The Effect of Deformation Temperature on Microstructure and Superplastic Flow in Al-Cu-Mg/Si₃N₄p Composites

Ha-Guk Jeong*, Kenji Hiraga*, Mamoru Mabuchi** and Kenji Higashi***

*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

**National Industrial Research Institute of Nagoya, Nagoya 462-0057, Japan

***Department of Metallurgy and Materials Science, College of Engineering,
Osaka Prefecture University, Sakai, Osaka 599-8231, Japan

The effect of deformation temperature on microstructure and superplastic flow in Al-Cu-Mg alloy composite reinforced with Si₃N₄ particles (Si₃N₄p), was investigated. The composites tensile-tested at 758, 773 and 788 K, exhibiting high-strain-rate superplasticity, show high resistance to grain growth during the tensile deformation. However, the composite tensile-tested at 798 K, exhibiting much lower elongation than the composites tensile-tested at lower temperatures, shows considerable grain growth during the tensile deformation. The EDS analysis shows that concentration of the additive elements occurs at the interfaces and the analytical values depend on the sampling points. The largest elongation is obtained at 788 K, which is close to the onset temperature for partial melting determined by DTA. On the other hand, the elongation significantly decreases at 818 K, which is close to the end temperature for partial melting.

(Received December 25, 1998; In Final Form January 19, 1999)

Keywords: deformation temperature, microstructure, high-strain-rate superplasticity, aluminum-alloy matrix composite, reaction phase, high-resolution electron microscopy

I. Introduction

It is well known that superplastic deformation strongly depends on the deformation temperature⁽¹⁾⁽²⁾. Some experimental investigations showed that a maximum elongation obtained at temperatures close to a partial melting temperature in aluminum-alloy matrix composites⁽³⁾⁻⁽⁶⁾. Recently, the nature of matrix/reinforcement interfaces in high-strain-rate superplastic Al-Mg-Si/Si₃N₄w composites was investigated by high-resolution electron microscopy (HREM) and energy dispersive spectroscopy (EDS) analysis⁽⁷⁾. The study revealed that reaction phases exist at interfaces between Si₃N₄ crystals and the matrix, and furthermore, these phases have relatively high magnesium and silicon contents. The result suggested that those reaction phases at the interfaces have a lower melting point than the matrix. The presence of the liquid boundaries was also supported experimentally by *in-situ* transmission electron microscopy and differential scanning calorimetry⁽⁸⁾. It has been suggested that a liquid phase assists both to relax the stress concentration and to limit the built-up of internal cavities and subsequent failure during superplastic flow⁽⁹⁾. However, the presence of a liquid phase does not always lead to high-strain-rate superplasticity, because too much liquid causes decohesion at a liquid phase⁽¹⁰⁾. Increasing the temperature leads to a decreased flow stress and to a displacement of the optimum strain rate range of superplastic deformation towards higher values of strain rate⁽¹¹⁾. However, such an effect of temperature is observed only in the range corresponding to a stable microcrystalline structure⁽¹¹⁾. In our experimental observation⁽¹²⁾, it is observed that a

rapid grain growth makes it impossible to obtain a large elongation in an Al-Cu-Mg/Si₃N₄p composite. Thus, it is important to examine the nature of microstructures of superplastic composites in order to understand the origin of superplastic flow related to fine structure of composites. The present work has paid attention to the change of microstructure in Al-Cu-Mg/Si₃N₄p composites tensile-tested at various temperatures.

II. Experimental

The composites used in this work were prepared by hot-pressing powders of an Al-Cu-Mg alloy (2124 Al) and Si₃N₄ particles at 873 K at a pressure of 390 Mpa for 1.2 ks in vacuum. The sintered samples were then extruded at 733 K with a reduction ratio of 100:1. Details have been described in a previous paper⁽¹³⁾. The extruded samples were tensile-tested at various temperatures.

The samples tensile-tested at 758, 773, 788 and 798 K, respectively, were used in this work. Tensile-tested samples at 758, 773 and 788 K, exhibit very high elongations of about 500, 720 and 840%, respectively, with strain rates of 10⁻¹, 2 × 10⁻¹ and 4 × 10⁻² s⁻¹, respectively. However, the remarkable decrease of elongation (~100%) was observed in the tensile-tested sample at 798 K. The samples were cooled by water soon after the samples were deformed to failure. The time required to cool down the samples from the test temperatures to room temperature was less than 10 seconds. The samples tensile-tested at various temperatures were cut into thin slices, perpendicularly to the extrusion axis. The slices were mechanically polished to a thickness of about 30 μm, and were punched out to 3 mm diameter discs by

an ultrasonic cutter. These disc-shaped samples were thinned with argon ions at an accelerating voltage of 3 kV with a glancing angle of 10–20 degrees. Transmission electron microscopy (TEM) and high-resolution electron microscopy (HREM) observations were performed using an 400 kV electron microscopy (JEM-4000EX) with a resolution of 0.17 nm. Chemical analyses at the interfaces were made with a 300 kV electron microscope (JEM-3000EX) equipped with energy-dispersive X-ray spectroscopy (EDS). The probe size used for the EDS analysis was 3–5 nm. A differential thermal analysis (DTA) was carried out for the as-extruded samples to investigate partial melting. The DTA runs from room temperature to about 1000 K with a constant heating rate of 20 K/min. The thermal analyzer was connected to a computer with a suitable interface, and the data from each run were continuously stored.

