Grain Size Variation during Low Temperature Creep and Tensile Deformation of Ultrafine-Grained Copper

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Creep and tensile tests at temperatures between 298 and 393 K were conducted on ultrafine-grained (UFG) copper processed by equal channel angular pressing (ECAP), accumulative roll-bonding (ARB) and high-pressure torsion (HPT). It is known that UFG metals show some unique mechanical properties that do not appear in coarse-grained (CG) metals, such as deviation from the Hall-Petch relationship, cyclic softening and inverse temperature dependence of activation volume. However, the mechanisms to cause these phenomena have not been fully understood yet.

In general, steady-state deformation behavior under constant stress and strain rate has been investigated to discuss the deformation mechanism. For this purpose, either stress (for creep) or strain rate (for tensile tests) is fixed and a rate equation of the form

\[ \dot{\varepsilon}_s = g(\sigma_s, T, S, K) \]  

is discussed. In eq. (1), \( \dot{\varepsilon}_s \) is the steady-state strain rate, \( \sigma_s \) the steady-state stress, T test temperature, S state variables to characterize microstructure and K the material constants. In the present study to treat UFG Cu, grain size is considered to be the most important variable of S. Although grain size is commonly regarded to be unchanged during deformation, recrystallization for UFG metals occurs much more easily than for CG metals. That is, grain size of UFG metals can change during deformation, in particular, at high temperatures. Blum et al. stated that grain coarsening during deformation of UFG Cu leads to increase in creep rate or decrease in flow stress. Thus, grain size variation needs to be taken into account in order to reveal the deformation mechanism.

In this study, creep and tensile tests of UFG Cu were conducted at relatively low temperatures, and grain size was measured by electron backscattering diffraction (EBSD) technique before and after these mechanical tests. From the experimental results, the deformation mechanism of UFG Cu will be discussed.

2. Experimental Procedure

CG Cu (99.99% purity, oxygen-free high-conductivity) rods were cut into cylinders of 10 mm diameter and 60 mm length and annealed at 873 K for 1 h. Each cylinder was subjected to 8 passes of ECAP via route Bc (rotation of the specimen by 90° in the same direction after every pass). After the ECAP deformation, the cylinder was cut into bone-shape specimens with 10–20 mm gage length and 3 × 1 mm2 cross section so that the specimen plane became parallel to the z-x plane in Fig. 1. These specimens will be referred to as as-ECAP Cu.

Creep tests were conducted under constant tensile stresses \( \sigma \) between 100 MPa and 350 MPa at various temperatures \( T \) between 323 and 393 K. To attain the constant-stress condition, applied tensile load \( P \) is controlled as \( P = P_0 / (1 + \varepsilon) \), where \( P_0 \) and \( \varepsilon \) are initial load and plastic tensile strain, respectively. Tensile tests were conducted by using an Instron-type testing machine (Minebea TG-50kN) with strain rate \( \dot{\varepsilon} \) of 2.42 × 10⁻⁵–1.69 × 10⁻² s⁻¹ at 298, 323 and 373 K. In these tests, strain gages (KYOWA KFG) were mounted on the specimen gage section to measure the true strain rate and temperature was controlled within ±1 K in an oil bath.

Fig. 1 A schematic of an ECAP die and specimens. The x direction is parallel to the extrusion direction.
As will be mentioned in section 3.2 and 3.3, steady-state deformation behavior with constant stress $\sigma_s$ and strain rate $\dot{\varepsilon}_s$ appeared during both creep and tensile tests. Therefore, mechanical tests were stopped when the steady-state deformation is reached. Grain morphology and grain size after deformation were measured by electron backscatter diffraction (EBSD) technique using a JSM-7001F (JEOL) scanning electron microscope with a field emission type gun. Observed direction of EBSD was parallel to the $y$-axis in Fig. 1.

3. Results

3.1 Grain size distribution of UFG Cu before deformation

Figure 2 shows a boundary map (a) and its trace image (b) obtained by the EBSD analysis on as-ECAP Cu. In Fig. 2(a), green lines are boundaries with misorientation angles (orientation gap between two measurement points) $\theta$ larger than 15 degrees (high angle grain boundary; HAGB) and red lines are those with $2^\circ < \theta < 15^\circ$ (low angle grain boundary; LAGB). Although the boundaries in Fig. 2(b) do not contain sub-boundaries with $\theta < 2^\circ$ because of the detection limit of EBSD, all the boundaries with $\theta > 2^\circ$ are regarded as grain boundaries (GBs) for the sake of convenience.

