Low Frequency Internal Friction Study of Sintered Ultra-Fine Aluminium Products

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The internal friction of sintered ultra-fine aluminium products (SUFAP) has been measured in an inverted torsion pendulum apparatus. A relaxation peak of a thermal activation type is observed at about 400 K. A possible mechanism for the peak is suggested; grain boundary sliding of fine grains of aluminium with some contribution of the diffusional relaxation along oxide or nitride particles. The results are compared with a mechanical property of SUFAP measured in a previous study.

(Received July 25, 1987)

Keywords: internal friction, anelasticity, grain boundary, sintered aluminium powder, mechanical properties

I. Introduction

The internal friction of Al–Al₂O₃ dispersion-strengthened alloys (SAP: sintered-aluminium powder) has been studied by many investigators(1)-(6). Although several internal friction peaks have been observed above room temperature, the obtained results differ among different investigators. The peaks have been interpreted by the following two mechanisms:

1) The relaxation due to grain boundary sliding(4)(5)
2) Dislocation damping(6)

Recently, Tsunekawa, Suzuki and Takei(7) have studied mechanical properties of a sintered aluminium alloy prepared from ultra-fine aluminium powder of about 0.1 μm in diameter; they called the material SUFAP (sintered ultra-fine aluminium powder). It has been found that the alloy exhibits a large elongation when deformed at temperatures around 450 K; which is interpreted as due to intergranular sliding. It seems interesting to measure the internal friction of such materials, since the behaviour of grain boundaries can be sensitively monitored.

In the present paper we report the result of low frequency internal friction measurements of SUFAP specimens. It is aimed to clarify the nature of the internal friction of SUFAP and to investigate the possible relationship between the internal friction and mechanical properties.

II. Experimental

Four kinds of ultra-fine aluminium powder (about 0.1 μm in diameter) produced by Vacuum Metallurgical Co. were sintered for 3.6 ks(1 h) in vacuum at 863 ~ 873 K. Sintered rods were rolled at 723 K, annealed at 723 K for 21.6 ks (6 h) and cut into final form (1 × 1 × 70 mm³) for internal friction measurements. Specimens contain impurities of Cu, N and O which entered during the production of the ultra-fine powder. The chemical analysis of specimens is given in Table 1. In this investigation, we used the same specimens as those used in the previous experiment on mechanical properties of SUFAP; samples 1, 2, 3 and 4 correspond to samples A, B, D and E in the previous study(7), respectively. A part of the data obtained in the previous study is also listed in the table; εₘₜₜ is the maximum fracture strain at about 450 K. The detailed description of the
preparation of the specimen and mechanical properties were given in the previous paper.

The internal friction was measured in an inverted torsion pendulum apparatus over the temperature range from 100 to 700 K. Heating rate was about 1 K/min and the measuring frequencies were between 0.5 and 10 Hz. The strain amplitude in the maximum surface shear strain was about $1 \times 10^{-5}$.

### III. Results

Figure 1 shows the internal friction and the vibrational frequency of a specimen 1 as a function of temperature. An internal friction peak is observed at about 400 K. The corresponding change of the vibrational frequency indicates that the peak is of a relaxation type.

Figure 2 shows the internal friction and the relative change of the rigidity modulus, $G/G_u$, of specimens listed in Table 1. $G$ is the rigidity modulus; the square of the measuring frequency, $f^2$, is proportional to $G$. $G_u$ is the unrelaxed modulus which is determined from an extrapolation of the linear part of the rigidity modulus (or frequency)-temperature curve below 250 K. All the internal friction vs temperature curves show maxima at about 400 K. Since these peaks are overlapped with an increasing background or a tail of another higher temperature peak, it is difficult to determine the position or the height of the peak. Therefore, we determined the peak temperature of the relaxation and the relaxation strength from the temperature derivative of the modulus is narrower than the peak of the internal friction. This is true even when the process involves multiple relaxation times. Therefore, the location of the peak and so the relaxation time can be more easily determined for the former.

(2) By taking the temperature derivative, the effect of high temperature background can be minimized.

