



ORIGIN OF SUPERPLASTIC ELONGATION IN ALUMINUM ALLOYS PRODUCED BY MECHANICAL MILLING

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Abstract—The effects of alloying (Mg, Cu, Ge and/or Si) and prior rolling and annealing on tensile stress–strain behaviors were examined for mechanically milled, powder metallurgy aluminum alloys at 748 K in the strain rate range of 1×10^{-4} – 7.5×10^2 /s. Lowering the temperature of prior rolling and increasing the anneal temperature and time result in an increase in the strain rate sensitivity, m . Especially alloying effectively increases the m value as well as the strain to fracture, ϵ_f , at the intermediate strain rate of approximately 10^0 /s; m and ϵ_f reach 0.4 and 440% for a case of alloying both 1.1 at.% Mg and 1.2 at.% Cu, though they are smaller than 0.1 and 100% at lower ($< 1 \times 10^{-2}$ /s) and higher ($> 5 \times 10^1$ /s) strain rates. TEM and SEM revealed that the microstructure of superplastic alloys, compared to pure Al, consists of thermally recovered, fine equiaxed grains (smaller than $1 \mu\text{m}$ in diameter) with smooth grain boundaries of large misorientation. It is then postulated that superplastic elongation occurs when smooth boundaries slide under a plastically stable condition due to a large m value. © 1998 Acta Metallurgica Inc. Published by Elsevier Science Ltd. All rights reserved.

1. INTRODUCTION

Mechanical alloying (MA) was originally developed to make oxide dispersion strengthened nickel-based superalloys [1], and later applied to aluminum-based alloys [2]. During MA of a reactive metal like aluminum, a process control agent (generally, organic matters such as stearic acid and ethanol) is added to maintain a balance between cold welding and fracture of aluminum powder particles [2]. This allows the development of extremely uniform dispersion of fine Al_2O_3 and Al_4C_3 particles [3] (a few 10 nm in size) which were formed due to reaction of Al with oxygen and carbon in the agent.

The dispersion of these thermally stable particles retards the coarsening of microstructure at high temperatures because of their pinning effects for dislocations and grain boundaries. Powder metallurgy aluminum alloys produced by mechanical milling§ of aluminum alloy powders (MM Al alloys) consist of fine grains (smaller than $1 \mu\text{m}$ in diameter) which are stable up to high temperatures [4]. Besides their excellent strength and creep resistance, they are expected to show the superplasticity due to fine grained structure at high temperatures and high

strain rates. In fact, a large elongation to fracture of several 100% has been reported in tension of some MM Al alloys at strain rates above 10^{-1} /s (e.g. Refs [5–11]). The high strain rate superplasticity has also been analyzed from a mechanistic viewpoint to discuss the mechanism [9].

A survey of previous work indicates that, without exception, the superplastic MM aluminum-based materials are made from aluminum alloys such as Al–Mg (IN9052) [10], Al–Mg–Cu (IN9021 and IN90211) [5, 6, 10] and Al–Mg–Li (IN905XL) [11]. On the other hand, a material made from mechanically milled powder of pure aluminum exhibited only a moderate elongation of less than 100%, despite its fine grain size (submicron) [12]. These facts suggest that some sort of alloying modifies the microstructure, without a notable change in grain size, so as to be suitable for superplastic deformation. Further, thermomechanical processing like rolling and annealing before deformation may possibly influence the elongation through a change in microstructure, as shown in ingot metallurgy Al–Mg alloys [13] and a Al–SiC_w composite [14].

At present, however, microstructural aspects of high strain rate superplasticity in MM Al alloys have not been fully understood. The purpose of the present work is, after clarifying the effects of alloy addition and prior rolling and annealing on flow stress and elongation to fracture, to discuss the origin of superplastic deformation from a microstructure viewpoint.

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§The terminology “mechanical milling (MM)” was used instead of mechanical alloying (MA), since starting materials themselves are atomized powders of aluminum alloys.

2. EXPERIMENTAL PROCEDURE

Atomized powders of aluminum alloys with the addition of Mg, Cu, Ge and/or Si, solely or in combination, were mechanically milled in an argon atmosphere by using a high energy ball mill. Magnesium and copper were chosen since the alloys containing both of these elements (IN9021, IN90211) have been known to exhibit superplasticity [5,6,10]. Germanium and silicon were chosen because they are, like Mg and Cu, larger and smaller than Al in atomic size, respectively. The amount of alloying element was limited to be within the solubility limit at the deformation temperature of 748 K. Ethanol was added as a process control agent during MM to form and disperse fine Al_2O_3 and Al_4C_3 particles (~ 35 nm in size and $\sim 7.5\%$ in volume fraction) in the matrix through subsequent high temperature treatments [15].