Grain size $w$ was measured by image processing where the two-dimensional shape of a grain was approximated by an ellipse and $w$ was defined as the length of the minor axis of the ellipse. Generally speaking, many kinds of uncertainty exist in determining "grain size" unless all the grains are the same in shape and equal in size. For the purpose of understanding one origin of the uncertainty which comes from the grain size distribution per se, we have considered both number-weighted and area-weighted averages in this study. Black solid curves in Fig. 3 show number-weighted and area-weighted cumulative fractions $F(w)$ based on the measurement of as-ECAP Cu. Here, the number-weighted cumulative fraction was defined as $F(w) = N(w)/N_0$ where $N(w)$ and $N_0$ are respectively the number of grains with its minor axis smaller than $w$ and that of all the grains. Similarly, the area-weighted cumulative fraction was defined as $S(w)/S_0$

$$f(w) = \frac{1}{\sqrt{2\pi s(w/w_0)}} \exp \left[ -\frac{\ln(w/w_0) - m}{2s^2} \right].$$

where $w_0$ is a constant to make $w$ non-dimensional and $m$ and $s$ are characteristic parameters in the log-normal distribution. By assigning $w_0 = 1$ nm, the cumulative function $F(w)$ derived from $f(w)$ can be expressed as

$$F(w) = \frac{1}{2} \text{erfc} \left[ -\frac{\ln(w/w_0) - m}{\sqrt{2}s} \right],$$

where erfc is the complementary error function. Green and red dashed curves in Fig. 3 were obtained from eq. (3) with $m = 5.62$ and $s = 0.443$ (for the green curve) and with $m = 5.99$ and $s = 0.459$ (for the red curve). The good agreement between experimentally measured curves (black solid curves) and those (green and red dashed curves) based
on eq. (3) indicates that the grain size distribution of the present as-ECAP Cu can be well represented by log-normal distributions. Therefore, the average grain size was evaluated from

\[
\bar{w} = \int_{0}^{\infty} w f(w) dw = w_0 \exp\left(\frac{m + s^2}{2}\right). \tag{4}
\]

We find \( \bar{w} = 444 \text{ nm} \) for the area-weight average and \( \bar{w}_N = 304 \text{ nm} \) for the number-weighted average, as shown in Fig. 3.

It is well known that the area-weight average is larger than the number weighted average.\(^{22,23}\) Furthermore, it has been pointed out\(^{23}\) that the average grain size determined by the so-called Heyn intercept method\(^{24}\) is slightly larger than the area-weighted average grain size. Therefore, although typical grain size of 200–300 nm\(^{25–27}\) for UFG Cu processed by ECAP is closer to the number-weighted average \( \bar{w} = 304 \text{ nm} \), it seems reasonable to deal with the area-weighted average \( \bar{w} = 444 \text{ nm} \). In any case, since \( \bar{w} \) and \( \bar{w}_N \) differ considerably, care must be taken to choose physically meaningful “average grain size”\(^{28,29}\) depending on the purpose of a particular research.

As mentioned above, the grain size \( w \) was evaluated by taking all the GBs with \( \theta > 2^\circ \) into account. Meanwhile, when another grain size \( d \) surrounded by only HAGBs (\( \theta > 15^\circ \)) is considered, it is naturally larger than \( w \). As can be seen from Fig. 2(a), it is rather difficult to approximate the two-dimensional grain shape surrounded by HAGBs as ellipses. Therefore, after careful observation and measurement of about four hundred HAGBs, \( d = 2.4\bar{w} \) and \( d = 2.4\bar{w}_N \) were obtained where \( d \) is the average grain size surrounded by only HAGBs.

### 3.2 Creep tests

Figures 4(a) and (b) respectively show creep strain–time (\( \varepsilon - t \)) curves and creep rate–time (\( \dot{\varepsilon} - t \)) curves at \( T = 373 \text{ K} \). In Fig. 4(b), the minimum strain rate in each curve was regarded as the steady-state strain rate \( \dot{\varepsilon}_s \). It can be seen from Figs. 4(a) and (b) that steady-state creep is attained before the creep strain reaches 0.05. Figure 5 shows a double logarithmic plot of \( \dot{\varepsilon}_s \) against \( \sigma_s \). When the relationship between \( \dot{\varepsilon}_s \) and \( \sigma_s \) is represented by a power law \( \dot{\varepsilon}_s \propto \sigma_s^{n} \), the data points at a given temperature lie on a straight line and the stress exponent \( n \) becomes

\[
n = \left( \frac{\partial \ln \dot{\varepsilon}_s}{\partial \ln \sigma_s} \right)
\]

as the slope of the straight line. Obtained \( n \) values are also shown in Fig. 5. We find that \( n \) decreases with increase in \( T \) except the data for \( \sigma_s \) smaller than 200 MPa and \( T = 373 \text{ K} \). Although these values of \( n \) are much larger than that \( (n = 4.8^{(13)}) \) of CG Cu at elevated temperatures, such large values have also been obtained for UFG Cu in other studies.\(^{14,30}\) In contrast, for \( \sigma_s \) smaller than 200 MPa and \( T = 373 \text{ K} \), the value of \( n \) was 3.35 and is closer to that of CG Cu.

