ABSTRACT

*In situ* magnetic measurement on pure Fe and A533B steel was performed at $T = 563$ K under neutron irradiation in a 50 MW nuclear reactor. Magnetic properties which are sensitive to lattice defects were obtained from the minor hysteresis analysis. In pure Fe, the minor loop coefficients show a monotonic increase with increasing neutron fluence in the range of $0$ to $5 \times 10^{19}$ cm$^{-2}$, which is due to the formation of dislocation loops during the irradiation. On the other hand, in A533B steel, the coefficients increase and show a maximum at the fluence of $1 \times 10^{19}$ cm$^{-2}$, followed by the slow decrease. The appearance of the maximum suggests that Cu precipitates in the vicinity of dislocations compensate the internal stress of dislocations so as to minimize the elastic energy.

INTRODUCTION

The quantitative evaluation of degradation in nuclear pressure vessel materials has been studied by the comparison of physical properties with the irradiated and the pre-irradiated states. The number of data points is limited in the currently employed measurements to know the neutron irradiation effects. In situ measurement of physical properties is generally difficult under neutron irradiation in the nuclear reactor. The magnetic properties, however, are possible of the realization of *in situ* measurement, though the magnetic method is unpopular in the engineering and scientific fields, especially in nuclear engineering. The relation of magnetic properties and neutron radiation low alloy steels was investigated by limited number of scientists [1-3]. However, we cannot find the consistent trend in the relation of their data; it is unclear whether coercive force increases or decreases by the neutron fluence. The different trend would attribute to the limited numbers of data points. The present in situ measurement would give us a reliable answer to the different trend [4].

The relation of coercive force and dislocations has been studied in ferromagnetic materials for long time and a simple relation was obtained by the Stuttgart group of Max-Planck Institute [5,6]. Coercive force increases in proportion to the square root of dislocation density. The irradiation damages, such as cooper precipitates would make coercive force increase though the magneto-elastic coupling. The lattice defects can be observed directly or indirectly after neutron irradiation; the direct observation is due to TEM, while the indirect one is due to the magnetic method [7], the atom-probe field ion microscope [8], positron annihilation [9], small angle neutron scattering [10], electric resistance [11] etc. Only the magnetic method can be technically applied for in situ measurement during neutron radiation in the nuclear reactor. Though the coercive force should increase with the increase of neutron fluence, it decreases. Of our interest is the confirmation of the different trend of coercive force by in situ measurement. The main purpose of the present study is the investigation of the different trend in coercive force.

Recently, an analysis method of minor hysteresis loops has been found in pure Fe and low carbon steels. Minor-loop coefficients deduced from simple relations between parameters of minor loops are much more sensitive to dislocations than coercive force [12,13]. In addition, one can obtain the minor-loop coefficients with much lower magnetic field compared with that of the major loop. For instance, 1 kA/m is sufficient for minor-loop analysis of A533B steel, though we need more than 10 kA/m for major loop. Such low field measurements are very useful for the present in situ magnetic measurements because the maximum applied field is limited to 2 kA/m owing to the limited space of an irradiation capsule of ring core samples.
ANALYSIS METHOD OF MINOR LOOPS

We found seven scaling rules between the minor-loop properties; minor-loop magnetization $M_a^*$, minor-loop coercive force $H_c^*$, minor-loop remanence $M_R^*$, minor-loop hysteresis loss $W_F^*$, minor-loop remanence work $W_R^*$, minor-loop stored energy $W_a^*$ and minor-loop susceptibilities $\chi_{H, R}^*$ and $\chi_a^*$ (see Fig. 1) [12, 14]. The scaling rules include

$$W_F^* = W_F^\alpha \left( \frac{M_a^*}{M_S} \right)^{n_F},$$

(1)

$$W_R^* = W_R^\alpha \left( \frac{M_R^*}{M_R} \right)^{n_R},$$

(2)

$$W_a^* = W_a^\alpha \left( \frac{M_a^*}{M_S} \right)^{n_a},$$

(3)

and

$$H_c^* = H_c^\alpha \left( \frac{M_R^*}{M_R} \right)^{n_c},$$

(4)

where $M_S$ and $M_R$ are saturation magnetization and remanence, respectively. Minor-loop coefficients, $W_F^\alpha, W_R^\alpha, W_a^\alpha$ and $H_c^\alpha$ are independent of the magnetic field amplitude $H_a$.

Figure 1 - Parameters of a minor hysteresis loop.

![Figure 1](image1.png)

Figure 2 - (a) A ring core sample and (b) a part of irradiation capsule.