The MM powders were first cold pressed in aluminum cans, and then degassed in vacuum, hot pressed and finally extruded at 773 K for consolidation. The uniform dispersion of such small particles of Al_2O_3 and Al_4C_3 was effective in suppressing the grain coarsening up to approximately 800 K. Alloy notation is, for example, "MM Al-1.1Mg-1.2Cu" for a material made from atomized powder of Al-1.1 at.% Mg-1.2 at.% Cu alloy. The "MM Al-Mg/Cu system" is also used when alloyed with Mg or Cu solely, or Mg and Cu in combination. For comparison, a material was prepared from atomized pure aluminum powder (purity in wt%: 99.7; main impurities are Fe 0.13, Si 0.06, Cu 0.001 and Mn 0.001) by the same process mentioned above. This is denoted "MM pure Al".

Plates machined from extruded rods were then rolled by 70% in thickness reduction at 423 K, parallel to the extruding direction. Specimens for the tension test (gauge size: 8 mm long, 4 mm wide and 1 mm thick) were machined from the rolled sheets to have the gauge length direction parallel to the rolling direction. They were annealed at 773 K for 3.6 ks and then water quenched. After heating up to 748 K within 20 min and holding for 10 min, they were deformed in tension at a nominal strain rate in the range of 1×10^{-4} – $7.5 \times 10^2/\text{s}$ on a hydraulic testing machine. Thin foils for TEM were prepared from specimens before deformation by mechanical and electrochemical polishing, and were examined by using an electron microscope with a goniometer stage operated at 300 kV. Specimen surfaces after deformation were observed by using SEM.

3. RESULTS AND DISCUSSION

3.1. Stress-strain behavior

Examples of true stress, σ_t , vs true strain, ϵ_t , curves at low ($1 \times 10^{-3}/\text{s}$), intermediate ($1 \times 10^0/\text{s}$) and high ($1 \times 10^2/\text{s}$) strain rates are given in

Fig. 1(a)–(c), respectively, for MM Al-Mg/Cu and MM Al-Ge/Si system alloys. The maximum flow stress increases with increasing strain rate. In contrast to room temperature deformation, it decreases by alloying in both alloy systems. Maximum elongation to fracture is obtained at the intermediate range of strain rate and increases when alloyed. The combined addition of 1.1 at.% Mg and 1.2 at.% Cu is notably effective for an increase in elongation ($> 400\%$ in nominal strain).

When tensile elongation is large at the intermediate strain rate [Fig. 1(b)] the flow stress remains almost constant during straining over a wide range of strain, as previously reported for IN90211 alloy containing Mg and Cu [16]. Continuous observation of deformation by using a high speed video camera (4500 frames/s) revealed that, under the superplastic

