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Quantitative analysis of the dependence of hardening on copper precipitate diameter and density in Fe–Cu alloys

Kimihiro Nogiwa *, Nobuyasu Nita, Hideki Matsui

Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan

Abstract

In order to investigate the dependence of hardening on copper precipitate diameter and density, in situ TEM observations during tensile tests of dislocation gliding through copper precipitates in thermally aged Fe–Cu alloys were performed. The obstacle strength has been estimated from the critical bow-out angle, ϕ_c, of dislocations. The obstacle distance on the dislocation line measured from in situ TEM observations were compared with number density and diameter measured by 3DAP (three dimensional atom probe) and TEM observation. A comparison is made between hardening estimation based on the critical bowing angles and those obtained from conventional tensile tests.

1. Introduction

It is well known that the radiation-induced defect clusters in metals are responsible for significant changes in mechanical properties. Examples of extended crystal defect clusters include precipitates, voids, dislocation lines and loops. Understanding the quantitative effect of such radiation-induced defect clusters on dislocation motion is important for construction of predictive models to estimate the lifetime of power plant components. Based on Orowan's simple model, the most commonly used expression for the change in shear stress, $\Delta\tau_c$, by a regular array of obstacles is given in the following equation: $\Delta\tau_c = \alpha\mu b(Nd)^{1/2}$, where μ is the shear modulus, b the magnitude of the dislocation Burgers vector of the dislocations, N the number density of defect cluster, d the defect cluster diameter and the square-root factor is the reciprocal of the average distance between obstacles. The α factor is known as the obstacle strength and is determined as $\alpha = \cos(\phi_c/2)$. The critical bow-out angle, ϕ_c, was taken as the angle between two tangent lines drawn from the cusp in the line tension approximation. However, relatively few works have been performed to determine the α, and, in practice, microstructure and mechanical property are merely compared to infer the α for different types of defect clusters [1].

In recent molecular dynamics (MD) simulation studies [2–4], the dependence of hardening on obstacles diameter and the distance between obstacles was investigated. However, it is difficult to compare MD results with experimental observations because obstacles responsible for hardening are often too small to observe by transmission electron microscopy (TEM). In our previous study [5], the in situ

* Corresponding author. Tel.: +81 22 215 2068; fax: +81 22 215 2066.
E-mail address: nogiwa@imr.tohoku.ac.jp (K. Nogiwa).

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TEM observation technique has proven to be quite useful for quantitative estimation of the dislocation–obstacle interactions.

In this study we investigate the dependence of hardening on copper precipitate diameter and number density by performing in situ TEM observations during tensile tests in thermally aged Fe–Cu alloys. The obstacle strength α was determined at the point of dislocation break away from the obstacle. Only a single type of precipitate is assumed as the obstacle, hence the angle, ϕ_c, depends on the diameter. The number density of obstacles was determined from the dislocation segment length L between two adjacent pinning points. The spatial randomness of the pinning point separation has been taken into account by using a relationship of Foreman and Makin [6,7]. Comparison is made between number densities estimated from in situ observations and those obtained by three dimensional atom probe (3DAP) and conventional TEM observations.

2. Experimental procedure

Fe–1.0 wt%Cu alloy was prepared by arc melting followed by cold rolling. The samples for in situ tensile tests were machined to coupons of 1.0 mm × 4.0 mm × 0.2 mm and samples for 3DAP were cut into small square rods of 0.25 × 0.25 × 7 mm which were subsequently electro-polished to sharp needles. Type SSJ plate tensile specimens (16 × 4 × 0.25 mm with a 1.2 mm × 5 mm gauge section) were used for macroscopic property tests. All samples were annealed at 825 °C for 4 h and subsequently quenched into ice water. Some of the samples were thermally aged at 525 °C for 20 min, 1 h, 10 h and 100 h. The coupons (for in situ tensile tests) were electro-polished to 30–40 μm thicknesses. The thin foil specimens were prepared by electropolishing with a twinjet Struers Tenupol-3 using a polishing solution of 5% perchloric acid in ethanol cooled to −20 °C.

