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The effect of copper and manganese on magnetic minor hysteresis loops in neutron irradiated Fe model alloys

S. Kobayashi [a,*], H. Kikuchi [a], S. Takahashi [a], Y. Kamada [a], K. Ara [a], T. Yamamoto [b], D. Klingensmith [b], G.R. Odette [b]

[a] NDE and Science Research Center, Faculty of Engineering, Iwate University, Morioka 020-8551, Japan
[b] Department of Mechanical and Environmental Engineering, University of California Santa Barbara, Santa Barbara, CA 93106, USA

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ABSTRACT

Changes of magnetic minor hysteresis loops in pure Fe, Fe–1 wt% Mn, Fe–0.9 wt% Cu, and Fe–0.9 wt% Cu–1 wt% Mn model alloys after neutron irradiation have been studied. Minor-loop coefficients which are obtained from scaling relations between minor-loop parameters and in proportion to internal stress, were found to decrease in all model alloys after the irradiation to a fluence of 3.32×10^{19} n cm^{-2}. The decrease of the coefficients is larger for alloys including Cu and is enhanced by 1 wt% Mn addition. Such decrease implying the reduction of internal stress during irradiation is in contrast with changes of yield strength after the irradiation that increase with Cu and Mn contents. A qualitative explanation was given on the basis of the preferential formation of Cu precipitates along pre-existing dislocations which reduces internal stress of the dislocations.

1. Introduction

Accurate evaluation of neutron irradiation embrittlement of nuclear reactor pressure vessel steels and prediction of their lifetime have a key role for integrity assessments of nuclear reactors during operation. Long-term irradiation with high-energy neutrons makes ductility of steels lower and susceptible to rupture [1–3]. Irradiation embrittlement is associated with changes in various mechanical properties such as an increase in ductile–brittle transition temperature (DBTT), hardness, yield strength as well as a decrease in upper shelf energy. Currently, the irradiation embrittlement of pressure vessels is characterized by a change of DBTT obtained by Charpy impact tests, which is a destructive testing. However, the diminishing stock of Charpy specimens preinstalled in the reactors has became an urgent problem for long-term operation of nuclear reactors beyond their designed lifetime. An establishment and practical realization of a reliable non-destructive testing method are required [4].

The mechanisms of irradiation embrittlement related to the material hardening have been extensively investigated with various experimental methods ranging from fundamental bulk measurements such as DBTT, yield strength, microhardness, electrical resistivity, Seebeck coefficient, to direct nanoscale measurements such as transmission electron microscopy, atom-probe tomography, small-angle neutron scattering, positron annihilation [1,3]. It has been shown that the hardening is primarily due to the formation of nanoscale defects such as Cu rich precipitates, dislocation loops, solute-vacancy clusters etc., which pin dislocations from movement [2,3]. The nucleation and growing processes of these defects during neutron irradiation was found to be strongly influenced by various factors including neutron flux, irradiation temperature, chemical composition, and heat treatment prior to irradiation [3,5,6].

The magnetic method using hysteresis loops is one of useful non-destructive methods to get information on lattice defects [7–18]. Since magnetic domain walls interact with lattice defects through magnetostatic, magnetoelastic couplings etc., their movement is largely disturbed by the defects. The effective strength of the interactions depends on the size, density, spatial distribution, and kinds of lattice defects, which is reflected in a shape of magnetic hysteresis loops. It was revealed in cold-rolled steels [16] that coercive force H_c, which is one of important structure-sensitive hysteresis properties, is proportional to hardness and DBTT. Such good relationship between magnetic and mechanical properties has been confirmed in various steels where defect density is controlled by plastic deformation [9,13], thermally ageing [10,11], pre-heat treatment [12], and chemical compositions [12].

In spite of the deep understanding of magnetic properties in these steels, however, results of magnetic properties in neutron irradiated steels are still controversial [4,19–26]. For instance, in A533B nuclear pressure vessel steels irradiated to the fluence of about 1×10^{19} n cm^{-2}, H_c was found to decrease after neutron irradiation [21–23], whereas some groups reported the increase or no

* Corresponding author. Tel./fax: +81 19 621 6350.
E-mail address: koba@iwate-u.ac.jp (S. Kobayashi).

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significant changes [19,20]. This discrepancy is mainly attributed to a slight difference in an experimental set-up as well as sample conditions including pre-heat treatment, chemical compositions. Systematic investigation of magnetic property changes and construction of their database are indispensable for the practical realization of the magnetic method for nondestructive evaluation of pressure vessels.

Quite recently, we performed measurements of a set of minor hysteresis loops in neutron-irradiated Fe–Cu–Ni–Mn model alloys, where chemical compositions of Cu (0.05–0.9 wt%) and Ni (0.8, 1.6 wt%) contents were systematically varied [18]. It was found that minor-loop coefficients which are in proportion to internal stress decrease after neutron irradiation to a fluence of 0.44×10^{19} n cm^{-2} for all alloys. The decrease of the coefficients strongly depends on both Cu and Ni contents and is enhanced for high-Cu high-Ni alloy. Further, the decrease of the coefficients is roughly in inverse proportion to changes of yield strength.