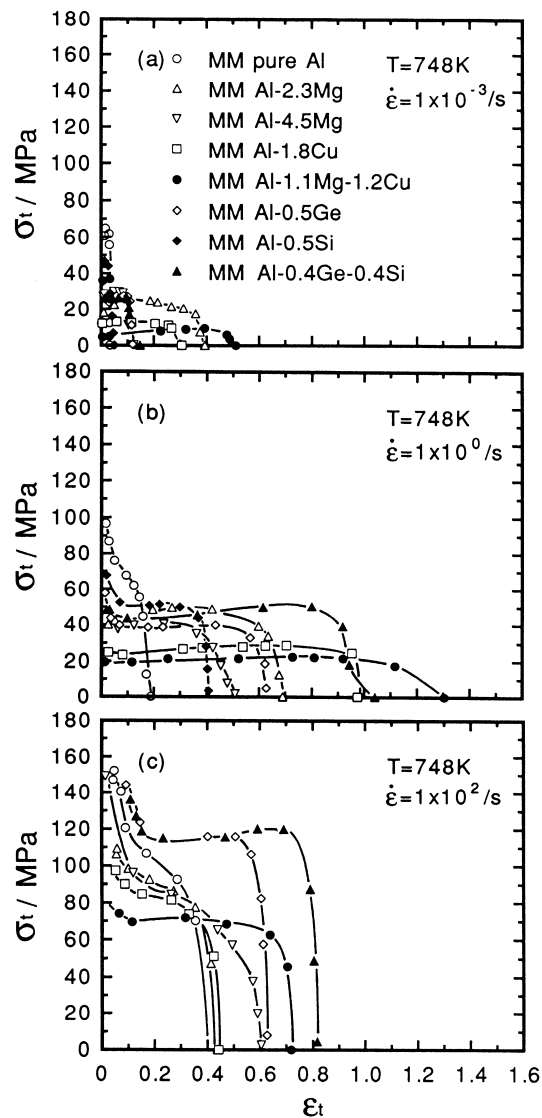


Fig. 1. True stress σ_t vs true strain ϵ_t curves for MM Al-Mg/Cu and MM Al-Ge/Si system alloys, $T = 748$ K: (a) $\dot{\epsilon} = 1 \times 10^{-3}/\text{s}$; (b) $\dot{\epsilon} = 1 \times 10^0/\text{s}$; (c) $\dot{\epsilon} = 1 \times 10^2/\text{s}$.

condition, deformation proceeds fairly uniformly along the gauge length and fracture occurs without clear local necking [17].

On the other hand, when the elongation is small, as in deformation of MM pure Al at all the strain rates and of MM Al alloys both at low and high strain rates (under a non-superplastic condition), local necking was produced at the early stage of deformation and the specimen fractured immediately at the neck part because of severe strain concentration. It is postulated that, in the present materials, the superplastic elongation occurs when the neck formation is delayed.

3.2. Strain rate dependence of tensile elongation and flow stress

Nominal strain to fracture, ϵ_f , and the maximum of true flow stress, σ_m , are plotted in Fig. 2(a) and (b), respectively, against nominal strain rate, $\dot{\epsilon}$, for MM Al–Mg/Cu system alloys. The value of ϵ_f is small at low strain rates ($\dot{\epsilon} < \sim 1 \times 10^{-2}/s$), takes maxima at intermediate strain rates ($\sim 1 \times 10^{-2}/s < \dot{\epsilon} < \sim 5 \times 10^1/s$) and reduces again at high strain rates ($\dot{\epsilon} > \sim 5 \times 10^1/s$). These three strain rate regions are hereafter called “regions I, II and III”, respectively. It is also seen that the value of ϵ_f increases when alloyed and reaches more than 400% in the case of the combined addition of 1.1 at.% Mg and 1.2 at.% Cu.

The value of σ_m at 748 K is lower when alloyed, compared to that of MM pure Al. The reason for

this will be discussed later. Figure 2(b) shows a log–log plot of σ_m vs $\dot{\epsilon}$, where the slope (the strain rate sensitivity, $m = d(\ln \sigma_m)/d(\ln \dot{\epsilon})$) is smaller both in regions I and III than in region II. The m value in region II increases with alloying, up to 0.4 for MM Al–1.1Mg–1.2Cu. This increase in the m value is an apparent reason for the occurrence of superplastic elongation in MM Al alloys.

Similar changes in ϵ_f and σ_m with alloying were also observed in MM Al–Ge/Si system alloys, though the attained value of ϵ_f (170% in MM Al–0.4Ge–0.4Si) was smaller compared to that in MM Al–1.1Mg–1.2Cu. Hence, the same phenomenological explanation could be applied for this alloy system, too.

It is noticed in Fig. 2 that the strain rate corresponding to a maximum m value does not coincide with that of maximum ϵ_f ; the former is slightly higher than the latter. This may be due to the fact that in Fig. 2(b) the flow stress is plotted against the nominal strain rate, not the true strain rate. It actually decreases with the progress of deformation in the present constant nominal strain rate test.

