	0.42	0.81	408±30	1.7

Now, we compare ΔW_F^P with a change of yield stress $\Delta\sigma_y$, which is related to irradiation hardening [8] due to the formation of irradiation defects. Figure 4 shows the relation between ΔW_F^P and $\Delta\sigma_y$ (T6 condition) for LV- and CWP-series alloys. For most alloys except for LA and LC alloys, ΔW_F^P has a good linear relationship with $\Delta\sigma_y$. This indicates that irradiation hardening due to the formation of Cu-rich precipitates can be evaluated by measuring the minor-loop coefficient. Note that a largely small value of ΔW_F^p for LC alloy may be related to pre-precipitation before irradiation [5].

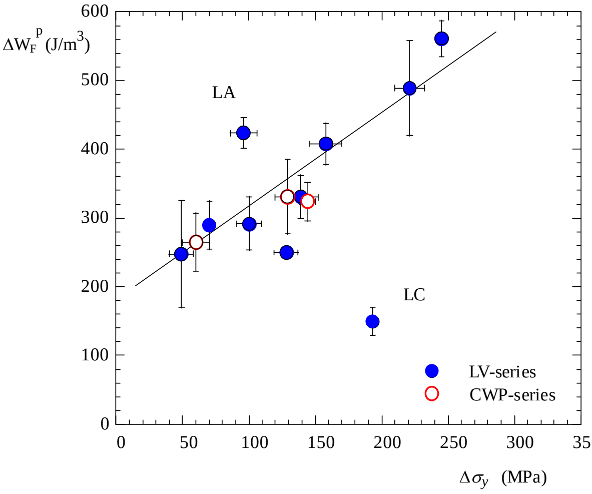

Fig. 4. The relation between ΔW_F^p and $\Delta\sigma_y$

$\Delta\sigma_y$ is generally a monotonically increasing function of neutron fluence and are a good indicator of irradiation hardening. On the other hand, a behavior of ΔW_F^0 is not simple and there exist at least two competing irradiation mechanisms. Which mechanism dominates the magnetic property changes seems to depend on the material initial condition (density of pre-existing dislocations, microstructure *etc*) as inferred from the comparison of the present results with our previous results for A533B-type RPV steels with a different heat treatment prior to irradiation [4]. Nevertheless, our model analysis revealed that an increase of the minor-loop coefficient, ΔW_F^p, shows a similar dependence on Cu and Ni contents irrespective of initial condition. This indicates that an increase of the minor-loop coefficient due to the formation of irradiation defects *ie* yield stress change $\Delta\sigma_y$ can be evaluated for any RPV steels by the model analysis.

Because of a maximum in ΔW_F^0, there are some neutron fluencies that give the same value of ΔW_F^0. For instance, for LJ alloy $\Delta W_F^0 = 0$ occurs at $\phi_t = 0$, ~0.5, and ~1.7×10^{19} n/cm^2. This means that $\Delta\sigma_y$ can not be estimated from a single measured value of ΔW_F^0. A combination of continuous measurements of ΔW_F^0 from the beginning of neutron irradiation and the model analysis would enable us to quantitatively evaluate $\Delta\sigma_y$ from the magnetic data. Further measurements on RPV steels to develop the database of the relation between ΔW_F^0 and $\Delta\sigma_y$ in various alloy-irradiation conditions taking account of material initial condition is now in progress and the results will appear elsewhere.

4 CONCLUSIONS

We have studied neutron irradiation effects on magnetic minor hysteresis loops for A533B-type nuclear reactor pressure vessel steels with a systematic variation of both alloy and irradiation conditions in order to develop the database of a relationship between minor-loop properties and irradiation embrittlement. A minor-loop coefficient, which is a sensitive indicator of internal stress, was found to decrease just after irradiation and then show a maximum in the intermediate fluence regime for most alloys. A model analysis assuming the Avrami-type growth for Cu-rich precipitates for the internal stress increase as well as a relaxation of internal stress which obeys an empirical logarithmic law showed that the increase of the minor-loop coefficient due to the formation of Cu-rich precipitates increases with Cu and Ni contents and is in linear proportion to a yield stress change. The results shows that the irradiation hardening can be generally evaluated by the magnetic method, but continuous magnetic measurements on RPV steels from the beginning of neutron irradiation is indispensable for a quantitative evaluation of the irradiation hardening due to Cu-rich precipitates.

Acknowledgement

We thank Prof. G. R. Odette of UCSB for allowing us to measure neutron irradiated samples, and Dr. T. Yamamoto, Dr. D. Klingensmith, Dr. D. Gragg of UCSB for their assistance of magnetic measurements. This work is supported by Grant-in-Aid for Young Scientists (B), Grant No. 20760588, from The Ministry of Education, Culture, Sports, Science and Technology, Japan.

REFERENCES

[1] ODETTE, G.R. – LUCAS, G.E.: Recent Progress in Understanding Reactor Pressure Vessel Steel Embrittlement, Radiat. Eff. Def. Solids. **144** (1998), 189-231

[2] KOBAYASHI, S. – KIKUCHI, H. – TAKAHASHI, S. – KAMADA, Y. – ARA, K. – YAMAMOTO, T. – KLINGENSMITH, D. – ODETTE, G.R.: The Effect of Copper and Manganese on Magnetic Minor Hysteresis Loops in Neutron Irradiated Fe Model Alloys, J. Nucl. Mater. **384** (2009), 109-114

[3] KOBAYASHI, S. – KIKUCHI, H. – TAKAHASHI, S. – CHIBA, K. – KAMADA, Y. – ARA, K.: Neutron Irradiation Effects in Fe-Cu-Ni-Mn Model Alloys Studied by Measurements of Magnetic Minor Hysteresis Loops, Philos. Mag. **87** (2007) 4047-4058

[4] KOBAYASHI, S. – KIKUCHI, H. – TAKAHASHI, S. – KAMADA, Y. – ARA, K. – YAMAMOTO, T. – KLINGENSMITH, D. – ODETTE, G.R.: Neutron Irradiation Effects on Magnetic Minor Hysteresis Loops in Nuclear Reactor Pressure Vessel Steels, Philos. Mag. **88** (2008), 1791–1800

[5] ODETTE, G.R. – LUCAS, G.E. – KLINGENSMITH, D. – WIRTH, B.D. – GRAGG, D.: The Effects of Composition and Heat Treatment on Hardening and Embrittlement of Reactor Pressure Vessel Steels, NUREG/CR-6778 (U.S. Nuclear Regulatory Commission, Washington, DC, 2003)

[6] TAKAHASHI, S. – KOBAYASHI, S. – KIKUCHI, H. – KAMADA, Y.: Relationship Between Mechanical and Magnetic Properties in Cold Rolled Low Carbon Steel, J. Appl. Phys. **100** (2006) 113908/1-6

[7] KRONMULLER, H. – Fahnle, M., *Micromagnetism and the Microstructure of Ferromagnetic Solids* (Cambridge University Press, Cambridge 2003)

[8] ODETTE, G.R. – LOMBROZO, P.M. – WULLAERT, R.A.: Relationship between Irradiation Hardening and Embrittlement of Pressure Vessel Steels, Effect of Radiation of Materials, ASTM STP **870** (1985) 840-860

Received 30 September 2010