In this paper, we focused on changes of magnetic minor hysteresis loops in neutron-irradiated pure Fe, Fe–1 wt% Mn, Fe–0.9 wt% Cu, and Fe–0.9 wt% Cu–1 wt% Mn model alloys with no Ni inclusion. The compositions are more fundamental compared with those of previous Fe–Cu–Ni–Mn model alloys, but the obtained results would give a deep insight into roles of each irradiation defect on minor-loop properties. The results were explained from the viewpoint of nanoscale features of irradiation defects which were revealed by atom-probe tomography, small-angle neutron scattering, positron annihilation using the same materials [27–29].

2. Experimental

We examined four model alloys listed in Table 1; Fe (VA), Fe–1.0 wt% Mn (VB), Fe–0.9 wt% Cu–1.0 wt% Mn (VD), and Fe–0.9 wt% Cu (VH) alloys. The alloys were solution treated for 17 h at 775 °C and then salt-bath quenched at 450 °C for 3 min, followed by slow cooling to room temperature in air.

Tensile test samples in the form of 24 mm × 5 mm × 0.5 mm 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 2. The maximum fluence ϕt was 3.32×10^{19} n cm^{-2} and the neutron flux ϕ was 0.77 or 0.97×10^{12} n cm^{-2} s^{-1}, which is categorized as high flux regime [6]. We examined the T6 condition for all model alloys, and also the K2 condition for the VD alloy. The irradiation temperature was 290 °C.

Magnetic hysteresis measurements were performed at room temperature using an apparatus designed for neutron-irradiated tensile test samples [18]. The sample was fixed by upper and lower yokes made of Fe–3 wt% 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. The details of the measurement system is given in Ref. [18].

A set of quasistatic magnetic minor hysteresis loops with various amplitude of a cyclic field, H_a up to 6 kA/m, was measured by increasing H_a, step by step. Before measuring each minor loop, the sample was demagnetized with decaying alternating magnetic field and then a minor loop, which is symmetric about the origin, was measured. These minor loops are different from the major loop, obtained with H_a large enough for saturation. In the present study, the hysteresis loop with H_a = 6 kA/m was assumed to be the major loop. The magnetic properties for each alloy-irradiation condition were obtained from 1 or 2 samples. For our measurement system, an experimental accuracy of magnetic properties was within 1 % for each sample and errors of magnetic properties are mainly due to their sample dependence.

Our analysis [14–16] showed that there exist several simple relations between parameters of minor loops in a limited range of H_a:

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

$$W_R^* = W_R^0 \left(\frac{M_R^*}{M_R}\right)^{n_R} \quad (2)$$

and

$$H_c^* = H_c^0 \left(\frac{M_R^*}{M_R}\right)^{n_c}. \quad (3)$$

Here, M_a^*, M_R^*, W_F^*, W_R^* and H_c^* are minor-loop magnetization, minor-loop remanence, minor-loop hysteresis loss, minor-loop remanence work, and minor-loop coercive force, respectively, as denoted in Fig. 1. M_s and M_R are saturation magnetization and remanence of the major loop, respectively. The relations of Eqs. (1)–(3) are valid for minor loops where irreversible movement of Bloch wall mainly contributes to magnetization. The exponents of the power laws, n_F, n_R, and n_c are nearly 1.5, 1.5, and 0.45, respectively, and are almost independent of kinds of materials. Note that the relation of Eq. (1) is known as Steinmetz law where $n_F \sim 1.6$ [30]. W_F^0, W_R^0, and H_c^0 are minor-loop coefficients independent of magnetic field and amplitude of a cyclic field. The coefficients are proportional to internal stress and more sensitive to it compared with H_c. In this study, these coefficients were used to investigate changes of internal stress in the model alloys after neutron irradiation.

3. Experimental results

To see changes of magnetic properties due to neutron irradiation, we first pay attention to major loops. Fig. 2(a)–(d) shows ma-

Table 1
Chemical compositions of measuring samples.

Sample	Cu (wt%)	Mn (wt%)	N (wt ppm)
VA			5
VB		1.0	40
VD	0.9	1.0	10
VH	0.9		20

Table 2
Neutron irradiation conditions in this study.

	Flux ϕ (10^{12} n cm^{-2} s^{-1})	Fluence ϕt (10^{19} n cm^{-2})
K2	0.77	0.55
T6	0.97	3.32

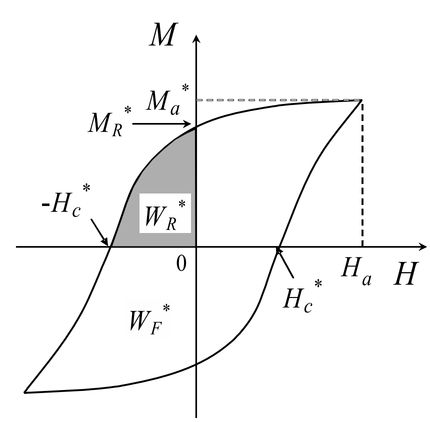

Fig. 1. Parameters of a minor loop.