Available online at www.sciencedirect.com
ScienceDirect
Scripta Materialia 60 (2009) 968–971
Scripta MATERIALIA
www.elsevier.com/locate/scriptamat

Low-temperature superplasticity of Al–Mg–Sc alloy produced by friction stir processing

F.C. Liu,[a] Z.Y. Ma[a,*] and L.Q. Chen[b]

[a]Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, 72 Wenhua Road, Shenyang 110016, China

[b]State Key Laboratory of Rolling and Automation, Northeastern University, Shenyang 110004, China

Received 13 January 2009; revised 4 February 2009; accepted 12 February 2009
Available online 20 February 2009

Ultrafine-grained (0.7 μm) Al–Mg–Sc alloy with an approximately random misorientation distribution and predominantly high-angle boundaries of 97% was produced by friction stir processing. A ductility of 235% was obtained at 200 °C. Increasing temperature from 200 to 300 °C resulted in an increase in superplasticity, optimum strain rate and strain rate sensitivity. Low temperature and high strain rate superplasticity with a ductility of 620% was achieved at 300 °C and $3 \times 10^{-2}\ s^{-1}$. Abnormal grain growth occurred at 350 °C, resulting in the disappearance of superplasticity.

Keywords: Superplasticity; Friction stir processing; Ultrafine-grained structure; Aluminum alloys; Friction stir welding

It is well documented that grain refining in materials results in either an increase in strain rate or a decrease in the temperature at which superplasticity appears [1–3]. By reducing the grain size to 0.3 μm, Valiev et al. [4] first reported the possibility of achieving low-temperature superplasticity (LTSP), which was defined as the occurrence of superplastic elongations at temperatures significantly lower than those generally required for superplasticity. In their work, Valiev et al. [4] obtained an elongation of ~250% in the ultrafine-grained (UFG) Al–Cu–Zr alloy at 220 °C corresponding to 0.53 T_m, where T_m is the melting temperature of the material, expressed by K. This provided a clear demonstration of the potential for achieving superplasticity at relatively low temperatures through refining the grains to the submicrometer level.

Subsequently, several studies have reported the occurrence of LTSP in aluminum alloys with ultrafine grain sizes, where grain refinement was achieved by using different processing techniques [5–9], such as thermomechanical processing (TMP) [5,6], equal-channel angular pressing (ECAP) [7,8] and high-pressure torsion (HPT) [9]. Those studies showed that highly refined grain sizes, sometimes below 1.0 μm in size, can be produced and that grain refinement enhances superplasticity.

Recently, a solid-state processing technique, friction stir processing (FSP), was developed as a generic tool for microstructure modification based on the basic principles of friction stir welding (FSW) [10–12]. Through a single-pass FSP, bulk fine-grained materials with predominantly high-angle grain boundaries (HAGBs) were produced. A previous study [3] has shown that FSP 7075Al alloy with an UFG structure of 0.8 μm exhibited superplastic ductility over a wide low temperature range of 200–350 °C. At 200 °C, a ductility of 350% was observed at a strain rate of $1 \times 10^{-5}\ s^{-1}$. Furthermore, LTSP was obtained at 175 °C (0.48 T_m) in an UFG Al–4Mg–1Zr prepared via FSP [13]. These results clearly show the effectiveness of FSP for producing UFG materials that are amenable to LTSP.

Al–Mg–Sc alloys were developed as a class of highly formable aluminum alloys with superior properties (especially yield strength) compared to conventional Al–Mg alloys with the same magnesium content [14]. They have the potential to be applied in the fabrication of airframes, stiffened panels and tanks for condensed gas storage. A previous study [15] indicated that LTSP was obtained at 200 °C in an Al–3Mg–0.2Sc alloy with a grain size of 0.2 μm prepared by ECAP. The alloys containing 3% Mg have only a limited commercial application due to their low strengths. Increasing the

* Corresponding author. Tel./fax: +86 24 83978908; e-mail: zyma@imr.ac.cn

1359-6462/$ - see front matter
doi:10.1016/j.scriptamat.2009.02.021

magnesium content in Al–Mg–Sc alloys increases their tensile strength significantly. Hence Al–Mg–Sc alloys with high magnesium content can be used as load-carrying structures.