3.3. Microstructure appropriate for superplastic elongation

Whether superplasticity appears or not should depend on the microstructure. Examples of TEM micrographs taken before deformation are given in Figs 3 and 4 for MM pure Al and MM Al alloys, respectively. The former showed only 30% elongation at most while the latter exhibited an elongation larger than 100%. Although the grain size tends to be slightly larger in the alloys, all the materials studied consist of fine equiaxed grains (<1 μm in diameter). The most notable change in microstructure produced by alloying is reflected in their thermal recovery behavior. The structure of MM pure Al could not be fully recovered even after annealing at 773 K; the crystal lattice is highly

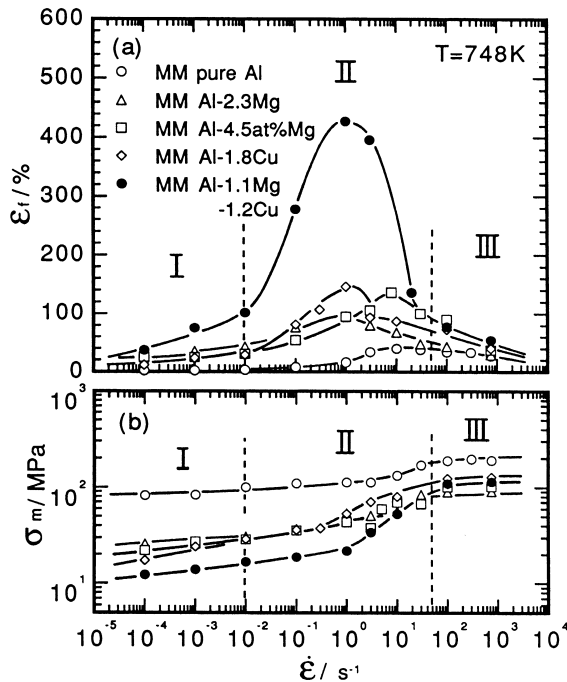


Fig. 2. Changes in (a) nominal strain to fracture ϵ_f and (b) maximum true flow stress σ_m with nominal strain rate $\dot{\epsilon}$ for MM pure Al and MM Al–Mg/Cu system alloys, $T = 748 \text{ K}$.

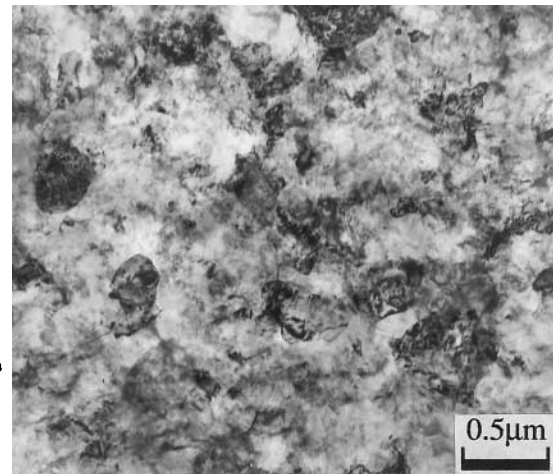


Fig. 3. TEM micrograph of MM pure Al taken before deformation.

