

# Effects of Mg and Cu Additions on Superplastic Behavior in MA Aluminum Alloys

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**Abstract** MA Al alloys are examined to determine the effects of alloying of Mg and Cu and rolling on tensile deformation behavior at 748 K over a wide strain rate range ( $10^{-4}$ - $10^3$ /s). A powder metallurgy aluminum alloy produced from mechanically alloyed pure Al powder exhibits only a small elongation-to-failure ( $\epsilon_f < \sim 50\%$ ) in high temperature (748 K) tensile deformation at high strain rates ( $\dot{\epsilon} = 1$ - $10^2$ /s).  $\epsilon_f$  in MA Al-0.5~4.0Mg alloys increases slightly with Mg content ( $\epsilon_f = \sim 140\%$  at 4 mass%). Combined addition of Mg and Cu (MA Al-1.5%Mg-4.0%Cu) is very effective for the occurrence of superplasticity ( $\epsilon_f > 500\%$ ). Warm-rolling (at 393-492 K) tends to raise  $\epsilon_f$ . Lowering the rolling-temperature is effective for increasing the ductility. The effect is rather weak in MA pure Al and MA Al-Mg alloys, but much larger in the MA Al-1.5%Mg-4.0%Cu alloy. Additions of Mg and Cu and warm-rolling of the alloy cause a remarkable reduction in the logarithm of the peak flow stress at low strain rates ( $\dot{\epsilon} < \sim 1$ /s) and sharpening of microstructure and smoothing of grain boundaries. Additions of Mg and Cu make the strain rate sensitivity (the  $m$  value) larger at high strain rates, and the warm-rolling may make the grain boundary sliding easier with less cavitation. Grain boundary facets are observed on the fracture surface when  $\epsilon_f$  is large, indicating the operation of grain boundary sliding to a large extent during superplastic deformation.

**Key words** tensile deformation, strain rate, superplasticity, cavitation, grain boundary sliding.

## 1. Introduction

Powder-metallurgy aluminum (Al) alloys produced from mechanically alloyed (MA) Al powders consist of fine (submicron in diameter) grains (or subgrains).<sup>1,2)</sup> Further, very fine (a few 10 nm in diameter) particles are generally distributed by a several volume% homogeneously in the matrix. They may be  $Al_2O_3$  and  $Al_4C_3$  formed by the reaction of Al with ethanol which was incorporated into the Al powder during MA process.<sup>3-5)</sup> Therefore, superplastic deformation due to fine grained structure is expected to occur at elevated temperatures under high strain rates. In fact, a large elongation-to-failure has been reported in tension of some MA Al alloys at extremely high strain rates above  $10^{-1}$ /s.<sup>6-8)</sup>

Previous works have shown that, without exception, the maximum value of tensile elongation is smaller in a MA Al-Mg alloy (IN9052) than in MA Al-Mg-Cu alloys (IN9021 and IN90211) despite their similar microstructure

(uniform dispersion of very fine carbides and oxides in a fine grained matrix).<sup>6-9)</sup> Further, it was found in the preliminary experiment that an alloy produced from MA pure Al powder, that is also similar in microstructure to the above alloys, exhibited only a small tensile elongation (a few 10% in maximum). These facts propose that some sort of alloying elements, which may be dissolved in the matrix at the deformation temperature, also affects the tensile elongation, probably through some modification of microstructure besides particle dispersion and grain size. Moreover, it has been found that thermomechanical processing like warm-rolling to deformation influences the amount of tensile elongation in an ingot-metallurgy Al-Mg alloys.<sup>10)</sup>

The purpose of the present paper is to examine the effects of alloy additions of Mg and Cu and warm-rolling on the high-temperature superplastic behavior in MA Al alloys, and to know the microstructure responsible for high strain rate superplasticity.