In this study, an Al–5.3Mg–0.23Sc alloy was subjected to FSP and superplastic investigation. The aim is (i) to understand the microstructural evolution of the Al–Mg–Sc alloy subjected to FSP with rapid cooling; (ii) to examine the possibility of achieving LTSP in the FSP UFG Al–Mg–Sc alloy with high Mg content at 200 °C; and (iii) to examine the thermal stability of the FSP UFG Al–Mg–Sc alloy at elevated temperatures.

A 300 mm (length) × 70 mm (width) × 8 mm (height) extruded plate with a composition of Al–5.33Mg–0.23Sc–0.49Mn–0.14Fe–0.06Zr (in wt.%) was used in this study. The detailed description of the extrusion process has been presented in a previous work [16]. A single pass of FSP was carried out on the extruded plate at a tool rotation rate of 400 rpm and a traverse speed of 100 mm min^{-1} with immediate room temperature water quenching of the plate. The total FSP length was 270 mm from pin entry to pin exit. A steel tool with a concave shoulder 14 mm in diameter, a threaded conical pin 5 mm in root diameter, 3.5 mm in tip diameter, and 4.5 mm in length was used. Microstructural characterization was performed on the cross-section of the stir zone (SZ) transverse to the FSP direction by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The samples for SEM were lightly electropolished to produce a strain-free surface. Electron backscatter diffraction (EBSD) orientation maps (with a resolution of 80 nm) were obtained using a Zeiss Supra 35, operated at 20 kV, and interfaced to an HKL Channel EBSD system. Thin foils for TEM were prepared by twin-jet polishing using a solution of 70% methanol and 30% nitric acid at −35 °C and 19 V.

Dogbone-shaped superplastic tensile specimens (2.5 mm gage length, 1.4 mm gage width, and 1.0 mm gage thickness) were electrodischarge machined from the SZ of the FSP sample transverse to the FSP direction. These samples were subsequently ground and polished to a final thickness of ~0.8 mm. Constant crosshead speed tensile tests were conducted using an Instron 5848 micro-tester. Each sample was held at the testing temperature for about 20 min in order to reach thermal equilibrium. To check the thermal stability of the UFG introduced by FSP, small samples with dimensions of 8 × 8 × 10 mm^3 cut from the FSP plate were statically annealed for 20 min at temperatures of 200, 300 and 350 °C, respectively, and then cooled rapidly in water to provide microstructural information of the FSP samples just before tensile test at various temperatures.

Figure 1a shows the microstructure of the FSP Al–Mg–Sc sample obtained by EBSD mapping. FSP resulted in the generation of the fully recrystallized microstructure with a uniform and equiaxed grain size distribution, and the average grain size was determined to be 0.7 μm. Microstructure evolution during FSW/FSP has been widely investigated in previous studies [17–19]. It was reported that the initial size of newly recrystallized grains was of the order of 25–100 nm and grew to 2–5 μm, a size equivalent to that found in

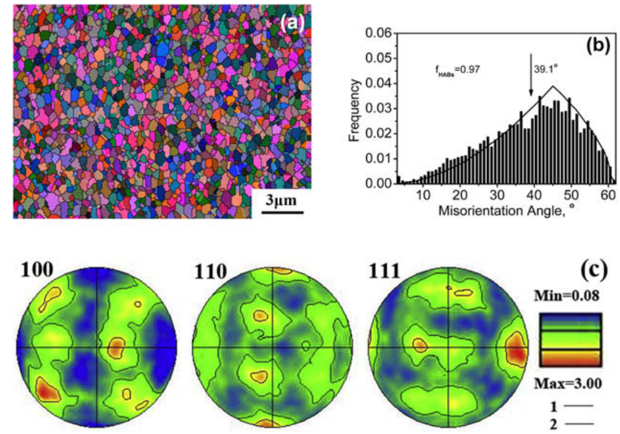

Figure 1. Microstructure of FSP Al–Mg–Sc alloy: (a) EBSD map, (b) boundary misorientation angle distribution, and (c) pole figures (sample surface was perpendicular to the FSP direction).

FSP aluminum alloys, when heated for 1–4 min at 350–450 °C [19]. Therefore, it is possible to produce an ultrafine-grained structure by inhibiting the growth of the recrystallized grains by cooling the FSP region immediately behind the pin tool. The production of the ultrafine grains in the FSP Al–Mg–Sc sample is attributed to active cooling and the small tool size used. Similar results have been reported previously in FSP 7075Al alloy [3].