(3) For a single relaxation process, the relaxation strength is estimated either from the height of internal friction peak or from the relative difference between the relaxed- and unrelaxed modulus. For a multiple relaxation process accompanied by a high temperature background, the relaxation strength is better estimated from the height of internal friction peak or from the relative difference between the relaxed- and unrelaxed modulus. For a multiple relaxation process accompanied by a high temperature background, the relaxation strength is better estimated from the height of internal friction peak or from the relative difference between the relaxed- and unrelaxed modulus.

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**Table 1** Chemical analysis, the maximum fracture strain and results of internal friction measurements.

| Specimen | Chemical analysis (mass%) | $e_{fs}$ | $\Delta$ | $E$ (eV) | $-\log \tau_0$ (s) |
|----------|---------------------------|---------|---------|--------|------------------|
| 1        | 2.01 2.42 4.90            | 32~38   | 0.132   | 1.5±0.3 | 20±3             |
| 2        | 1.94 4.85 0.25            | −8      | 0.064   | 1.2±0.3 | 18±3             |
| 3        | 0.58 4.54 0.41            | −8      | 0.055   | 1.2±0.3 | 18±2             |
| 4        | 0.24 6.43 12.1           | −1      | 0.051   | 1.0±0.1 | 15±1             |

† Ref. (7). Data of $e_{fs}$ were obtained with strain rate of $2.1 \times 10^{-3}$ s$^{-1}$ at 450 K.
evaluated by integrating the peak area of the temperature derivative curve of the modulus (i.e. the vibrational frequency). The relaxation strengths thus evaluated are listed in Table 1.

Similar measurements to those in Fig. 2 were made at different frequencies. Peak positions are plotted as a function of the measuring frequency in Fig. 4, together with the results for the peaks in pure aluminium and SAP reported by previous investigators. The activation energy, $E$, and the pre-exponential factor of the relaxation time, $\tau_0$, are determined from the figure, and are also listed in Table 1.

Fig. 2 Temperature dependence of (a) $G/G_0$ and (b) internal friction of specimens listed in Table 1.

Fig. 3 Temperature dependence of $-d(G/G_0)/dT$ of specimens listed in Table 1.

Fig. 4 Frequency dependence of peak temperature, $T_p$, for SUFAP specimens (1, 2, 3, 4). The results for SAP (4 mass% Al$_2$O$_3$) and pure aluminium in the literature are also shown. SAP: G-Gelli$^{29}$, S-Seemann$^{30}$, MP-Mezzetti and Passari$^{40}$ (two kinds of specimens were used; CW-cold worked, EX-extruded), GTE-Gondi et al.$^{6}$ (four peaks, P1, P2, P3 and P4, were observed). Pure Al: K-Ke$^{10}(11)$, CS-Cordea and Spretnak$^{12}$, WL-Williams and Leak$^{13}$. 
IV. Discussion

1. Mechanism of the 400 K peak

Since the present specimens contain a fairly large amount of copper (0.25 ~ 12.1 mass%), we will first discuss the possible effect of this element on the relaxation spectrum. Several investigators have measured the internal friction of Al–Cu alloys:

1) In an Al–4 mass%Cu alloy, Berry and Nowick(14) found a peak at 450 K (~1 Hz). This peak was observed in an as-quenched state, and its height decreased on aging; the peak was interpreted as the Zener relaxation(15). On aging, a new broad peak is developed at 410 K, which was referred to as the $\delta'$-peak; the peak was considered to be associated with the transitional $\delta'$ phase. On further aging at higher temperatures, both peaks disappeared.

2) Lei(16) investigated the effect of the addition of copper (0.09 ~ 3.08 mass%) on the grain boundary peak in Al. The peak decreases with increasing the Cu content, whereas the peak temperature (about 500 K for 1 Hz) remains unaffected.

3) For an Al–3.4 mass%Cu, Kiss et al.(17) found a "pseudo peak" around 750 K, and suggested that the peak might be related to the dissolution of precipitates. For this concentration the grain boundary peak is completely suppressed.