In situ tensile deformation experiments were performed at room temperature in a JEOL 4000FX TEM operating at 400 kV with a single tilt straining holder. The motion of dislocations was recorded by a CCD camera with a time resolution of 1/30 s. The slip planes had to be determined and the bow-out angles of dislocation should be measured in slip plane. Slip systems being activated were determined to be $\langle 111 \rangle$ $\{10\bar{1}\}$ or $\{11\bar{2}\}$. The method for determination of the actual slip system was described in detail in Ref. [5]. The α was determined by a frame-by-frame analysis of the interaction between a moving dislocation and the copper precipitates, in which the critical angle was assessed at the configuration just before the dislocation broke away. Tensile tests were carried out at a strain rate of $6.67 \times 10^{-4}\ s^{-1}$ at room temperature in air. The 3DAP measurements were performed with the energy compensated position-sensitive atom probe facilities which consist of a reflectron energy compensator, a position-sensitive detector and a high-resolution flight time detector [8]. Measurements were carried out at a sample temperature of 60 K with the pulse fraction of 0.15 under ultrahigh vacuum conditions.

3. Results and discussion

Fig. 1 shows the typical morphology of dislocations during an in situ TEM observation of Fe–1.0 wt%Cu after aging 20 min, 1 h, 10 h and 100 h at 525 °C. Dislocations are along $\langle 111 \rangle$ direction and anchored at pinning points. After 20 min and 1 h aging, copper precipitates were not observed by TEM in Fig. 1(a) and (b). On the other hand, copper precipitates were clearly observed at pinning points and in the matrix after 10 h and 100 h aging. Table 1 shows the average diameters and number densities of copper precipitates measured by 3DAP and TEM observations. The diameter was about 1 nm after 20 min and 1 h aging. In our previous study of coincidence Doppler broadening measurements [5], it is confirmed that copper precipitates are larger than about 0.6 nm after 20 min aging at 525 °C. The copper precipitates diameter increased with the aging time. The homogeneous distributions of copper precipitates were observed by TEM. It has been reported that precipitates of b.c.c. clusters are formed at first, then their transformation to 9R structure occurs when they grow beyond a critical diameter of 4 nm [9]. Therefore under the present experimental conditions, copper precipitates are deduced to be a coherent b.c.c. structure after 20 min and 1 h aging. On the other hand, it is considered that the precipitates were in the stage of transformation from b.c.c. structure to 9R structure after 10 h aging. However, it is difficult to observe ultrafine precipitates of b.c.c. structure by TEM. Therefore most of the copper precipitates observed by TEM were expected to have 9R structure and small invisible b.c.c. copper precipitates might also exist after 10 h aging. There is a peak in number density of copper precipitates at 1 h aging.