The misorientation angle histogram of the FSP Al–Mg–Sc sample is shown in Figure 1b. This misorientation distribution is very close to a random grain assembly predicted by Mackenzie for randomly oriented cubes [20]. The fraction of HAGBs was 97%, the same as that predicted for a truely random grain assembly [20]. The average misorientation angle was determined to be 39.1°, which is very close to the 40.7° predicted by Mackenzie for a random misorientation distribution [20]. Furthermore, it was revealed that the FSP Al–Mg–Sc sample exhibited a very weak texture component as indicated by the pole figures in Figure 1c. This weak texture component is near to rotated cube orientation $\{001\}\langle 110\rangle$, or so-called shear component, which comes from the remnant of shear texture. The shear component was usually formed with inhomogeneous shear deformation due to the FSP.

The misorientation distribution for the FSP Al–Mg–Sc sample is somewhat similar to the previous report for FSW 7071Al which contained 80–90% of HAGBs [21]. However, the present result is different from the previous reports about cold-rolled Al–6Cu–0.4Zr and Al–5Mg–0.2Sc alloys in which two peaks were observed at 10–15° and 50–60° in the misorientation distribution histograms [22,23]. During superplastic deformation, grain boundary sliding (GBS) deformation accommodated by random grain rotation resulted in an essentially random misorientation distribution. Compared with the previous reports, it can be concluded that FSP is an effective method of producing fine-grained materials predominated by HAGBs. The approximately random misorientation distribution of the FSP Al–Mg–Sc alloy may be suitable for superplastic deformation associated with GBS. Thus, if the ultrafine grains with predominant

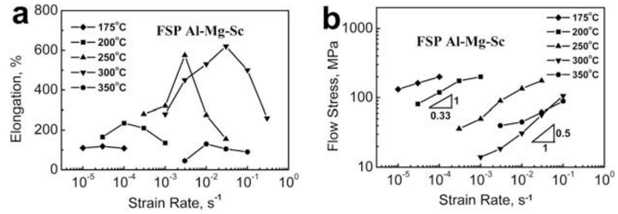

Figure 2. Variation of (a) elongation and (b) flow stress with initial strain rate for FSP Al–Mg–Sc alloy.

HAGBs have the capability to resist intergranular separation and significant grain growth at high temperature, it will be possible to achieve superplastic flow accommodated by GBS.

Figure 2a shows the variation in elongation with initial strain rate for the FSP Al–Mg–Sc sample. Maximum elongation of 115% was observed at 175 °C. This means that the FSP Al–Mg–Sc sample did not exhibit superplasticity at a low temperature of 175 °C. At 200 °C (0.51 T_m), a ductility of 235% was obtained at a low strain rate of 1×10^{-4} s^{-1}. A temperature increase from 200 to 300 °C resulted in an increase in the optimum strain rate for superplasticity as well as the maximum elongation. Maximum superplasticity of 620% was achieved at a low temperature of 300 °C and a high strain rate of 3×10^{-2} s^{-1}. This result is attractive because a single-pass FSP could induce the occurrence of LTSP in the Al–Mg–Sc alloy at a high strain rate. However, at 350 °C, the elongations dropped drastically and were consistently lower than 150% for the various strain rates.

Figure 2b shows the variation of flow stress (at true strain of 0.1) with the initial strain rate for the FSP Al–Mg–Sc sample. An m value of 0.3 or more and an elongation >200% has been considered to be a rough indicator of the appearance of superplasticity [24]. At 175 °C, the strain rate sensitivity (m) was consistently lower than 0.2, which is consistent with the absence of superplasticity, as shown in Figure 2a. At 200 °C, an m value of 0.33 was achieved at 1×10^{-4} s^{-1}, indicating that LTSP was developed at 200 °C in the FSP UFG Al–Mg–Sc alloy. The strain rate sensitivity for maximum superplasticity increased with increasing temperature from 200 to 300 °C. At 300 °C, the m value increased to ~0.5 at the strain rate of 1×10^{-2}–1×10^{-1} s^{-1}, the condition for GBS dominating the flow process. At 350 °C, the m values were consistently lower than 0.3 for various strain rates. The low m values correspond to the low elongation in the FSP sample at 350 °C.

