Bimodal size distribution of Cu precipitates in Fe-1.0%Cu aged at 773K

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1. Introduction

The fracture toughness decrease of pressure vessel steels in nuclear reactors is associated with the changes in microstructure occurring under the combined influence of elevated temperature and neutron irradiation. One of the reasons is the formation of copper-rich precipitates and point defect clusters, which are obstacles to the movement of dislocations under stress, thereby reducing ductility and increasing yield stress. Therefore, It is important to study the behavior of Cu impurities in Fe matrix. For the thermally aged Fe-1.0% alloys, the most notable results include the evident observation of a bimodal distribution of precipitate sizes. It means that a special technique, such as the Maximum Entropy Method is necessary for the interpretation of SANS(small angle neutron scattering) data.

2. Method and Results

2.1 Experimental

The starting material for this study was a binary Fe-1.0%Cu alloy. The alloy was made through melting with pure Fe and pure Cu. It was confirmed that impurities were negligible through a chemical analysis. Plates of 10 mm in thickness were cold rolled into 1 mm and square samples (10 mm x 10 mm) were cut from the plates. After samples were solution-treated at 1,123K for 5 hrs in the vacuum condition, they were water-quenched. Samples were isothermally aged at 773K.

Vickers hardness was measured on the polished surface after aging for checking the effect of Cu precipitates growth. To measure the size and its distribution of precipitates, methods of SANS and TEM were selected. SANS measurements were performed using the instrument at HANARO. For examination in electron microscope, discs of diameter 3 mm were punched from aged samples. Thin areas were obtained by electropolishing in a Struers twin-jet electropolisher using a solution of 5% perchloric acid in methanol cooled at 228K with an applied potential of about 15V. The sizes of Cu precipitates were evaluated with the data of SANS and TEM analyses.

2.2 Mechanical Properties (Hardness)

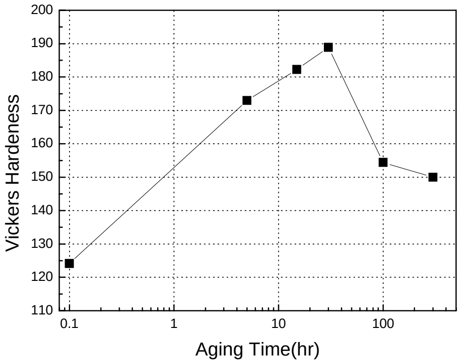


Fig.1. Change in microhardness during aging at 773K.

The increase in hardness of samples as a function of aging time at 773K is shown in Fig. 1. It shows a hardness increase by precipitation of 65 HV, after approximately 30 hours. After a comparatively short time (5 hours), the hardness has increased to a substantial amount (about 74% of maximum increase). The precipitates responsible for this initial stage must be very small since they are not detected in TEM analysis. After 30 hours, a decrease in hardness shows over-aging. The analysis of the hardness change in FeCu alloys is usually explained by using of Russel and Brown model[1]. Assuming a linear relationship between the shear stress and the hardening component due only to the precipitates H_{vp}, the expression given by Russel and Brown can be written

$$H_{vp} = \frac{Gbf^{1/2}}{R} F(R) \qquad (1)$$

where G is shear modulus of matrix, b the Burgers vector of the dislocation f the volume fraction of precipitate, R the radius of precipitate and F a function of the precipitate radius. Therefore, the smaller and the more the precipitates are, then the larger the increase of hardness becomes.

2.3 SANS

Fig. 2 shows the nuclear scattering cross sections for aged 5 hr and 30 h samples. The scattering data shows a coarsening of precipitate size distribution. The scattering intensity has following relationship.

$$\ln I(Q) \approx \ln I(0) - Q^2 R_g{}^2 / 3 \qquad (2)$$

where Q is scattering vector and R_g is the radius of gyration. When the precipitate is assumed as sphere, the radius of precipitate is

$$R_g{}^2 = \frac{3}{5} R^2 \qquad (3)$$

R_g can be determined from the slope of the Guinier plot. However, it was difficult to get the size of precipitate, because the data of samples aged over 15 hrs didn't show linear portion. To solve the problem, the indirect Fourier transformation method by Glatter[2] was used. The size of precipitate is shown in table 1.

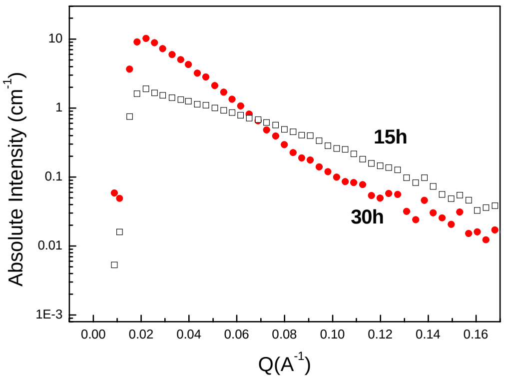


Fig. 2. Nuclear scattering cross sections for Fe1.0%Cu thermally aged for 5 and 30 h at 773K.

Table 1. Size of Cu precipitates calculated from SANS and TEM measurements.

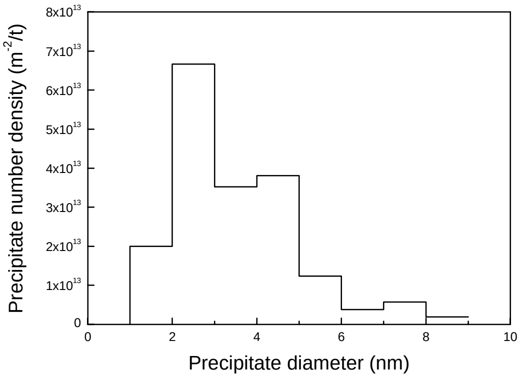
Aging Time(hr)	Size of Cu ppts (SANS), nm	Size of Cu ppts (TEM), nm
5	2	-
15	4	2(p1) 4(p2)
30	4.5	3(p1) 5(p2)
100	7	8(p1) 10(p2)
300	8	10(p1) 16(p2)

show linear portion. The size of precipitate is shown in table 1.

2.4 TEM

The sample of 5h aging has not shown any evidence of precipitation. This is thought to reflect the limit of observable size at TEM, about 2 nm. Other specimens showed bimodal distribution of precipitate size like Fig. 3 of 30 hr aging.

It has been revealed that Cu precipitates show Oswald ripening mechanism. This means that the smallest particles dissolve and the largest grow. Especially the driving force for the process is known as the diffusion of Cu atoms through vacancies**.**



coherency, transforming first to a twinned 9R structure

Fig. 3. Bimodal size distribution of Cu precipitates in Fe-1.0% Cu aged for 30 hr at 773K.

Copper precipitates show the phase transformation from a bcc structure to an fcc structure following a complicated sequence involving the formation of two intermediate structures: bcc → 9R → 3R → fcc[3]. After reaching a critical size, they lose their full coherency, transforming first to a twinned 9R structure (fcc with stacking faults) and then to 3R structure (a distorted fcc) and This can explain about the phenomenon of bimodal distribution of Cu precipitates. The critical size of bcc-9R transformation is known as about 4 nm and 9R-3R as 8 nm. In 100hr aged specimen, The fringe marks in precipitate image of 100hr aged specimen means the formation of 9R structure.

3. Conclusion

The bimodal size distribution suggests that the interpretation of SANS data needs a special explanation method such as the Maximum Entropy method. Or Lifshitz-Slyosov-Wagner method[4]. When a single distribution is assumed, the measured size of precipitates by SANS is small relatively, as shown above.

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