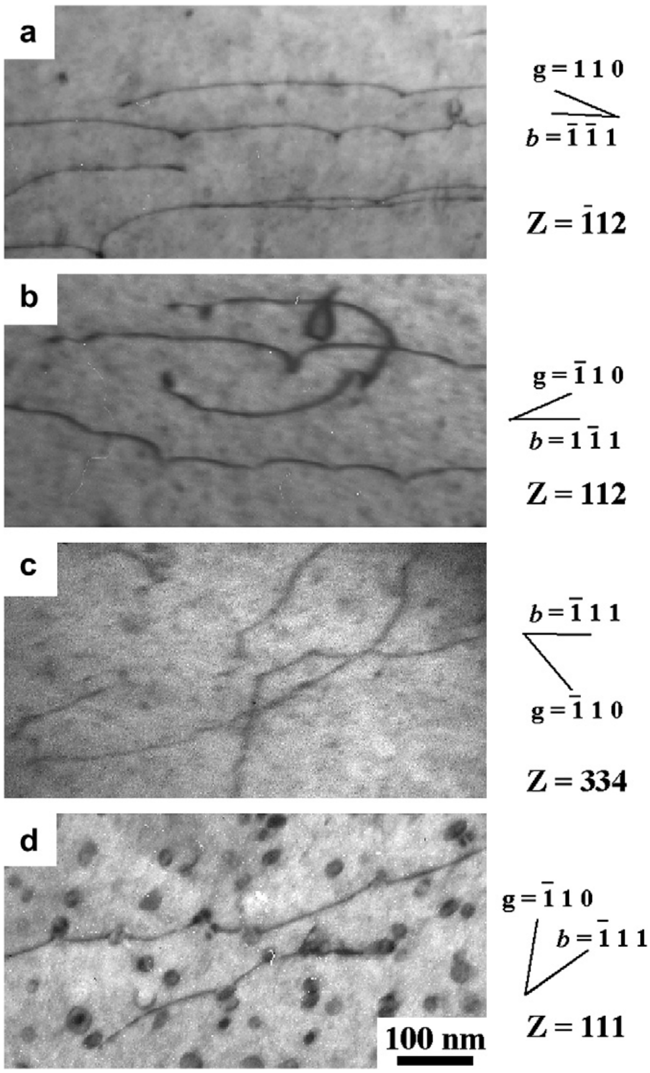

Fig. 1. The typical morphology of dislocations under stress during in situ TEM observations in Fe–1.0 wt%Cu aged 20 min (a), 1 h (b), 10 h (c) and 100 h (d) at 525 °C.

Table 1
Number density and mean diameter of Cu precipitates measured by 3DAP and TEM observations, in thermal aged Fe–1.0 wt%Cu alloy

	3DAP		TEM	
Aging time	20 min	1 h	10 h	100 h
Diameter (nm)	1.2 nm	1.2 nm	5 nm	20 nm
Number density (m^{-3})	4.5×10^{23}	6.6×10^{23}	5.5×10^{21}	1.1×10^{21}

Figs. 2 and 3 show the snapshots of in situ TEM observation of Fe–1.0 wt%Cu after 1 h and 100 h aging at 525 °C. Small defect clusters with a black dot contrast were observed after 1 h aging, which were not observed before straining. It is believed that these defect clusters are debris defects left after cross slips of dislocations. The arrows indicate where dislocations broke away from pinning points. In Fig. 2, the dislocations were pinned at copper precipitates which are too small to be observed by TEM. The larger copper precipitates pinning dislocations are clearly observed in Fig. 3. The obstacle strength was measured by dislocation with screw component, in other words, most of dislocations interacting with obstacles were of screw type.

Fig. 4 shows the histograms of the obstacle strength parameter, $\cos(\phi_c/2)$ (a) and obstacle distance, L (b). The average obstacle strength was almost same after 20 min and 1 h aging. For longer aging times of 10 h and 100 h, the average obstacle

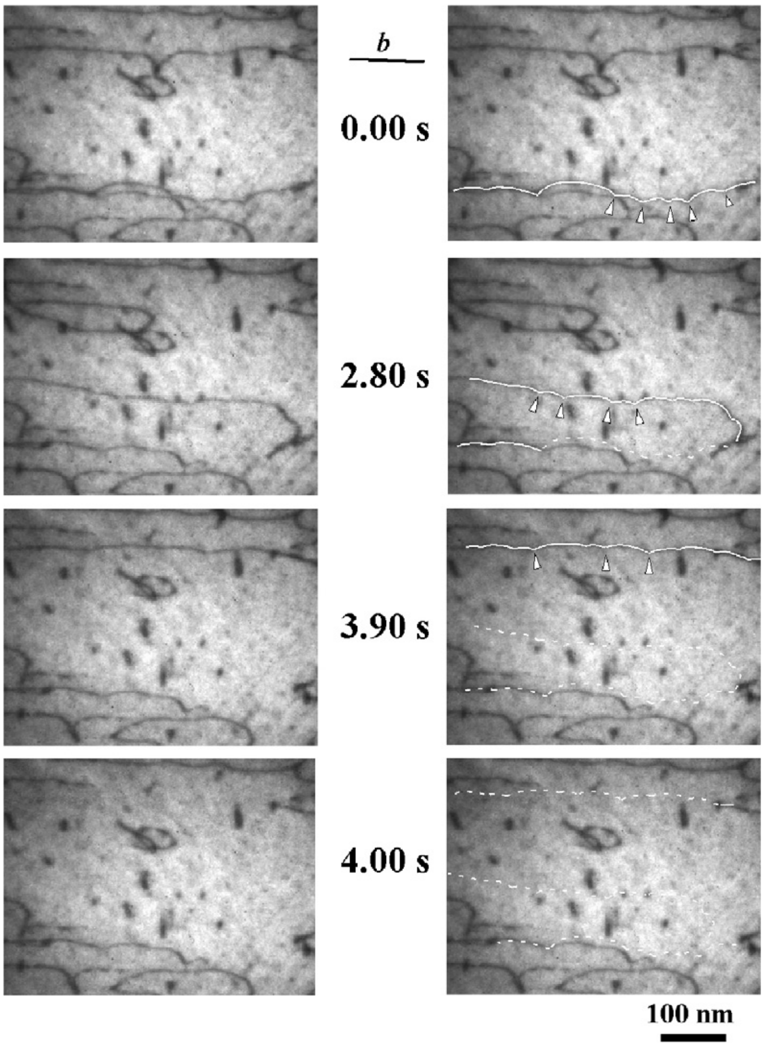

Fig. 2. In situ TEM observation of deformation dislocations in Fe–1.0 wt%Cu alloy after 1 h aging at 525 °C.