Figure 3 shows the TEM microstructures of the as-FSP and annealed FSP Al–Mg–Sc samples. The as-FSP sample contained a low density of dislocations in the large grains. For the grains smaller than 200 nm in size, few dislocations were observed in the grains. This dislocation distribution is somewhat similar to that in the UFG 7075Al prepared by FSP with rapid cooling [18]. Figure 3b and c show that the grains in the FSP Al–Mg–Sc alloy exhibited a slowly and essentially uniform growth when annealed at 200 and 300 °C due to the presence of fine Al_3Sc precipitates both at the grain boundaries and within the grain interiors. The measured average grain sizes in the annealed FSP samples are ~1.0 and ~1.2 μm for 200 and 300 °C, respectively. Such a fine and stable microstructure is suitable for superplastic deformation at these temperatures. Furthermore, it is noted that with the increase of the temperature, the density of dislocations in the grains reduced and the grain boundaries became more distinct. When annealed at 350 °C for 20 min, the FSP sample displayed a duplex microstructure with a small number of large grains being surrounded by arrays of fine grains, indicating the occurrence of abnormal grain growth (AGG). Such a microstructure no longer meets the requirements for superplastic flow and accounts for the disappearance of superplasticity at 350 °C (Fig. 2a).

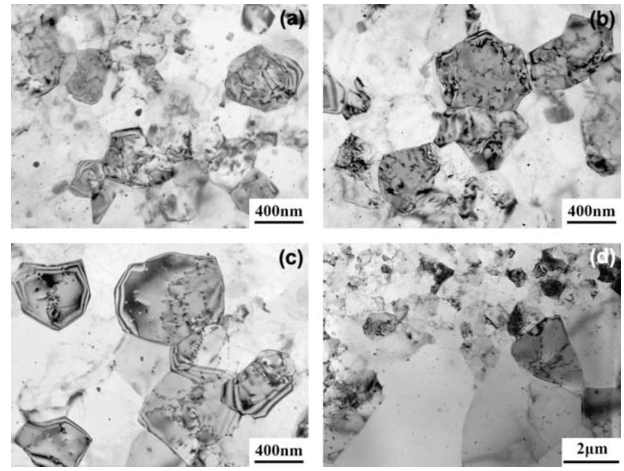

Figure 3. TEM microstructures of (a) as-FSP Al–Mg–Sc alloy and (b–d) FSP Al–Mg–Sc alloy annealed for 20 min at (b) 200 °C, (c) 300 °C and (d) 350 °C.

In the previous reports, AGG in FSP/FSW aluminum alloys was often observed to initiate at the SZ/base metal (BM) interfaces, the material surface in contact with the FSP tool or the root of the SZ [21,25–28]. In this study, when the as-FSP sample was annealed at

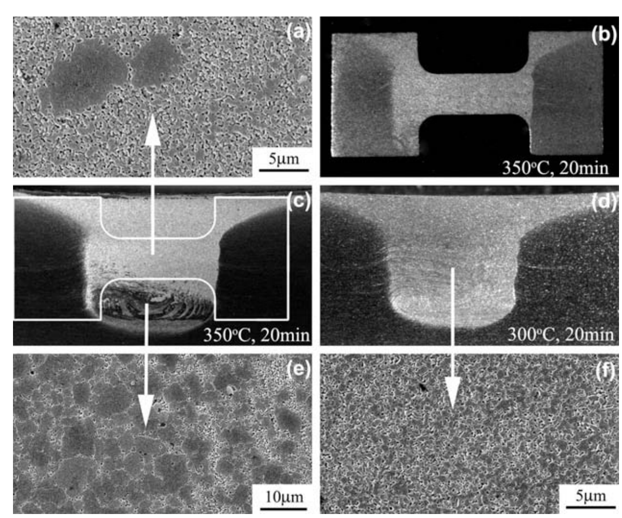

Figure 4. Microstructure evolution in FSP Al–Mg–Sc alloy annealed at different temperatures.

