Analysis of Magnetic Minor Hysteresis Loops in Thermally Aged and Cold-rolled Fe-Cu Alloys

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Abstract. Neutron irradiation causes the formation of Cu precipitate in reactor pressure vessel steel and makes the steel susceptible to rupture. In the present study, we have examined magnetic minor hysteresis loops of Fe-1wt%Cu alloy after thermally ageing at 753 K and subsequent cold rolling to elucidate the effects of Cu precipitation on magnetic properties. Minor-loop coefficients, obtained from scaling power laws between field-dependent parameters of minor hysteresis loops, decrease with ageing time and show a local maximum around 200 min, reflecting the growth of Cu precipitates with ageing. For the alloy cold-rolled after ageing, the minor-loop properties linearly increase with reduction and show a good relationship with mechanical properties such as DBTT and hardness. These observations indicate that the analysis method using magnetic minor loops can be an useful technique of nondestructive evaluation of irradiation embrittlement and subsequent deformation hardening in reactor pressure vessel steels.

1. Introduction

Formation of Cu precipitate causes embrittlement of iron-based materials such as reactor pressure vessel (RPV) [1]. Neutron irradiation enhances the nucleation and formation of nano-size defects such as Cu-rich precipitates (CRPs) and increases in matrix damage. Both CRPs and the matrix damage impede the dislocation motion, resulting in undesired changes in mechanical properties such as a decrease in toughness and ductility, and an increase in ductile-to-brittle transition temperature, hardness. Hence, the embrittlement of RPV steels is a main concern for the nuclear industry for a long-term operation of nuclear power plants.

Magnetic method using minor hysteresis loops is one of useful one to get information on lattice defects [2,3]. It was revealed that there exist some universal scaling power laws between the field-dependent parameters and the coefficients are sensitive to defect density compared with coercivity. In addition, the coefficients were obtained with very low measurement field, being advantageous for the on-site integrity assessment.

Traditionally, thermally aged Fe-Cu alloys with Cu precipitates have been extensively studied to elucidate the detailed mechanism of Cu precipitation because of great cost in time and expense for preparation of neutron irradiated RPV steel samples as well as a difficulty of the handling of the irradiated samples. In this study, we have developed the magnetic method to investigate the effects of formation of Cu precipitates and subsequent cold rolling in Fe-1wt.% Cu alloys where nanometer Cu precipitates are induced by thermal ageing.

2. Experimental

2.1 Materials

Fe-1wt%Cu alloy was annealed at 1123 K for 5 hours, followed by a water quench. The alloy was then cold rolled (CR) up to 10% reduction in order to introduce dislocations. The alloy was thermally aged at 753 K up to a desired time (0, 20, 200, 1800 min) and subsequently cold rolled up to 20%
reduction. The thermal ageing was performed to induce Cu precipitation and simulate irradiation embrittlement, whereas the subsequent cold rolling was performed to simulate the effect of plastic deformation such as due to earthquake on irradiated RPV steels during operation.

Charpy impact test and hardness measurements were performed to investigate mechanical property changes due to ageing and subsequent cold rolling. Charpy impact test was performed with a pendulum of 26.6 kG and life angle of 138.5° in the temperature range of -100~200 °C. Ductile-brittle transition temperature (DBTT) was determined from a midpoint between low toughness brittle and high toughness ductile fracture regimes in the absorption energy curve. Vickers hardness was measured with the standard indentation technique. The applied load was 300 gf and 10 indents were taken for each sample.

2.2 Magnetic measurements
We prepared samples with different sample geometries; toroid (external and internal diameters of 20 and 18 mm, respectively, and thickness of 3 mm), Charpy (10×10×55 mm), disk (φ100×10 mm). For the toroidal sample, 110-turn exciting and 114-turn detecting coils were wound around the sample, whereas for the Charpy and disk samples a magnetic yoke was used to form a magnetically closed circuit. After demagnetizing the sample, a set of magnetic minor hysteresis loops was measured with step-by-step increasing field amplitude $H_a$. The major hysteresis loop was obtained with $H_a = 5$ kA/m.