strength increased with the aging time. This behavior is similar to the aging time dependence of the average copper precipitates diameter as shown in Table 1. A broader distribution with higher obstacle strength was obtained with increasing the aging time. The obstacle strength estimated from in situ TEM observations is depended on distribution of precipitate diameter and 'the impact parameter' which is the distance from the center of an obstacle to a glide plane [2]. The situation where a dislocation penetrates the precipitate center is rather a special case, because the relative position of a precipitate to a glide plane may be arbitrary. Therefore, it is considered that the impact parameter should be always taken into account when the average obstacle strength was measured by in situ TEM observations.

The average obstacle distance is smallest at 10 h aging as shown Fig. 4(b). In contrast, the number density measured by 3DAP and TEM observations have a peak at 1 h aging as shown in Table 1. The number density is depended on obstacle distance and diameter. Assuming a square lattice array, the number density, N_v, is given by the equation:

$$N_v = 1/dL_0^2, \tag{1}$$

where d is the obstacle diameter, and L_0 is the obstacle distance. At very low stresses or when the dislocation is nearly straight, the effective obstacle spacing is much larger than the square lattice spacing. Foreman and Makin [6,7] propose the following empirical expression:

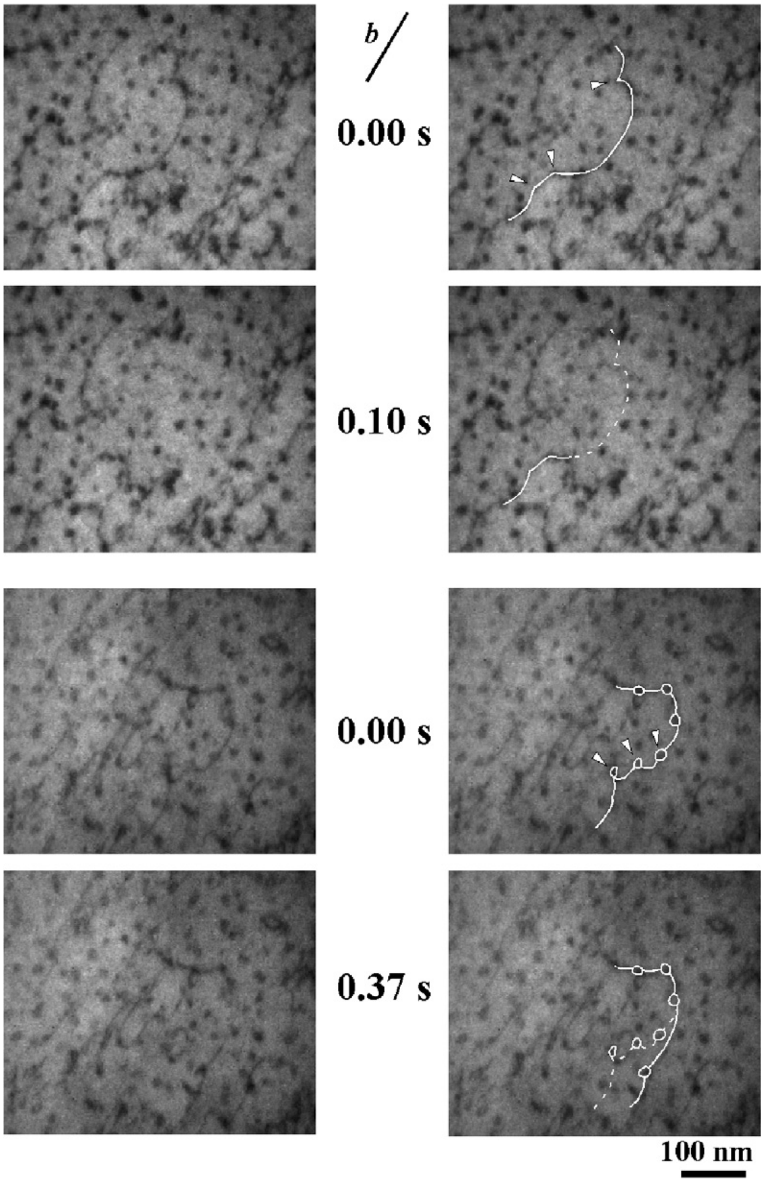

Fig. 3. In situ TEM observation of deformation dislocations in Fe–1.0 wt%Cu alloy after 100 h aging at 525 °C.