In the present experiment, all the specimens were subjected to annealing at 723 K for 21.6 ks (6 h) prior to measurements. Most of copper atoms are considered to exist as CuAl$_2$ precipitates segregated at the surface of a "striation structure", as reported in the previous paper. Therefore, the 400 K peak observed in the present experiment does not seem to correspond to any of the peaks described above. As seen in Fig. 2(b), copper seems to have an effect on the high temperature background rather than the 400 K peak. In what follows we shall discuss the origin of the 400 K peak, assuming that the effect of copper can be neglected.

Before discussing the mechanism of the 400 K peak, it seems pertinent to review briefly the present status of understanding of the origin of the grain boundary peak. In 1947, Ke(10)(11) first observed a large relaxation peak in polycrystalline aluminium but not in a single crystalline specimen. The peak was well explained by the model of the grain boundary sliding proposed by Zener(18) and was called "the grain boundary peak". Since then, the peak had long been believed to be due to the relaxation associated with grain boundaries. However, Woirgard et al.(19) threw a doubt on such an interpretation, on the basis of the observation that similar peaks appear in several pure fcc metals when single crystalline specimens are slightly deformed. These peaks were explained to be due to lattice dislocations. Many other experiments(20)-(27) also supported such an explanation. On the other hand, Iwasaki(28) recently found that the peak consists of two different components which are overlapped each other in the same temperature range. The components were interpreted as due to the grain boundary sliding and the movement of lattice dislocations. He pointed out that the conflict on the grain boundary peak arose from a lack of recognition of the existence of the two components.

Thus, although the agreement on the origin of "the grain boundary peak" has not yet been fully achieved among the researchers, it seems reasonable to consider that the grain boundary does contribute to some extent to the grain boundary peak of the internal friction. In this understanding we discuss the origin of the 400 K peak below.

The peak temperatures for relaxation peaks observed for SAP and pure aluminium are plotted as a function of measuring frequencies in Fig. 4. A set of data characterizing these peaks are listed in Table 2. Although the peak temperatures for the same frequency range are widely different, the slope of the lines in the figure or the activation energies are not much different, which suggests that the relaxation processes are controlled by the basically same mechanism. We shall consider the source of the wide variation of the peak temperatures.

The important differences in the microstructure of various specimens are the grain size and the presence of oxide particles along grain boundaries. The effect of the second phase par-
particles on the grain boundary sliding was theoretically investigated by Mori et al.\(^{(30)(31)}\). The particles suppress the boundary sliding through the internal stress due to blocking effect. According to their analysis, the relaxation time and the relaxation strength are given by

\[
\tau = \alpha \tau_R(a/a_R), \\
\Delta = \alpha \Delta_R,
\]

where \(\tau_R\) and \(\Delta_R\) are, respectively, the relaxation time and the relaxation strength of a particle-free specimen with grain diameter, \(a_R\); the suffix "\(\text{R}\)" means "reference", since we will evaluate \(\tau\) and \(\Delta\) of relevant specimens with those of pure aluminium as the standard. \(\alpha\) is a geometrical factor given by

\[
\alpha = \left(1 + \pi a r / \lambda^2\right)^{-1},
\]

where \(a\) is the radius of grain, \(\lambda\) is the interparticle distance and \(r\) is the average radius of the blocking particles. Equation (1) shows that the blocking effect reduces \(\tau\) and \(\Delta\) by the factor \(\alpha\) but the associated activation energy remains unchanged.

In the present paper the value of \(\alpha\) is evaluated from the relaxation strength, eq. (2), by assuming that the grain boundary in pure aluminium is particle-free i.e. \(\alpha = 1\); the values of \(\alpha\) are listed in Table 2. The relaxation strength can be determined fairly accurately from the temperature dependence of the measuring frequency. By combining eq. (1),

\[
\tau = \alpha \tau_R(a/a_R) = \alpha \tau_{0R}(a/a_R) \exp \left(\frac{Q}{kT}\right) \quad (1)',
\]

and the relation

\[
\omega \tau = 2\pi f\tau_0 \exp \left(\frac{Q}{kT}\right) = 1, \quad (4)
\]

one obtains

\[
\ln(2\alpha f) + \frac{Q}{kT_R} = \text{const} \quad (5)
\]