![Figure 2](image2.png)
and have a linear relation with coercive force. It was revealed that the exponents $n_F$, $n_R$, $n_a$ and $n_C$ are constant independent of lattice defects, applied stress, temperature and kinds of materials; $n_F = n_R = 3/2$, $n_a = 1$ and $n_C = 0.45$. The minor-loop coefficients give a lot of information on the microstructure including dislocations and show a monotonic increase with the increasing dislocation density. Further, the coefficients have a simple relationship with mechanical properties such as ductile-brittle transition temperature and Vickers hardness [13]. Note that the scaling law with $n_F = 1.6$ between $W_F^*$ and $M_s^*$ was discovered by Steinmetz about one century ago and is well-known as the Steinmetz law [15].

**EXPERIMENTAL PROCEDURE**

The experimental procedure was already described in detail in the reference [4]. Magnetic measurements of ring cores under neutron radiation were performed in Japan Materials Testing Reactor at Japan Atomic Energy Agency. For in situ magnetic measurements, the ring cores with exciting coil, pickup coil, and thermocouple, were installed into an irradiation capsule as shown in Fig. 2. Mineral insulated cables, which have high durability against neutron irradiation, were used for the coil. This mineral insulated cable was wound on the ring core with 100 turns, which yields a maximum applied field of 2 kA/m. The cables extended from the capsule were connected to an electronic circuit installed outside the reactor.

**EXPERIMENTAL RESULTS**

Figure 3(a) shows minor-loop coefficients, $W_F^0$, $W_R^0$ and $H_c^0$, as a function of neutron fluence in pure Fe. One can see that all coefficients increase monotonically with the increase of neutron fluence. Their increase corresponds to the increase of stress field and is consistent with the fact that the irradiation damage increases by neutron radiation. One can admit that the minor-loop coefficients have a kink around $2 \times 10^{19}$ cm$^{-2}$ and their increasing rate becomes lower above the fluence.

In A533B steel, however, the dependence of minor-loop coefficients on neutron fluence is quite different from that of pure Fe. As shown in Fig. 3(b), minor-loop coefficients, $W_F^0$, $W_R^0$ and $H_c^0$, sharply increase by neutron irradiation at first and show a maximum at the fluence of $1 \times 10^{19}$ cm$^{-2}$. With increasing the fluence, the coefficients slowly decrease and seem to have another local maximum around $3 \times 10^{19}$ cm$^{-2}$. The local maximum indicates that there exist two opposite influences on the microstructure under the neutron radiation; one is the operation to increase the internal stress and the other is that to decrease it. The later operation is unexpected since the mechanical properties increase by neutron radiation without exception. The mechanism of decreasing internal stress is discussed in the next section.

**DISCUSSION**

The neutron radiation induces several kinds of lattice defects such as vacancies, interstitial atoms, voids, dislocation loops and copper precipitates. It was revealed that the copper precipitates with the size of 2-3 nm, which are nucleated and grown by the irradiation, make materials brittle in A533B steel with high copper content. The copper precipitates would play the main role to the change of minor-loop coefficients in A533B steel.

In A553B steel, minor-loop coefficients show a maximum at the fluence of $1 \times 10^{19}$ cm$^{-2}$, followed by the slow decrease with the increase of neutron fluence. This change in the minor-loop coefficients indicates that the residual internal stress decreases and obstacles to the 180$^\circ$ wall motion become small above the fluence of $1 \times 10^{19}$ cm$^{-2}$. The contradiction would be explained by the introduction of an idea that the copper precipitates grow up in the vicinity of dislocations above the fluence.
Copper precipitates and dislocation loops in the matrix have stress field in themselves and make minor-loop coefficients increase. On the other hand, the stress field is reduced and minor loop coefficients decrease when these lattice defects exist in the vicinity of dislocations. Both radiation defects will compensate the stress field of dislocations by their stress field and the compensation brings about the decrease of minor-loop coefficients. The emulation of these two effects of stress field would result in the maximum of minor-loop coefficients. These radiation defects fixed around dislocations strongly disturb the dislocation movement and make mechanical properties increase as was observed.

The irradiation damage in pure Fe would be due to dislocation loops, vacancies and interstitial atoms. Since the size of vacancies and interstitial atoms is too small to exert the influence on 180° wall motion, only dislocation loops would play an important role for obstacles. The number and size of dislocation loops increase with the increase of neutron fluence, resulting in the monotonic increase in minor-loop coefficients. At an early stage of neutron irradiation, dislocation loops nucleate and grow up independently or combing with each other. This nucleation process brings about the monotonic increase of minor-loop coefficients. After reaching the certain critical size, however, the stress field of dislocation loops would overlap with that of dislocations and the stress fields of dislocation loops and dislocations would be compensated partially. The increasing rate of minor-loop coefficients changes around the fluence of $2 \times 10^{19}$ cm$^{-2}$.

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