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## 2. Experimental Procedure

Chemical compositions of MA Al alloys examined in the present work are, by mass%, Al-1.7%O-1.1%C, Al-0.5%Mg-1.7%O-1.1%C, MA Al-2.0%Mg-1.7%O-1.1%C, Al-4.0%Mg-0.8%O-1.1%C and Al-1.5%Mg-4.0%Cu-0.8%O-1.1%C. The last two are IN9052 and IN9021, respectively, produced by Incomap Co. The alloys will be designated MA pure Al, MA Al-0.5Mg, MA Al-2.0Mg, MA Al-4.0Mg and MA Al-1.5Mg-4.0Cu in this paper. The difference in volume% of fine particles due to the difference in oxygen content was  $\sim 2\%$ . Plates (4 mm thick) were machined from extruded rods of these alloys and rolled to 1 mm thickness (reduction in thickness:  $\sim 75\%$ ), parallel to the extruding direction, at three different temperatures: 393, 423 and 493 K. Specimens for tension tests (length 10 mm, width 5 mm, thickness 1 mm) were machined from the rolled sheets, parallel to the rolling direction. Specimens were finally annealed at 823 K for 1 h and then quenched in iced-water. After heating quickly to 753 K (deformation temperature) and holding at the temperature for 10 min, specimens were deformed at constant nominal strain rates ( $1 \times 10^{-4} - 1 \times 10^3/s$ ) with hydraulic testing machines. The matrices of the alloys are supposed to be in a state of solid solution at the deformation temperature;  $Al_3Mg_2$  and  $CuMgAl_2$  phases in MA Al-Mg alloys and MA Al-1.5Mg-4.0Cu, respectively, may have been mostly dissolved.

Thin foils for TEM observation were prepared from specimens by spark-erosion machining and standard twin-jet electro polishing, and examined in a JEM-3010 electron microscope operated at 300 kV. Fracture surfaces were inspected by SEM (JSM-890S).

## 3. Results and Discussion

### 3.1 Stress vs. Strain Behavior

Examples of nominal stress,  $\sigma_n$ , vs. nominal strain,  $\epsilon_n$ , curves obtained at 753 K under extremely high and low strain rates ( $5$  and  $1 \times 10^{-4}/s$ ) are given in Fig. 1 for MA pure Al, MA Al-4.0Mg and MA Al-1.5Mg-4.0Cu.  $\sigma_n$  vs.  $\epsilon_n$  curves for MA Al-0.5Mg and MA Al-2.0Mg were located between those for MA pure Al and MA Al-4.0Mg. The following can be seen from the figure: (1) Both maximum flow stress and elongation-to-failure are larger at higher strain rate; (2) In contrast to the deformation at room temperature, the maximum flow stress decreases with the alloy additions of Mg and Cu; (3) Elongation-to-failure increases with the alloying. It may be noteworthy that the tensile elongation of MA pure Al is very small ( $<1\%$ ) at low strain rate.

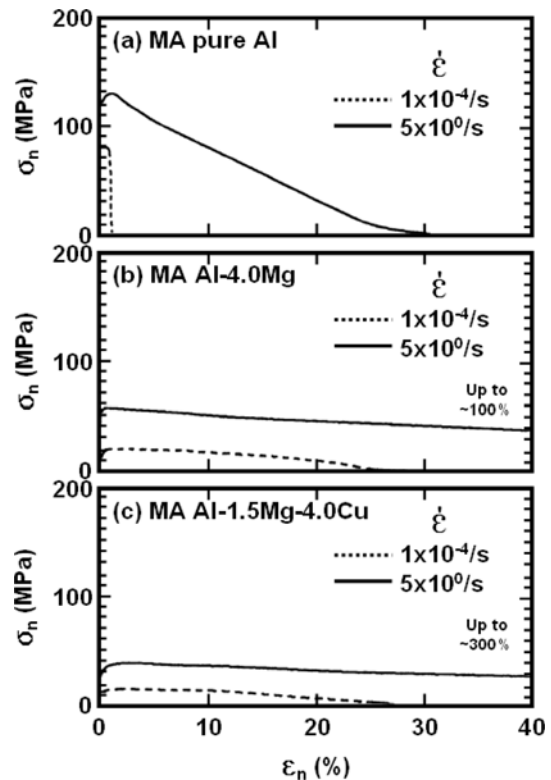


Fig. 1. Nominal stress,  $\sigma_n$  vs. nominal strain,  $\epsilon_n$ , curves at 748 K for (a) MA pure Al, (b) MA Al-4.0Mg and (c) MA Al-1.5Mg-4.0Cu rolled at 423 K and finally annealed at 773 K.