### Table 2 Internal friction peaks for SAP(4 mass\%Al\(_2\)O\(_3\)), SUFAP and pure aluminium.

| Ref. | \(T_p\) (K) | \(f\) (Hz) | \(Q\) (eV) | \(-\log(\tau_0)\) (s) | \(Q_{\alpha}^{-1}\) (10\(^{-4}\)) | \(\Delta(1/T)^{\alpha}\) (10\(^{-4}\) K\(^{-1}\)) | \(\beta^{b)}\) (10\(^{-4}\)) | \(\Delta^{c)}\) (10\(^{-4}\)) | \(2a\) (\(\mu\)m) | \(\alpha^{d)}\) |
|------|-------------|-------------|-------------|----------------------|---------------------|----------------------|---------------------|---------------------|---------------------|---------------------|
| SAP  | (2)         | 640         | 35500       | 60                   | 0.38                | 4.8                  | (2100)              | 0.35                | 0.53                |
| SAP  | (3)         | 600         | 50000       | 60                   | 0.57                | 5.4                  | (830)               | 0.9                 | 0.21                |
| SAP(CW)\(^a\) | 453         | 0.83        | 1.3         | 360                  | 0.58                | 4.8                  | (2100)              | 0.35                | 0.53                |
| SAP(EX)\(^b\) | 444         | 0.25        | 1.3         | 125                  | 0.64                | 5.4                  | (830)               | 0.9                 | 0.21                |
| SAP  | (5)         | 538         | 1000        | 1.2                  | 11.3                | 0.45                 | (210)               | 0.5                 | 0.052               |
| SAP\(^c\) | 570         | 1000        | 1.3         | 11.7                | 58                   | 0.19                 | (120)               | 0.5                 | 0.031               |
| SUFAP-1 | 389         | 2           | 1.5 \(\pm\) 0.3 | 20 \(\pm\) 3 | 150                  | 7.5                  | 1320                | 0.1                 | 0.33                |
| SUFAP-2 | 361         | 2           | 1.2 \(\pm\) 0.3 | 18 \(\pm\) 3 | 75                   | 7.3                  | 640                 | 0.1                 | 0.15                |
| SUFAP-3 | 385         | 2           | 1.2 \(\pm\) 0.3 | 18 \(\pm\) 2 | 50                   | 9.5                  | 550                 | 0.1                 | 0.15                |
| SUFAP-4 | 379         | 2           | 1.0 \(\pm\) 0.1 | 15 \(\pm\) 1 | 50                   | 8.8                  | 510                 | 0.1                 | 0.13                |
| pure Al\(^d\) | 553 \(\sim\) 588 | 1           | 1.5         | 14 \(\sim\) 15 | 600                  | 5.5                  | 4000\(^{g)}\)       | 100                 | 1                  |

\(^a\) Half width of the peak in \(Q^{-1}\) vs \(1/T\) curves.

\(^b\) Distribution parameter in the relaxation time when the lognormal distribution is assumed\(^{(29)}\).

\(^c\) Relaxation strengths in parentheses are calculated from \(Q_{\alpha}^{-1}\) and \(\beta\) by using a method presented by Nowick and Berry\(^{(29)}\).

\(^d\) \(\alpha\) is evaluated from a ratio of peak height to that of pure aluminium.

\(^e\) Cold worked

\(^f\) Extruded

\(^g\) Data for peak P3

\(^h\) Typical parameters for the grain boundary peak listed in Ref. (29).

\(^i\) Ref. (12).