$$\Delta\tau_c = \frac{\mu b}{L_f}\cos\left(\frac{\phi_c}{2}\right) = \frac{\mu b}{L_0}\left[\cos\left(\frac{\phi_c}{2}\right)\right]^{\frac{3}{2}}\frac{4\pi+\phi_c}{5\pi}$$
$$\Longleftrightarrow L_f = L_0\left[\cos\left(\frac{\phi_c}{2}\right)\right]^{-\frac{1}{2}}\frac{5\pi}{4\pi+\varphi_c}, \qquad (2)$$

where $\Delta\tau_c$ is the increase in shear stress and L_f is the obstacle distance along dislocation line. Fig. 5 shows the number density as a function of aging time. The number density estimated from the average obstacle strength and obstacle distance using Eqs. (1) and (2) was compared with the number density obtained by 3DAP and conventional TEM observations. The number density estimated from in situ observations using random lattice array is larger than square lattice array and is good agreement with the number density measured by 3DAP and conventional TEM observations. However, the precipitate number density estimated from in situ TEM observation is larger than the TEM observations after 10 h aging. This discrepancy may be explained by underestimation of the number density measured by TEM observation. It seems that there exist both b.c.c. and 9R copper precipi-

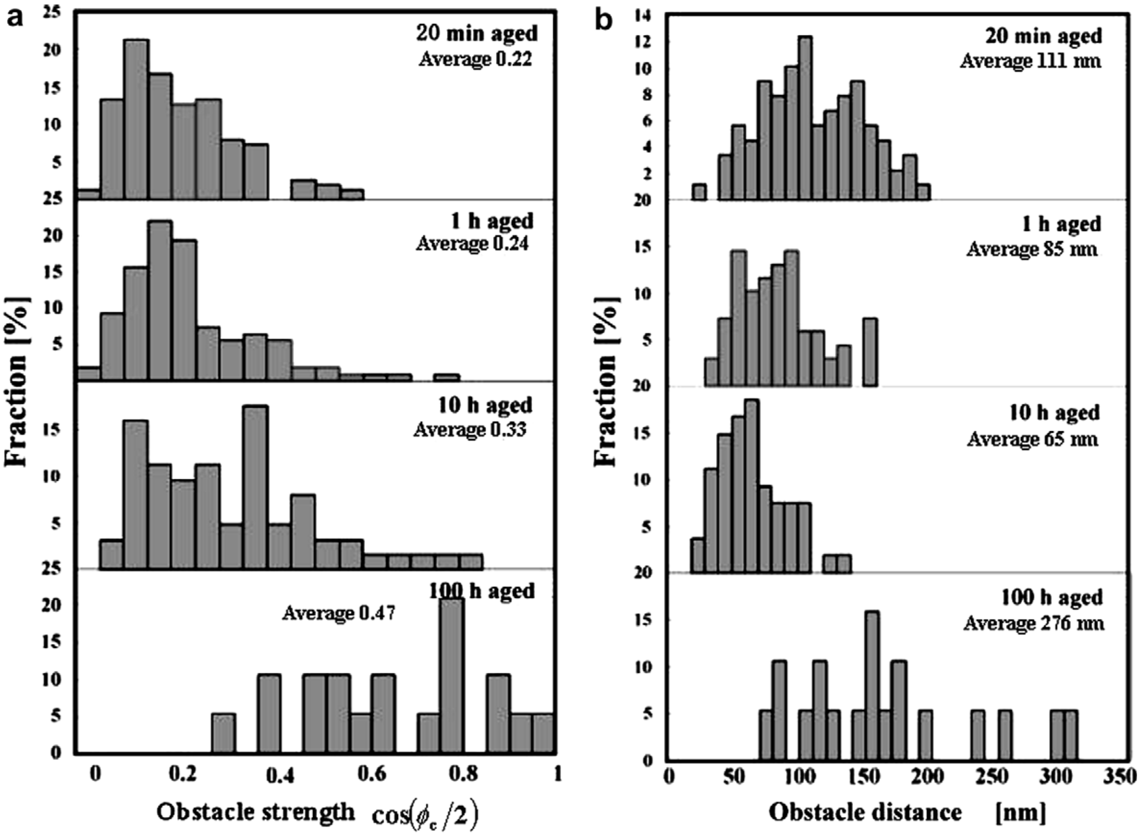

Fig. 4. Histogram of the measurement of the obstacle strength (a) and the obstacle distance (b) in Fe–1.0 wt%Cu alloy thermally aged at 525 °C.

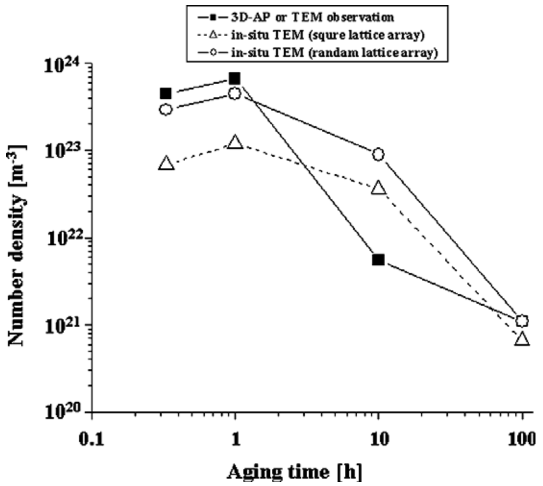

Fig. 5. Precipitate number density measured from 3DAP and TEM observations and estimated from in situ TEM observations using Eq. (1).

tates after 10 h aging. It is noted that the ultrafine copper precipitates which are too small to be observed by conventional TEM were detected by in situ TEM observation.