III. Results and Discussion

1. Microstructure

Figure 1 shows a typical TEM image of the as-extruded sample. The image shows an average grain size of matrix of $\sim 1.5 \mu\text{m}$ and a uniform distribution of Si₃N₄ particles in the matrix. **Figures 2 (a), (b), (c) and (d)** show TEM images of the as-tensile tested samples at 758, 773, 788 and 798 K, respectively. In the Figs. 2 (a), (b) and (c), one can see that the grain size of the matrix is still small of $\sim 1.5 \mu\text{m}$ in order, indicating no grain growth during the

tensile deformation at 758, 773 and 788 K. This result indicates that there was high resistance to grain growth during the tensile deformation at the above temperature range. The grain size is an important factor to keep superplasticity, because a stable microcrystalline structure is known to be an essential condition for realizing structural superplasticity⁽¹¹⁾. On the other hand, the TEM observation (Fig. 2 (d)) of the sample tensile-tested at 798 K shows that an average grain size of the matrix is $4 \mu\text{m}$, which is larger than those of the samples tensile-tested at lower temperatures up to 788 K. Tensile-testing at 798 K exhibits a much lower elongation than those of tensile-testing at 758, 773 and 788 K. It is important to note that

Fig. 1 TEM image of the as-extruded Al-Cu-Mg/Si₃N₄p sample.

Fig. 2 TEM images of the as-tensile tested Al-Cu-Mg/Si₃N₄p (a) at 758, (b) at 773, (c) at 788 and (d) at 798 K.

the rapid grain growth of the matrix occurs during the tensile deformation process at 798 K, and consequently it leads to the reduction of elongation. From the observation results, it is suggested that the highest superplasticity appears at an optimum temperature, at which the fine grains are kept, and the superplastic property reduces rapidly above the temperature (798 K), at which rapid grain growth occurs.

2. DTA investigation and chemical analysis at interfaces

Figure 3 shows heat flow and elongation at a strain rate of 10^{-1} s^{-1} for the Al-Cu-Mg/Si₃N₄p composites. In the Fig. 3, one can see an endothermic peak, which indicates partial melting at a temperature range of 790–813 K in the composite. The onset temperature for the partial melting is 790 K, which was determined from the intercept of two dotted lines shown in Fig. 3. As can be seen in Fig. 3, a large elongation is obtained at 788 K, which is close to the onset temperature determined by DTA (=790 K) for partial melting in the composite, but the elongation significantly decreased at 798 K, which is close to the bottom of the peak for partial melting determined by DTA (=803 K), as indicated by an arrow. It is clear that the optimum temperature for superplasticity is close to the onset temperature for partial melting. The same trend was found for other high-strain-rate superplastic Al-Mg-Si/Si₃N₄ composites⁽¹⁰⁾.

In the metal-ceramics composites, stress concentration can be considered to occur at interfaces of metal/ceramics during deformation, so cavitation is excessively developed at the interfaces, resulting in premature fracture. In a previous paper⁽⁹⁾, the accommodation process by a liquid phase has been proposed from the viewpoint of the mechanisms with a direct observation of a melting phase along grain boundaries and interfaces using an *in-*

Fig. 3 Heat flow and elongation at a strain rate of 10^{-1} s^{-1} for the Al-Cu-Mg/Si₃N₄p composites⁽¹²⁾.

situ TEM technique. However, the presence of a liquid phase does not always lead to high-strain-rate superplasticity, because too much liquid causes decohesion at a liquid phase⁽¹⁰⁾. This indicates that there is basically a unique role of liquid phase for developing high-strain-rate superplasticity in these materials. From the present investigations, it is inferred that high-strain-rate superplasticity does not appear in the temperature range higher than the onset temperature of partial melting, but appears only at an optimum temperature, which is close to the onset temperature of partial melting in the composite. This indicates that a part of reaction phases is concerned in the partial melting at the optimum superplastic condition. In the authors' previous papers⁽⁷⁾⁽¹⁴⁾, the presence of reaction phases at interfaces between the matrix and Si₃N₄w was reported, and the reaction phases with a relatively large amount of Mg and Si in the as-tensile tested Al-Mg-Si/Si₃N₄w and Al-Cu-Mg/Si₃N₄w composites. In these studies, the reaction phases were found to be a primary solid solution of Al containing

Fig. 4 (a) TEM image of a Si₃N₄ particle embedded in the matrix in the as-tensile tested Al-Cu-Mg/Si₃N₄p sample at 788 K. (b) HREM image of the place indicated by a rectangle in Fig. 4 (a) and sampling points in Table 1.