\(^{f\text{}}\) The value of \(\alpha\) may be alternatively calculated from \(\tau_0\) (eq. 1) by knowing the grain radius of the specimens. However, the value of \(\tau_0\) and also the value of activation energies \(Q\) are usually determined from measurements in the limited frequency range so that these values are not highly reliable. In fact, one has to assign the value of \(\alpha\) larger than unity for the data of SAP in Refs. (5) and (6). Therefore, we adopt the value of \(\alpha\) determined from the relaxation strength.
where \( f \) is the measuring frequency, \( a \) is the average grain size and \( \tau_0 = \alpha \tau_{R0}(a/a_R) \). In Fig. 5, \( \ln (2af\alpha) \) calculated for various peaks in Table 2 is plotted against \( 1/T \). Almost all data points for pure aluminium and SAP located on a straight line, which suggests that eq. (5) is valid for these materials; the activation energy, \( Q \), evaluated from the inclination of the line is 1.3 eV, which is close to that of the grain boundary peak for pure aluminium (\( \sim 1.5 \) eV). However, the peak temperatures for SUFAP are about 50 K lower than that expected from the line in the figure.

The results of the above analysis shows that the 400 K peak of SUFAP does not follow the simple model for the grain boundary sliding proposed by Mori et al. Although at the present stage of the investigation the reason for the deviation from the theory is not clear, we make a brief discussion on this point in the following.

One of the most notable differences between SUFAP and pure aluminium or SAP is that of the distribution of the relaxation time. The distribution parameter, \( \beta \), for SUFAP is far larger than the others (see Table 2), two possible origins of which are:

1. The structure of the grain boundary for SUFAP is considered to be somewhat different from those for the others. According to the previous study, the density of SUFAP is only 75–85\% of the theoretical one, which suggests that the grains are loosely bound to each other or there are many cavities between them. Further, the present specimens contain a fairly large amount of particles of oxide (\( Al_2O_3 \)) and nitride (\( AlN \)). If these particles exist in the form of plates or films, the model by Mori et al. cannot directly be applied to the 400 K peak. The existence of various sizes and forms of cavities and particles along the grain boundaries may cause a wide distribution of the relaxation time.

2. Mori et al. suggested that the diffusion around second phase particles also causes an internal friction peak. In the present specimens, it is expected that the diffusional relaxation occurs along \( Al_2O_3 \) or \( AlN \). The wide distribution of relaxation times may be a result of such contributions.

In conclusion, it is suggested from the above consideration that the features of the 400 K peak are basically understood as due to the grain boundary sliding with contributions from the diffusional relaxation along \( Al_2O_3 \) and/or \( AlN \) particles.

It is added here that the P3 peak observed by Gondi et al. has a peak temperature higher than those of other peaks of SAP specimens (see Fig. 5) and the distribution parameter, \( \beta \), is much smaller than that for pure aluminium (see Table 2). Therefore, the mechanism of the P3 peak is considered to be different from that for the other peaks in Table 2.

2. **Comparison with mechanical properties**

As described in Introduction, SUFAP exhibits a large elongation around 450 K, which was considered to be due to the grain boundary sliding of fine aluminium grains. If the internal friction peak of SUFAP is originated from the grain boundary sliding, it is expected that some relationship exists between the internal friction and mechanical properties.

In Fig. 6, the maximum fracture strain, \( \varepsilon_{\text{fr}} \), of various specimens are plotted as a function...
of the relaxation strength, $\Delta$. An almost linear relationship exists between $\Delta$ and $\varepsilon_{\text{FS}}^M$; the specimen which has larger internal friction shows larger elongation. It seems interesting to investigate further such correlations of the high temperature mechanical property and internal frictions.

V. Conclusion

The low frequency internal friction of SUFAP has been measured in an inverted torsion pendulum apparatus. A relaxation peak in internal friction was observed at about 400 K. The relaxation strength, the activation energy and the frequency factor were evaluated from the temperature derivative of rigidity modulus vs temperature curves. The peak is considered to be associated with the grain boundary sliding of fine grains of aluminium and the diffusional relaxation along $\text{Al}_2\text{O}_3$ and/or AlN particles. A linear relationship was observed between the relaxation strength of the 400 K peak and the magnitude of the elongation in tensile test measured in the previous study.

Acknowledgement

The authors would like to express their gratitude to Professor T. Mori of Tokyo Institute of Technology for useful suggestion and stimulating discussion.

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