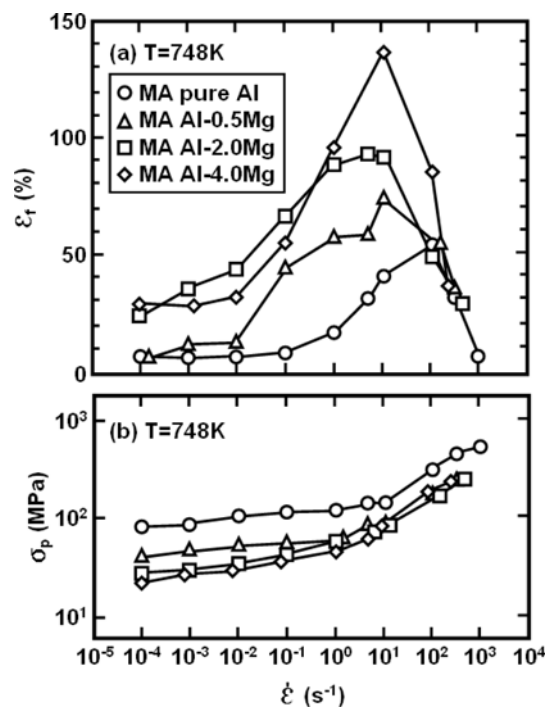
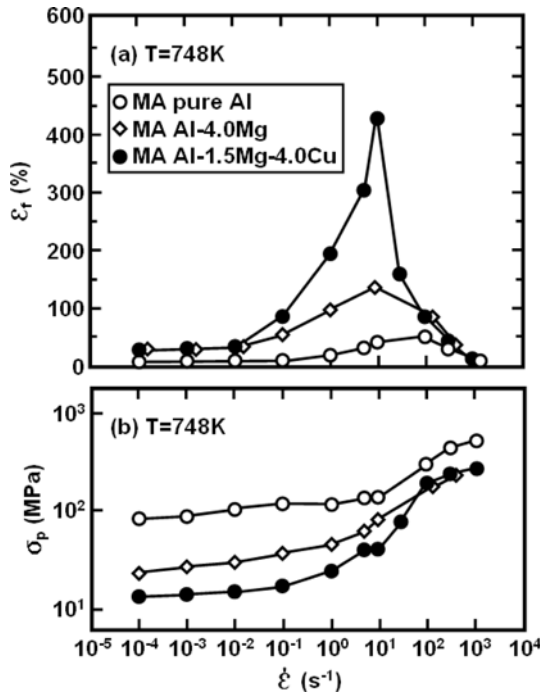


Fig. 2. Effect of Mg addition on (a) elongation-to-failure,  $\epsilon_f$  vs. nominal strain rate,  $\dot{\epsilon}$ , and (b) peak flow stress,  $\sigma_p$  vs.  $\dot{\epsilon}$  relations in MA Al-Mg alloys rolled at 423 K and finally annealed at 773 K.



**Fig. 3.** Effect of combined addition of Mg and Cu on (a) elongation-to-failure,  $\epsilon_f$  vs. nominal strain rate,  $\dot{\epsilon}$ , and (b) peak flow stress,  $\sigma_p$  vs.  $\dot{\epsilon}$  relations in Al-1.5Mg-4.0Cu rolled at 423 K and finally annealed at 773 K. Results on MA pure Al and MA Al-4.0Mg are also shown.