### 3.3 Tensile tests

Figure 6 shows true stress-true strain curves of tensile tests at \( T = 373 \text{ K} \). The fixed values of strain rate are shown in the figure. Similar to the creep tests, steady-state deformation with constant stress \( \sigma_s \) appeared before plastic strain reaches 0.05 in all the tensile tests. Figure 7 shows a double logarithmic plot of \( \dot{\varepsilon}_s \) against \( \sigma_s \) obtained from the tensile tests. The values of \( n \) are even larger than those for the creep tests (Fig. 5).
3.4 Activation energy of deformation

From the temperature dependence of steady-state creep rate and the temperature and strain-rate dependence of flow stress, plastic deformation of UFG Cu is known to be thermally activated. The activation energy (though more strictly, the activation enthalpy) $Q$ of deformation is obtained from

$$Q = -R \cdot \left\{ \frac{\partial \ln \dot{\varepsilon}_s}{\partial (1/T)} \right\}_{\sigma_s},$$

where $R$ is the gas constant. From Figs. 5 and 7, the variation of $\dot{\varepsilon}_s$ with $1/T$ at a constant stress $\sigma_s$ can be evaluated and the activation energy $Q$ can be estimated. Figure 8 shows the obtained relationship between $Q$ and $\sigma_s$. Only one $Q$ value could be obtained from the tensile test data for $\sigma_s = 410$ MPa in Fig. 7. $Q$ values obtained by Li et al.\textsuperscript{30)} are also shown in Fig. 8. Although $Q$ tends to decrease with increase in $\sigma_s$, there exists a plateau range with values around 80–90 kJ mol$^{-1}$. Interestingly, these $Q$ values are close to the activation energy of GB self-diffusion for Cu (84.8 kJ mol$^{-1}$).\textsuperscript{33)} This will be discussed again in Section 4.

3.5 Grain size variation after deformation

It is generally known in CG metals that the size of subgrains formed by deformation is inversely proportional to the applied stress.\textsuperscript{34,35)} The grain size $w$ in this study includes the contribution of subgrains formed by deformation. With this in mind, Fig. 9 shows area-weighted average grain size $w$ measured after creep and tensile deformation, as a function of $G/\sigma_s$, where $G$ is the shear modulus of Cu. It can be seen that the data points for larger $\sigma_s$ values (larger than about 200 MPa) lie on a straight line drawn from the origin, i.e.,
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where \( k_w \) is a dimensionless constant and \( b \) is the length of the Burgers vector. With \( G = 47.1 - 0.0168 \cdot (T/K) \) GPa and \( b = 0.256 \) nm, the slope of the straight line becomes \( k_w = 11 \). This value of \( k_w \) is considered reasonable since values such as 10\(^{30}\) or 14\(^{37}\) were reported previously. If the number-weighted average grain size \( \bar{w} \) were adopted in the left-hand side of eq. (7), the dimensionless constant becomes \( k_{wN} \approx 7 \) and the agreement with previous studies becomes less satisfactory.

In the lower \( \sigma_s \) range (smaller than about 200 MPa), on the other hand, \( \bar{w} \) is nearly equal to that of as-ECAP Cu (444 nm). This means that the grain size did not change during creep under the lower stresses. Figure 10 shows the boundary map of annealed as-ECAP Cu for 10\(^5\) s at 373 K without the application of stress. As can be seen, stress-free annealing caused extensive grain coarsening. This in turn means that deformation suppressed grain coarsening. In this respect, Miyajima et al. suggested that dynamic recovery during deformation decreases dislocation density and leads to the reduction of the driving force of grain coarsening.\(^{38}\) Therefore, it is concluded that \( \bar{w} \) under lower stresses deviates from eq. (6) because of dynamic recovery. In addition, we found that \( d = 2.4w \) is also true after deformation (Section 3.1).

4. Discussion

Figure 11 shows the relationship between \( \dot{\epsilon} \) and \( \sigma_s \) at \( T = 373 \) K. The data of other previous studies\(^{14,37,39}\) are also included. As can be seen, the present data of open and filled circles are not only in good agreement with those of previous studies but also cover much wider stress ranges.