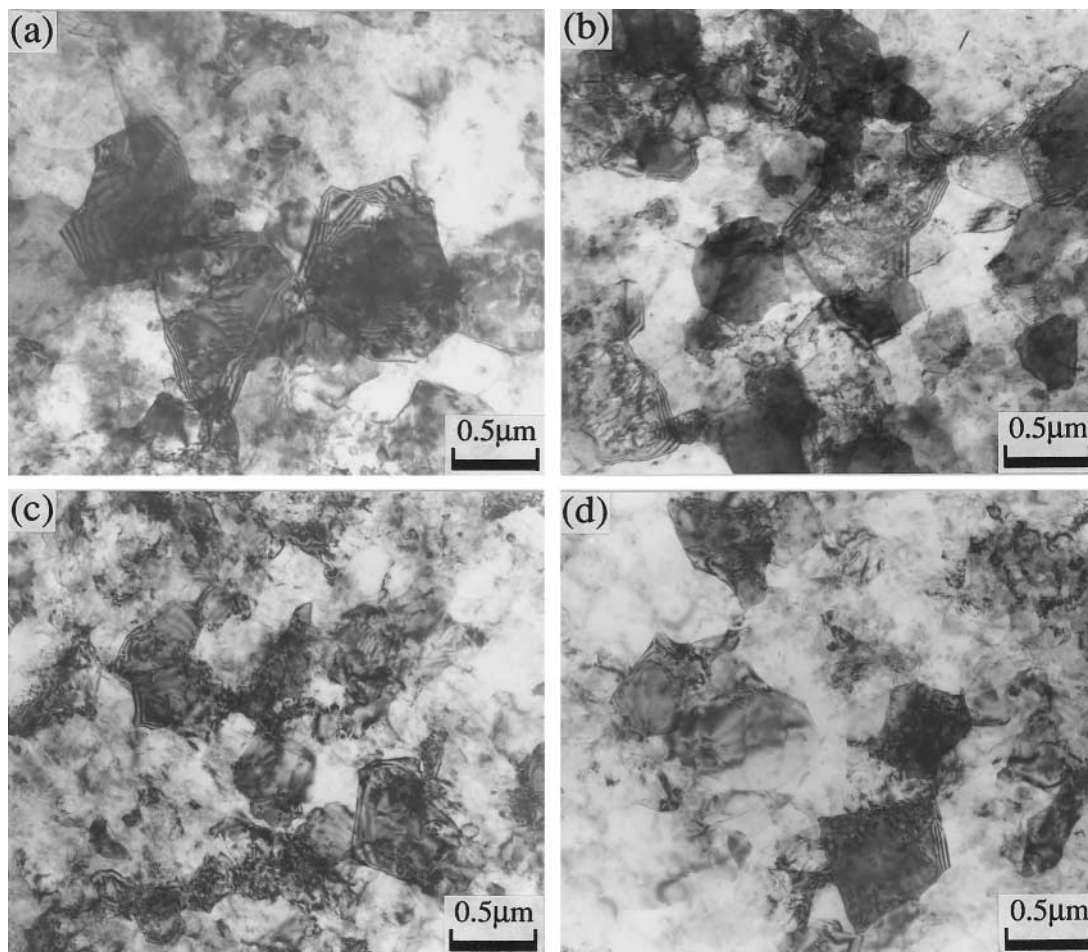


Fig. 4. TEM micrographs of MM Al alloys taken before deformation: (a) MM Al-1.8 at.% Cu; (b) MM Al-1.1 at.% Mg-1.2 at.% Cu; (c) MM Al-0.5 at.% Ge; (d) MM Al-0.4 at.% Ge-0.4 at.% Si.

distorted and the grain boundary is uneven as evidenced by complex contrast in grain interiors and few thickness fringes at grain boundaries (Fig. 3). Thermal recovery progressed more rapidly in MM Al alloys (Fig. 4). It is typically seen in Fig. 4(b) for MM Al-1.1Mg-1.2Cu that exhibited the largest elongation. The structure was almost fully recovered when highly alloyed; the grain interior is rather free from dislocations and the grain boundary is smooth as indicated by clear thickness fringes. Further, misorientation at grain boundaries estimated by an electron diffraction method tended to be larger in the alloys than in MM pure Al: about 10 deg or slightly larger in MM Al-1.1Mg-1.2Cu while a few degrees in MM pure Al.

To verify the TEM results more macroscopically, X-ray diffraction patterns were taken by using Cu-K_α radiation. Part of a Debye ring is shown in Fig. 5 for MM pure Al and MM Al-1.1Mg-1.2Cu that exhibited the smallest and the largest

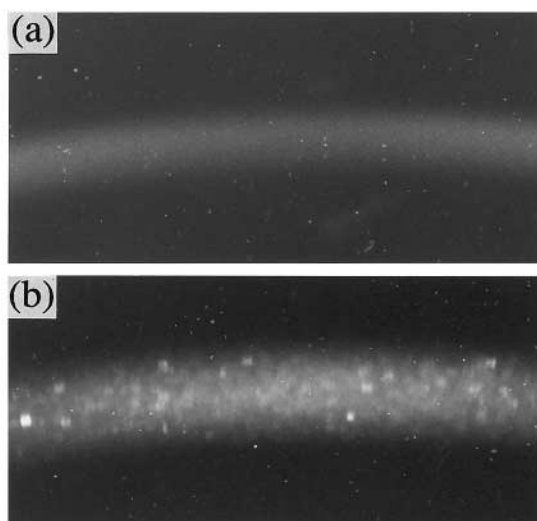


Fig. 5. X-ray Debye rings taken before deformation: (a) MM pure Al; (b) MM Al-1.1 at.% Mg-1.2 at.% Cu.

elongation, respectively. The contrast is diffuse in the former, but is spotty in the latter. This confirms a more advanced state of thermal recovery in the latter.