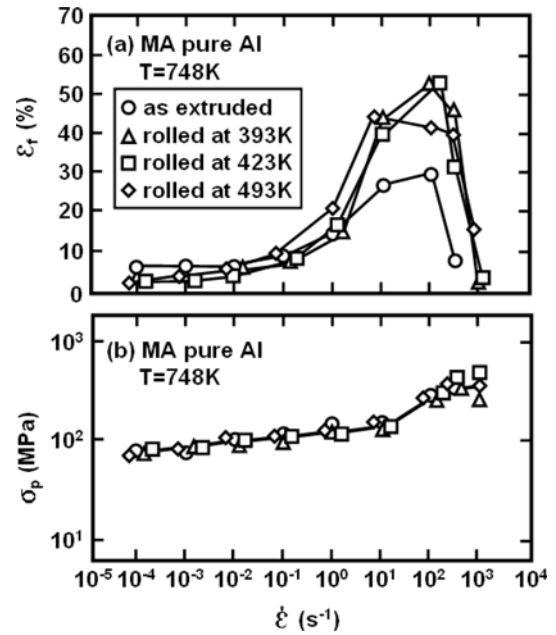
### 3.2 Tensile Elongation and Strain Rate Sensitivity

#### 3.2.1 Effect of addition of alloying elements, Mg and Cu

Figure 2 shows changes in the elongation-to-failure,  $\epsilon_f$ , and the peak flow stress,  $\sigma_p$ , as a function of the nominal strain rate,  $\dot{\epsilon}$ , in some MA Al-Mg alloys. The value of  $\epsilon_f$  reaches a maximum in the neighborhood of  $\dot{\epsilon} = 10$ /s. The strain rate sensitivity,  $m = d(\ln \sigma_p)/d(\ln \dot{\epsilon})$ , a parameter governing the rate of neck development, correspondingly takes a large value (0.3-0.4) in high strain rates ( $\dot{\epsilon} > \sim 1$ /s), while it is less than 0.1 at lower ones ( $\dot{\epsilon} < \sim 1$ /s). The maximum values both in  $\epsilon_f$  and  $m$  increase with the increase in the amount of alloying element, Mg.

Figure 3 shows  $\epsilon_f$  vs.  $\dot{\epsilon}$  and  $\sigma_p$  vs.  $\dot{\epsilon}$  relations in MA Al-1.5Mg-4.0Cu along with the results on MA pure Al and MA Al-4.0Mg for comparison. The tendency observed in the addition of Mg becomes more pronounced by the combined addition of Mg and Cu. Variation in the deformation behavior with the alloying can be characterized by the features: an increase in the maximum of  $\epsilon_f$  at high strain rates (around  $\dot{\epsilon} = \sim 10$ /s), a remarkable decrease in  $\ln \sigma_p$  at low strain rates ( $\dot{\epsilon} < \sim 1$ /s), and a slight decrease in the strain rate at which  $\epsilon_f$  takes the maximum.

At least from a phenomenological viewpoint, one can say that the decrease in  $\ln \sigma_p$  at low strain rates leads to the increase in the  $m$  value and consequently to the



**Fig. 4.** Effect of Warm-rolling on (a) elongation-to-failure,  $\epsilon_f$  vs. nominal strain rate,  $\dot{\epsilon}$ , and (b) peak flow stress,  $\sigma_p$  vs.  $\dot{\epsilon}$  relations in MA pure Al finally annealed at 773 K.

increase in  $\epsilon_f$  at high strain rates. The third feature may be related to the increase in the grain size former to deformation, as will be shown later.