The relationship between the increase in shear stress and the increase in yield stress, $\Delta\sigma$, is given by following equation using the Taylor factor for polycrystals ($T = 3.06$) [10]:

$$\sigma_y = \sigma_0 + \Delta\sigma_y = \sigma_0 + T\Delta\tau_c, \tag{3}$$

where σ_y is yield stress, σ_0 is yield stress for Fe–1.0 wt%Cu alloy as quenched without precipitation hardening. Fig. 6 shows the yield stress measured by conventional tensile tests and calculated from in situ TEM observations using Eqs. (2) and (3). The yield stress by tensile tests was in qualitative agreement with that estimated from the microstructural information.

4. Conclusions

In situ TEM observation during tensile test was performed to investigate the dependence of hardening on copper precipitates diameter and number

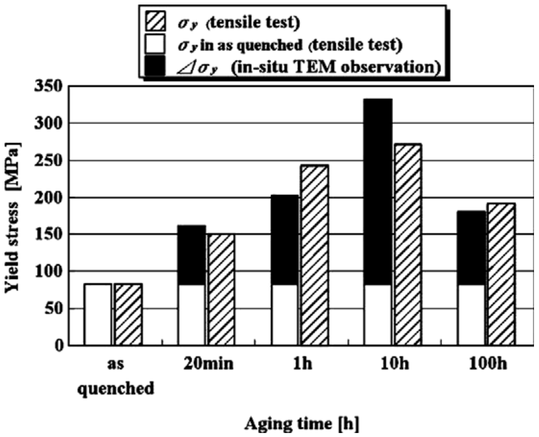

Fig. 6. Precipitate number density measured from 3DAP or TEM observation and estimated from in situ TEM observations. The yield stress measured from tensile tests and values calculated from the experimentally obtained microstructural data, using Eqs. (2) and (3).

density in thermally aged Fe–Cu alloys. A measurement of obstacle strength and distance were used to calculate the number density and the increase in shear stress.

The obstacle strength increased with the obstacle diameter. Further, the distribution of obstacle strength was broader with increasing the obstacle diameter. These results experimentally support the importance of the impact parameter.

The number densities obtained by in situ experiments were in a qualitative agreement with that measured by 3DAP and conventional TEM observations. The increase in yield stress estimated from in situ observation was in a qualitative agreement with the yield stress measured by tensile test.

Acknowledgments

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