3.4. On necessity of alloying and prior rolling and annealing for modification of microstructure

3.4.1. Role of alloying. One might wonder why thermal recovery that accompanies density reduction and rearrangement of dislocations [18] is accelerated with alloying, since solute atoms would pin dislocations. As reported in Al–Mg alloys [19], however, there is an opposite possibility that it occurs rapidly in a solid solution, even faster than in pure aluminum, because of a larger stored energy introduced during prior straining.

To verify the faster and/or more extended recovery in the present MM Al alloys, a change in Vickers hardness H_V with temperature was measured in as-rolled specimens by using a high temperature hardness tester. The results are presented in Fig. 6, where the H_V value is higher in the MM Al alloys than in MM pure Al at low temperatures. However, the H_V value for the MM Al alloys reduces more rapidly with increasing temperature, and at temperatures above ~500 K it becomes much lower than that for MM pure Al. Since no evidence of recrystallization was observed even after 773 K annealing, the result of Fig. 6 confirms the acceleration of thermal recovery with alloying. The higher H_V values for the MM Al alloys at the rolling temperature of 423 K suggest that the faster recovery in the alloys originates from a larger stored energy resulting from a higher rolling stress.

3.4.2. Role of prior rolling and annealing. If the above explanation for rapid recovery is valid, lowering the rolling temperature may accelerate thermal recovery and hence lead to a larger elongation to

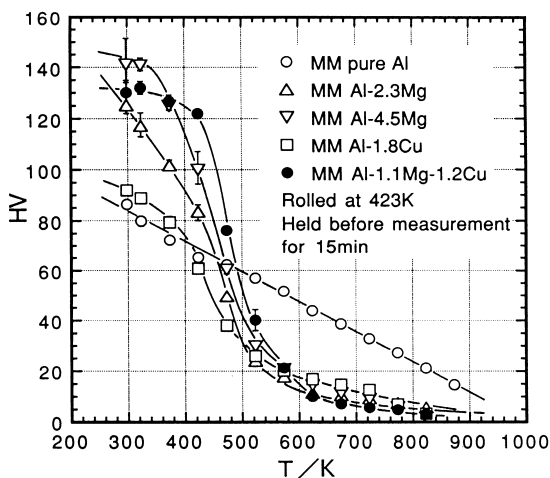


Fig. 6. Changes in Vickers hardness H_V with temperature in MM pure Al and MM Al–Mg/Cu system alloys after rolling of 70% in thickness reduction at 423 K.

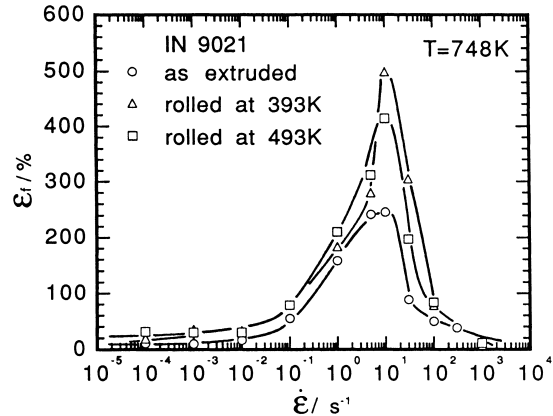


Fig. 7. Effect of temperature in prior rolling (thickness reduction: 70%) on strain to fracture ϵ_f in IN9021 alloy.

fracture. As shown in Fig. 7 for IN9021 alloy that was made from mechanically milled powder and is similar to MM Al–1.1Mg–1.2Cu in composition, a lower rolling temperature results in a larger elongation, indeed.

Further, increases in the annealing temperature and time after rolling would stimulate thermal recovery of microstructure and again lead to a larger elongation. To ascertain this, the elongation and microstructure of MM pure Al were examined after annealing at various conditions. Results are given in Fig. 8; even MM pure Al exhibited an elongation as large as 120% [Fig. 8(a)] when microstructure had been thermally recovered [Fig. 8(b)]. A decrease in the optimum strain rate for the largest elongation with increasing temperature and time of prior annealing is a result of the grain coarsening during annealing.

Tsuzaki *et al.* [20] reported for $(\alpha + \gamma)$ microduplex stainless steel that a boundary misorientation increases by repeating cold or hot rolling and annealing and subgrain boundaries change to grain boundaries. Accordingly, the situation suitable for grain boundary sliding, that is believed to be necessary for superplastic deformation, is created [21]. The increase in misorientation was ascribed to the absorption of dislocations introduced during rolling into subgrain boundaries. The previously stated increase in strain to fracture by prior rolling and annealing (Figs 7 and 8) is considered to be a result of the similar microstructural evolution.