#### 3.2.2 Effect of rolling

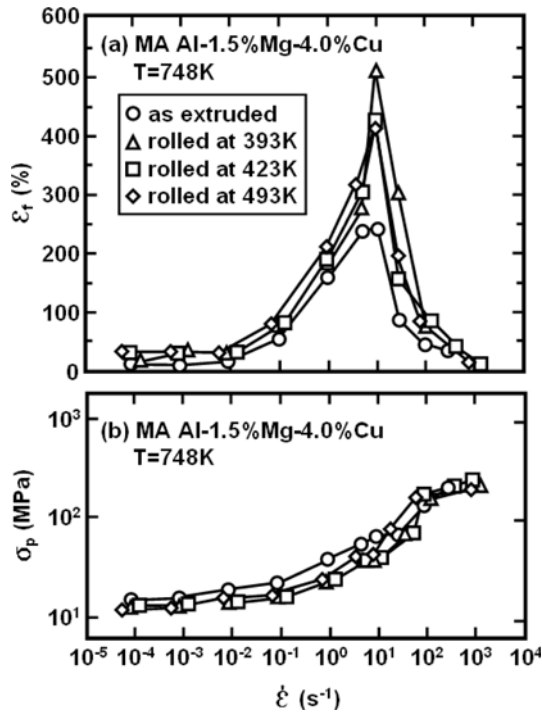
Figure 4 shows changes in  $\epsilon_f$  vs.  $\dot{\epsilon}$  and  $\sigma_p$  vs.  $\dot{\epsilon}$  relations with warm-rolling former to deformation in MA pure Al that exhibited the most poor ductility among the alloys examined. The maximum value of  $\epsilon_f$  observed around  $\dot{\epsilon} = \sim 50$ /s becomes larger by the rolling, particularly at low temperature (Fig. 4(a)). However, the amount of the increase in  $\epsilon_f$  is considerably small ( $\sim 25\%$ ), reflecting no notable change in the  $\sigma_p$  vs.  $\dot{\epsilon}$  relation (Fig. 4(b)). In MA Al-4.0Mg, too, the effect of warm rolling was found to be rather small.

In MA Al-1.5Mg-4.0Cu that exhibited the largest elongation, on the other hand, the tensile ductility can be largely improved by the rolling; the increase in the peak value of  $\epsilon_f$  reaches  $\sim 300\%$  after rolling at the lowest temperature (393 K) (Fig. 5(a)). This may be resulted from the increase in the  $m$  value caused by the reduction in  $\ln \sigma_p$  at low strain rates below 1/s (Fig. 5(b)).

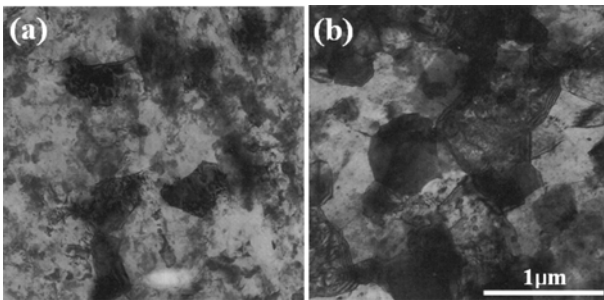
### 3.3 Microstructure

Let us consider here, from a viewpoint of microstructure, the reason for the increase in tensile ductility by the alloy addition and the prior warm-rolling.

Examples of TEM micrographs of MA pure Al and MA Al-1.5Mg-4.0Cu after rolling at 423 K and subsequent

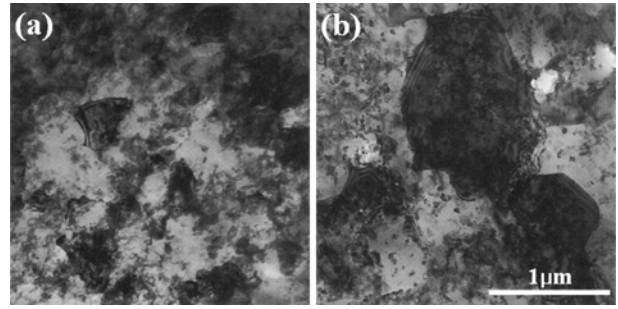


**Fig. 5.** Effect of Warm-rolling on (a) elongation-to-failure,  $\epsilon_f$  vs. nominal strain rate,  $\dot{\epsilon}$ , and (b) peak flow stress,  $\sigma_p$  vs.  $\dot{\epsilon}$  relations in MA Al-1.5Mg-4.0Cu finally annealed at 773 K.