Table 1 EDS data (at%) at the sampling points in Fig. 4 in the as-tensile tested Al-Cu-Mg/Si₃N₄p sample.

Sample point	Domains	Al	Mg	Cu	Mn	Si	Fe	Zn
1	Al-matrix	97.7	—	0.7	—	1.2	0.3	—
2	Si ₃ N ₄ p	19.2*	—	—	—	80.1	—	—
3	Reactant	88.2	9.7	0.6	—	1.5	—	—
4	Reactant	81	9.2	3.6	—	6.1	—	—
5	Reactant	90.2	3.4	1.1	—	4.7	—	—

*The Al content in Si₃N₄p results from overlapping of the Al-matrix.

supersaturated Mg and Si. The primary solid solution containing Mg and Si has a lower melting point than the matrix, and is considered to transform to a liquid phase at the tensile-testing temperature.

The TEM observation of the as-tensile tested Al-Cu-Mg/Si₃N₄p shows interface structures similar to those of the as-tensile tested Al-Mg-Si/Si₃N₄w and Al-Cu-Mg/Si₃N₄w. **Figure 4** shows a HREM image of Al-Cu-Mg/Si₃N₄p, where EDS analysis was carried out. In the image, the places at which analysis was performed, are numbered. The analytical data for these sampling points are listed in **Table 1**. The result shows that concentration of the additive elements occurs at the interfaces and the analytical values depend on the sampling points. It implies that the observed reaction phases does not have a single chemical composition. Therefore, it is inferred that the reaction phases start to melt gradually with an interval at elevated temperature and a part of reaction phases is concerned in the partial melting at the optimum superplastic condition.

IV. Conclusions

The effect of deformation temperature on microstructure and superplastic flow in Al-Cu-Mg/Si₃N₄p composites has been studied. The obtained results are summarized as follows:

(1) The composites tensile-tested at 758, 773 and 788 K, which exhibit high-strain-rate superplasticity, show high resistance to grain growth during tensile deformation. However, the composite tensile-tested at 798 K, exhibiting much lower elongation, shows considerable grain growth during tensile deformation. In this case, the grain growth occurring during the deformation process can be one of the reasons for the loss of superplasticity of the material. This indicates that a stable microcrystalline structure can be an essential condition for realizing structural superplasticity.

(2) For the Al-Cu-Mg/Si₃N₄p composite, it is clearly shown that a large elongation was obtained at around the onset temperature for partial melting in the composite. On the other hand, the elongation significantly decreased at beyond the onset temperature of partial melting in the composite. Therefore, it is suggested that a small quantity of liquid is may be concerned in high-strain-rate superplastic flow of the Al-Cu-Mg/Si₃N₄p composite.

REFERENCES

- (1) O. A. Kaibyshev: *Superplasticity of Alloys, Intermetallides and Ceramics*, Springer-Verlag Berlin Heidelberg, (1992), p. 3.
- (2) K. Matsuki and M. Yamada: *J. Japan Inst. Met.*, **37** (1973), 448.
- (3) M. Mabuchi and K. Higashi: *Mater. Sci. Eng.*, **A179/A180** (1994), 625.
- (4) M. Mabuchi and K. Higashi: *Mater. Trans., JIM*, **35** (1994), 399.
- (5) K. Higashi, T. G. Nieh, M. Mabuchi and J. Wadsworth: *Scripta Metall. Mater.*, **32** (1995), 1079.
- (6) J. Koike, M. Mabuchi and K. Higashi: *J. Mater. Res.*, **10** (1995), 133.
- (7) H.-G. Jeong, K. Hiraga, M. Mabuchi and K. Higashi: *Philos. Mag. Lett.*, **74** (1996), 73.
- (8) J. Koike, M. Mabuchi and K. Higashi: *Acta Metall. Mater.*, **43** (1995), 199.
- (9) M. Mabuchi and K. Higashi: *Philos. Mag. Lett.*, **70** (1994), 1.
- (10) M. Mabuchi, H. Iwasaki, H.-G. Jeong, K. Hiraga and K. Higashi: *Mater. Res.*, **12**, 9 (1997), 2332.
- (11) O. A. Kaibyshev: *Superplasticity of Alloys, Intermetallides and Ceramics*, Springer-Verlag Berlin Heidelberg, (1992), p. 8.
- (12) H.-G. Jeong: Doctor dissertation, Tohoku University, (1998).
- (13) M. Mabuchi, K. Higashi and T. G. Langdon: *Acta Metall. Mater.*, **5** (1994), 1739.
- (14) H.-G. Jeong, K. Hiraga, M. Mabuchi and K. Higashi: *Acta Metall. Mater.*, **46** (1998), 6009.