2.3 Analysis method of minor loops

Our analysis method of minor loops is based on our previous experimental results for Fe single crystals [2,3]. As shown in Fig. 1, several minor loop properties are introduced; minor-loop magnetization $B_a^*$, minor-loop coercive force $H_c^*$, minor-loop remanence $B_R^*$, minor-loop hysteresis loss $W_F^*$, and minor-loop remanence work $W_R^*$. The magnetization process before saturation can be divided conveniently into three stages. With increasing a field amplitude $H_a$ after demagnetizing the sample, $M_a^*$ shows a gentle increase in the first stage and then steeply increases in the second stage, followed by a gradual increase of $M_a^*$ toward the saturation in the third stage. The most part of magnetization proceeds in the second stage through the movement of Bloch walls. We found that there exist several scaling laws between a pair of minor-loop parameters in the first or second stage as follows.

$$W_F^* = W_F^0 \left( \frac{B_a^*}{B_i} \right)^{n_F}$$

(1)
The exponents $n_F$, $n_R$, and $n_C$ are independent of materials; $n_F \sim n_R \sim 1.5$ and $n_C \sim 0.45$. Here, Eq. (1) is well known as the Steinmetz [4]. $W_F^0$, $W_R^0$, and $H_C^0$ are minor-loop coefficients and sensitive indicators of internal stress. In this study, the behaviour of the coefficients after ageing and cold rolling was investigated in detail.

3. Experimental result and discussion

Figure 2 shows a set of minor loops before and after thermal ageing and subsequent cold rolling. Loop width of each minor loop becomes narrower after thermal ageing, whereas it largely becomes wider after cold rolling due to the increase of density of dislocations which disturb the Bloch wall movement. Figures 3(a) and 3(b) show double logarithmic plots of $W_F^* - B_a^*$ and $W_R^* - B_R^*$ curves, respectively, before and after thermal ageing and subsequent cold rolling. As is seen in the linear relationship on a double logarithmic scale, there exist a power-law relation in these curves. Least-squares fits of $W_F^*$, $B_a^*$ and $W_R^* - B_R^*$ curves to Eqs. (1) and (2), respectively, yielded minor-loop coefficients $W_F^0$ and $W_R^0$.

Figure 2. A set of minor loops for (a) $t = 0$ min and CR=0%, (b) $t = 1800$ min and CR=0% and (c) $t = 1800$ min and CR=10%. (toroid samples)

Figure 3. Double logarithmic plot of (a) $W_F^* - B_a^*$ curves and double logarithmic plot of (b) $W_R^* - B_R^*$ curves. (toroid samples)
Figure 4. (a) Dependence of $W_F^0$ on aging time for CR=0%, (b) Dependence of $W_F^0$ on rolling reduction, taken at various ageing time. (toroid samples)

Note that minor loops with $B_a$ in the range of 0.1-1.0 T were used for the fits. Since both the coefficients show a similar dependence to ageing time and rolling reduction, we will only show the behaviour of $W_F^0$.

First, we show the dependence of $W_F^0$ on aging time before cold rolling in Fig. 4(a). $W_F^0$ steeply decreases at an early stage of ageing, followed by a local maximum around 200 min. The initial decrease might be due to the relaxation of residual stress, while the local maxima reflects the domain-wall pinning due to Cu-rich precipitates [5]. With increasing rolling reduction after thermal ageing, $W_F^0$ monotonically increase for each ageing time. These behaviors were commonly observed for all the sample geometries (toroid, Charpy, and disk). These observations reflect the sensitivity of minor-loop properties to lattice defects i.e. nano-size Cu precipitates and dislocations.

Figure 5 shows the relation between $W_F^0$ and DBTT for each ageing time. $W_F^0$ is almost in a linear proportion to DBTT for all the ageing time and the relation shifts toward a higher DBTT with increasing ageing time. This shift reflects the formation and growth of Cu precipitates due to thermal ageing. Such a good relationship was also observed for the relation between $W_F^0$ and hardness.

Figure 5. Relation between $W_F^0$ and DBTT for each ageing time. (toroid samples)
4. Conclusion
We analyzed a set of minor hysteresis loops for thermally aged and cold rolled Fe-1wt%Cu alloy in order to investigate the effect of irradiation embrittlement and subsequent plastic deformation due to such as earthquake on magnetic property changes. Minor-loop properties change depending on ageing time as well as rolling reduction, and show a good relationship with DBTT and hardness, which are a sensitive indicator of irradiation embrittlement in RPV steels. These findings indicate that the magnetic method using minor hysteresis loops can be an useful technique for nondestructive evaluation of degradation in nuclear RPV.

5. References
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