**Fig. 6.** TEM micrographs of (a) MA pure Al and (b) MA Al-1.5Mg-4.0Cu rolled at 423 K and finally annealed at 773 K.

annealing at 823 K are given in Fig. 6. Even MA pure Al, which exhibited very poor ductility, consists of fine grains(or subgrains) indeed. However, the internal structure of this alloy has not yet been recovered before deformation (after the above thermomechanical treatment); the crystal lattice is highly strained and the grain boundaries are irregular as indicated by complex bend contours in the few thickness fringes and grain interiors at grain boundaries, respectively. On the other hand, MA Al-1.5Mg-4.0Cu, which exhibited large elongation, has been almost fully recovered; the grain size is larger compared to that of MA pure Al, the grain interiors are rather free from dislocations and clear thickness fringes can be seen at grain boundaries. The internal structure of MA Al-4.0Mg was found to be in an incompletely recovered state.

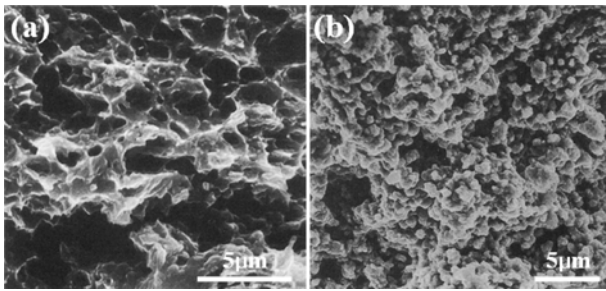


**Fig. 7.** TEM micrographs of (a) MA pure Al and (b) MA Al-1.5Mg-4.0Cu after high-temperature extrusion.

These observations may suggest that the characteristic changes in deformation behavior with the alloying of Mg and Cu, namely the decrease in  $\ln \sigma_p$  at low strain rates below 1/s and the increase in  $\epsilon_f$  at higher strain rates around 10/s, are ascribed to the smoothening of grain boundaries; more smooth boundaries would be able to slide under lower stresses with less cavitation. Further, the strain rate giving the peak of  $\epsilon_f$  showed a tendency to decrease with the alloying(Fig. 3). This might be related to the increase in grain size by the alloying(compare Figs. 6(a) and 6(b)). At present, we do not know the exact reason for the development of more well-defined grain(or subgrain) structure with the alloying. It might be due to the increase in the self diffusion of Al by the solute atoms, Mg and/or Cu, as suggested by Bieler *et al.*<sup>9)</sup>

Figure 7 shows TEM micrographs taken from the as-extruded MA pure Al and MA Al-1.5Mg-4.0Cu. One can see the effect of warm rolling on the evolution of microstructure by comparing micrographs in Fig. 6 with those in this figure. In MA pure Al, the effect is small; the internal structure is highly strained in both the extruded and the subsequently rolled states. In MA Al-1.5Mg-4.0Cu, on the other hand, the effect is considerably large; the partially recovered structure consisting of rather coarse grains in the as-extruded state can be made more sharp(more well-defined) in nature and finer in scale by the rolling. Taking into account that the sharpening of structure involves an increase in the boundary misorientation due to the incorporation of excess dislocations, which have been induced during rolling, into pre-existing boundaries, the enhancement of superplastic deformation by the rolling observed in MA Al-1.5Mg-4.0Cu is reasonably considered to be a reflection of the sharpening and refinement of microstructure.<sup>10)</sup>

SEM micrographs of fracture surface in MA pure Al and MA Al-1.5Mg-4.0Cu deformed at  $T = 753$  K and  $\dot{\epsilon} = 10$ /s are given in Fig. 8. In the former( $\epsilon_f = \sim 40\%$ ), the surface is wavy and many dimples are formed, indicating that the fracture is essentially transgranular. In the latter( $\epsilon_f = \sim 400\%$ ), grain boundary facets can be seen