It is therefore postulated that not only the smallness of grains but also a favorable character of grain boundaries (smoothness and large misorientation) are necessary for the occurrence of superplastic elongation at high strain rates; the prior rolling and annealing are useful for producing such a well-defined, fine microstructure in MM Al alloys.

3.5. Deformation mode

To understand the difference in deformation mode under superplastic and non-superplastic con-

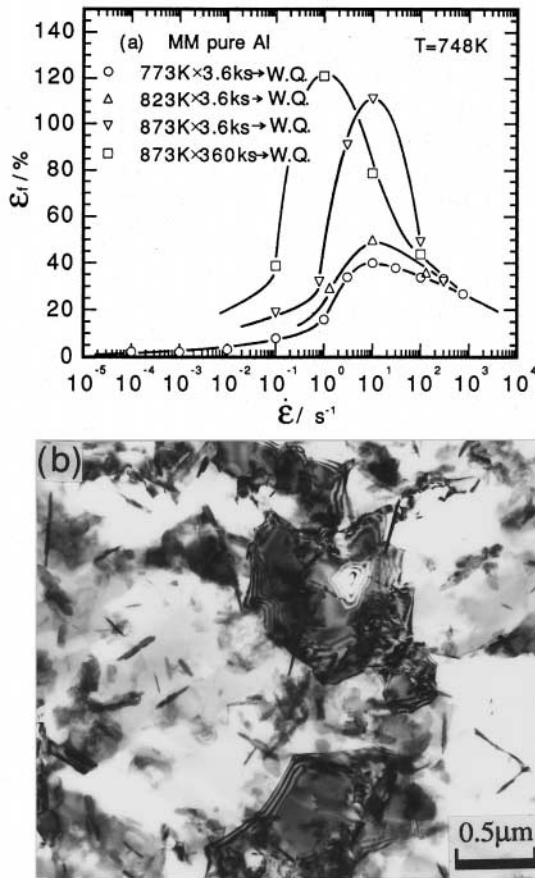


Fig. 8. (a) Effect of temperature and time in prior annealing on strain to fracture ϵ_f . (b) TEM micrograph after annealing at 873 K for 100 h, in MM pure Al, after prior rolling of 70% in thickness reduction at 423 K.

ditions, electrochemically polished surfaces of the specimen were examined with SEM after deformation. Figure 9 shows micrographs of the surface in MM pure Al and MM Al-1.1Mg-1.2Cu; the former exhibited the smallest elongation and the latter the largest elongation, respectively.

In MM pure Al, the surface is fairly flat after deformation in all the regions I, II and III even in an area of local necking. This implies that the main mode of deformation is the grain deformation due to dislocation motion. The surface condition is similar even for MM Al-1.1Mg-1.2Cu when deformed in region III. However, after deformation in regions I and II, the surface is uneven in a unit of a grain or several grains. Hence, it is reasonably considered that grain boundary sliding occurred during superplastic deformation in region II. At present, however, a contribution of grain boundary sliding to total elongation and a reason for the difference in elongation between regions I and II still remain unknown.

4. CONCLUDING REMARKS

On the basis of experimental results, the origin of superplastic elongation in MM Al alloys could be explained as follows.

Alloying of Mg, Cu, Ge and/or Si, especially the combined addition of Mg and Cu, and prior rolling accelerate thermal recovery when annealed at high temperature. Microstructure changes from the highly strained state in MM pure Al to well-defined but still fine grained structure in MM Al alloys in which dislocation density within grains is low and

	Region I ($\dot{\epsilon} = 1 \times 10^{-3}/s$)	Region II ($\dot{\epsilon} = 1 \times 10^0/s$)	Region III ($\dot{\epsilon} = 7.5 \times 10^2/s$)
MM pure Al			
MM Al-1.1Mg -1.2Cu			

Fig. 9. SEM micrographs of specimen surface after deformation in MM pure Al and MM Al-1.1 at.% Mg-1.2 at.% Cu.

grain boundaries are smooth and of large misorientation. Such sharpening of the microstructure reduces flow stresses due to both grain deformation at high strain rates ($> \sim 5 \times 10^1/s$) and grain boundary sliding at low strain rates ($< \sim 1 \times 10^{-2}/s$). Probably because a decrease in the logarithm of flow stress due to the latter mechanism is larger than that due to the former, the strain rate sensitivity (the m value) increases at intermediate strain rates ($1 \times 10^{-1} - 1 \times 10^1/s$). This large m value suppresses the occurrence of local necking, and superplasticity appears at strain rates around $10^0/s$.