To explain the steady-state deformation of UFG metals quantitatively, Blum and Zeng have proposed a model (B-Z model).\(^{40-42}\) In the B-Z model, creep deformation of UFG Cu is considered to be controlled by the climb of dislocations accumulated near GBs. Their rate equation is expressed as follows.

\[
\dot{\epsilon} = \frac{AG\delta D_{GB}}{k_B T} \left( \frac{d}{b} \right)^4 \left( \frac{\sigma_s}{G} \right)^8 f(\xi),
\]

where \( \delta \) is the thickness of GBs, \( D_{GB} \) the coefficient of GB self-diffusion, \( k_B \) the Boltzmann constant, \( \nu \) the Poisson ratio, \( M \) the Taylor factor, \( c \) and \( \alpha \) dimensionless constants with values from zero to unity and \( h_{spon} \) the maximum athermal spacing which is necessary for spontaneous annihilation of edge dislocations at HAGBs. The data in Fig. 11 were analyzed using the B-Z model. In eq. (7)–(10), \( c, \alpha \) and \( h_{spon} \) can be handled as adjustable parameters. Even if \( d \) and \( \alpha \) are fixed, these parameters affect \( \dot{\epsilon} \) sensitively.\(^{41}\) The black solid curve in Fig. 11 is drawn based on the B-Z model. The assigned values are shown in Table 1. For \( d \), the results shown in Fig. 9 were used together with \( d = 2.4w \). On the other hand, the black dashed curve in Fig. 11 was calculated by assigning a fixed value of \( d = 1.1 \) µm in the B-Z model. It can be seen that both solid and dashed curves reproduce \( \dot{\epsilon} \) for \( 200 \) MPa < \( \sigma_s < 300 \) MPa. This reproducibility is naturally understood since the adjustable parameters were so chosen as to fit the experimental data points of this stress range. However, since the B-Z model gives a nearly linear relationship between \( \ln \dot{\epsilon} \) and \( \ln \sigma_s \), the deviation from the data points becomes apparent for \( \sigma_s < 200 \) MPa and \( \sigma_s > 300 \) MPa.

As mentioned above, it is found that the B-Z model can reproduce the creep behavior of UFG Cu only for an intermediate stress range, i.e., \( 200 \) MPa < \( \sigma_s < 300 \) MPa in the present analysis. Interestingly, this stress range corresponds to that of the plateau range of the activation energy in...
and is still smaller than that of lattice diffusion for Cu ($Q_\text{L} = 197\text{kJ mol}^{-1}$). Although it is difficult to identify the deformation mechanism at the lower stress range, it can be said that both relatively larger grain size and larger activation energy suggest the increasing contribution of in-grain recovery-controlled processes. The small value of stress exponent is also consistent with this interpretation. Further study is necessary to reveal the steady-state deformation mechanisms of UFG Cu.

### 5. Conclusions

Creep and tensile tests under various temperatures and stresses were conducted on UFG Cu processed by ECAP, and the grain size after deformation was measured by the EBSD technique. The following conclusions were obtained.

1. Under all the present deformation conditions, the steady state of plastic deformation, i.e., minimum strain rate for stress-fixed creep tests and constant stress for strain-rate-fixed tensile tests, was realized before the plastic deformation reaches 0.05.

2. When the deformation behavior is described as power-law creep, the stress exponent of UFG Cu was generally much larger than that of CG Cu. In addition, the stress exponent increased with increase in applied stress.

3. The activation energy of deformation is a gradually decreasing function of applied stress with a plateau at an intermediate stress range of 200 MPa $<$ $\sigma_s$ $<$ 300 MPa.

4. At 373 K, grain size after creep deformation decreased with increase in the applied stress. However, when applied stress is lower than 200 MPa, gain size remained constant during creep.

5. It is suggested that three deformation mechanisms of UFG Cu are operative depending on the stress level: in-grain recovery-controlled creep at a lower stress range of $\sigma_s$ $<$ 200 MPa, a climb-controlled process of dislocations at GB at an intermediate stress range of 200 MPa $<$ $\sigma_s$ $<$ 300 MPa, and a dislocation glide-controlled process at a higher stress range of $\sigma_s$ $>$ 300 MPa.

### Acknowledgements

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### REFERENCES


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**Table 1** Parameter values used to apply the B-Z model.

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<th>Parameter</th>
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<td>$\delta D_{\text{eq}}$/m$^2$s$^{-1}$</td>
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