**Fig. 8.** SEM micrographs of fracture surface in (a) MA pure Al and (b) MA Al-1.5Mg-4.0Cu rolled at 423 K and finally annealed at 773 K.  $T = 748$  K,  $\dot{\epsilon} = 10/s$ .

and the fracture is intergranular. These facts may suggest that the large elongation due to high strain rate superplastic deformation in MA Al alloys is mostly caused by the grain boundary sliding, as reported previously.<sup>7)</sup>

#### 4. Conclusions

(1) Powder metallurgy aluminum(Al) alloy produced from mechanically alloyed pure Al powder(MA pure Al) exhibits only a small elongation-to-failure( $\epsilon_f < \sim 50\%$ ) in high temperature(748 K) tensile deformation at high strain rates( $\dot{\epsilon} = 1-10^2/s$ ).  $\epsilon_f$  in MA Al-0.5~4.0Mg alloys increases slightly with Mg content( $\epsilon_f = \sim 140\%$  at 4 mass%). Combined addition of Mg and Cu(MA Al-1.5%Mg-4.0%Cu, IN9021) is very effective for the occurrence of superplasticity( $\epsilon_f > 500\%$ ).

(2) Warm-rolling(at 393-492 K) tends to raise  $\epsilon_f$ . Lowering rolling-temperature is effective for the ductility increase. The effect is rather weak in MA pure Al and MA Al-Mg alloys, but much larger in MA Al-1.5%Mg-4.0%Cu alloy.

(3) Alloy additions of Mg and Cu and warm-rolling

cause a remarkable reduction in the logarithm of peak flow stress at low strain rates( $\dot{\epsilon} < \sim 1/s$ ), and sharpening of microstructure and smoothening of grain boundaries. The former makes the strain rate sensitivity(the m value) larger at high strain rates, and the latter may make the grain boundary sliding easier with less cavitation.

(4) Grain boundary facets can be observed on the fracture surface when  $\epsilon_f$  is large, indicating the operation of grain boundary sliding to a large extent during superplastic deformation.

#### References

1. I. E. Anderson and J. C. Foley, *Surf. Interface Anal.*, **31**, 599 (2001).
2. D. Jiang and T. Imai, *Mater. Chem. Phys.*, **80**, 15 (2003).
3. M. Besterci, O. Velgosová and L. Kováč, *Mater. Lett.*, **54**, 124 (2002).
4. T. Ishikawa, N. Yukawa, Y. Yoshida and K. Murakami, *J. Mater. Proc. Tech.*, **113**, 632 (2001).
5. T. Fujii, S. Sodeoka and K. Ameyama, *J. Jpn. Inst. Light Metals*, **47**, 329 (1997).
6. T. G. Nieh, P. S. Gilman and J. Wadsworth, *Scripta Metall.*, **19**, 1375 (1985).
7. T. R. Bieler, T. G. Nieh, J. Wadsworth and A. K. Mukherjee, *Scripta Metall.*, **22**, 81 (1988).
8. K. Higashi, T. Okada, T. Mukai and S. Tanimura, *Proc. Conf. on Superplasticity in Advanced Materials*, ed. by S. Hori, M. Tokizane and N. Furushiro, p.569, JSRS, Osaka (1991).
9. T. R. Bieler, S. F. Meagher, J. A. Diegel and A. K. Mukherjee, *Proc. Conf. on Hot Deformation of Aluminum Alloys*, ed. by T. G. Langdon, H. D. Merchant, J. G. Morris and M.A. Zaidi, p.297, TMS, Warrendale (1991).
10. M. A. Garcia-Bernal, R. S. Mishra, R. Verma and D. Hernandez-Silva, *Mater. Sci. Eng., A*, **534**, 186 (2012).