Grain boundary sliding certainly occurs during superplastic deformation, though its role for the appearance of superplasticity remains unknown in detail. It is concluded that not only the smallness of grains but also the smoothness of grain boundaries are indispensable for the occurrence of superplasticity in MM Al alloys.

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REFERENCES

1. Benjamin, J. S., *Metall. Trans.*, 1970, **1**, 2943.
2. Benjamin, J. S. and Bomford, M. J., *Metall. Trans.*, 1977, **8A**, 1301.
3. Singer, R. F., Oliver, W. C. and Nix, W. D., *Metall. Trans.*, 1980, **11A**, 1895.
4. Hasegawa, T. and Miura, T., in *Proc. Int. Conf. on Structural Applications of Mechanical Alloying*, ed. F. H. Froes and J. J. deBarbadillo. ASM Int., Metals Park, 1990, p. 213.
5. Nieh, T. G., Gilman, P. S. and Wadsworth, J., *Scripta metall.*, 1985, **19**, 1375.
6. Bieler, T. R., Nieh, T. G., Wadsworth, J. and Mukherjee, A. K., *Scripta metall.*, 1988, **22**, 81.
7. Higashi, K. and Mabuchi, M., in *Proc. Int. Conf. on Advanced Composite Materials*, ed. T. Chandra *et al.* TMS, Warrendale, 1993, p. 35.
8. Mishra, R. S., Bieler, T. R. and Mukherjee, A. K., *Acta metall. mater.*, 1995, **43**, 877.
9. Mukherjee, A. K., Mishra, R. S. and Bieler, T. R., *Mater. Sci. Forum*, 1997, **233-234**, 217.
10. Higashi, K., Okada, T., Mukai, T. and Tanimura, S., in *Superplasticity in Advanced Materials ISCAM-91*, ed. S. Hori, M. Tokizane and N. Furushiro. Japan Soc. Res. on Superplasticity, 1991, p. 569.
11. Higashi, K., Okada, T., Mukai, T. and Takamura, S., *Scripta metall. mater.*, 1992, **26**, 761.
12. Hasegawa, T., Komatsu, T., Miura, T. and Takahashi, T., in *Proc. Int. Conf. on Aspects of High Temperature Deformation and Fracture in Crystalline Materials*, ed. Y. Hosoi *et al.* Japan Inst. Metals, Sendai, 1993, p. 439.
13. Hales, S. J. and McNelley, T. R., in *Proc. Conf. on Superplasticity in Aerospace*, ed. H. C. Heikkinen and T. R. McNelley. TMS, Warrendale, 1988, p. 61.
14. Mishra, R. S., Echer, C., Bampton, C. C., Bieler, T. R. and Mukherjee, A. K., *Scripta mater.*, 1996, **35**, 247.
15. Hasegawa, T., Miura, T., Takahashi, T. and Yakou, T., *ISIJ Int.*, 1992, **32**, 902.
16. Bieler, T. R. and Mukherjee, A. K., *Mater. Sci. Engng*, 1990, **A128**, 171.
17. Yasuno, T., Hasegawa, T. and Takahashi, T., *Proc. Int. Symp. on Microstructure, Micromechanics and Processing of Superplastic Materials*. Mie Academic Press, in press.
18. Cahn, R. W., in *Physical Metallurgy*, ed. R. W. Cahn and P. Haasen. Elsevier Science, Amsterdam, 1996, p. 2399.
19. Barioz, C., Brechet, Y., Legresy, J. M., Cheynet, M. C., Courbon, J., Guyot, P. and Ratnaud, G. M., *Proc. 3rd Int. Conf. on Aluminium*, Trondheim, 1992, p. 347.
20. Tsuzaki, K., Xiaouxu, H. and Maki, T., *Acta mater.*, 1996, **44**, 4491.
21. Tsuzaki, K., Matsuyama, H., Nagao, M. and Maki, T., *Mater. Trans. JIM*, 1990, **31**, 983.