350 °C for 20 min, distinct AGG occurred at the bottom of the SZ (Fig. 4c and e), whereas in the middle of the SZ, the annealed FSP sample displayed a duplex microstructure with a small number of large grains being surrounded by arrays of fine grains (Fig. 4a). Similar microstructure was also observed at the gage of the tensile specimen which was annealed at 350 °C for 20 min (Fig. 4b). This duplex microstructure is also revealed by the TEM examination (Fig. 3d). The FSP sample annealed at 300 °C did not show any AGG phenomena (Fig. 4d and f). It was reported that AGG initiated at the root of the SZ developed into the central region of the SZ upwards with increasing annealing time [21]. Therefore, for the tensile specimen with the gage being machined from only the middle part of the SZ as shown in Figure 4, the effect of AGG in the bottom of the SZ on AGG development in the gage of the tensile specimen might be significantly reduced.

ECAP and cold-rolled UFG Al–Mg–Sc alloys with similar compositions as the present alloy did not show AGG at testing temperatures as high as 450 °C [29,23]. Therefore, the development of AGG at 350 °C in the FSP UFG Al–Mg–Sc alloy is somewhat surprising. For FSP/FSW alloys, onion rings or banded structures with alternately changed grain size or particle distribution are a prominent feature of the SZ due to the geometric effect resulting from the tool movement [30]. AGG occurring in the root of the SZ showed distinct growth characteristics along the onion rings (Fig. 4c). This result is consistent with previous studies [21,27]. Rios [31] believed that the local environment is constant only on average, and is locally subjected to microstructural fluctuations about this average. These microstructural fluctuations can consist in either a locally lower pinning force or a locally higher driving force. Further, the FSP region contained some large grains that have seven or more faces surrounded by much smaller grains (Fig. 3). From the topological theory of grain growth, these large grains have a higher thermodynamic force for grain growth than the rest of the grains [32]. These pre-existing large grains and local microstructural fluctuations may be responsible for the AGG in the tensile specimens.

In summary, the following conclusions are reached:

(1) Fine grains 0.7 μm in size with predominantly (97%) HAGBs were obtained in Al–Mg–Sc alloy by FSP with rapid cooling. The misorientation distribution was very close to a random grain assembly for randomly oriented cubes.
(2) Superplasticity of 235–620% was achieved at low temperatures of 200–300 °C. A temperature increase from 200 to 300 °C resulted in an increase in the optimum strain rate as well as the maximum elongation. The FSP Al–Mg–Sc exhibited a ductility of 620% at a lower temperature of 300 °C and a higher strain rate of $3 \times 10^{-2}\ s^{-1}$.
(3) The FSP material kept a fine grain size of ∼1 μm just before the tensile test at 200–300 °C. However, AGG occurred before the tensile test at 350 °C, resulting in the disappearance of superplasticity.

This work was supported by the National Natural Science Foundation of China under Grant Nos. 50671103 and 50871111, the National Basic Research Program of China under Grant No. 2006CB605205, the National Outstanding Young Scientist Foundation under Grant No. 50525103, and the Hundred Talents Program of Chinese Academy of Sciences.

[1] R.Z. Valiev, D.A. Salimonenko, N.K. Tsenev, P.B. Berbon, T.G. Langdon, Scripta Mater. 37 (1997) 1945.
[2] N. Tsuji, K. Shiotsuki, Y. Saito, Mater. Trans. 40 (1999) 765.
[3] F.C. Liu, Z.Y. Ma, Scripta Mater. 58 (2008) 667.
[4] R.Z. Valiev, O.A. Kaibyshev, R.I. Kuznetsov, R.Sh. Musalimov, N.K. Tsenev, Dokl. Akad. Nauk SSSR 301 (1988) 964.
[5] T.R. McNelley, R. Crooks, P.N. Kalu, S.A. Rogers, Mater. Sci. Eng. A 106 (1993) 135.
[6] I.C. Hsiao, J.C. Huang, Metall. Mater. Trans. A 33 (2002) 1373.
[7] S. Komura, Z. Horita, M. Furukawa, M. Nemoto, T.G. Langdon, Metall. Mater. Trans. A 32 (2001) 707.
[8] K.T. Park, D.Y. Hwang, S.Y. Chang, D.H. Shin, Metall. Mater. Trans. A 33 (2002) 2859.
[9] Z. Horita, T.G. Langdon, Scripta Mater. 58 (2008) 1029.
[10] W.M. Thomas, E.D. Nicholas, J.C. Needham, M.G. Murch, P. Templesmith, C.J. Dawes, G.B. Patent Application No. 9125978.8, December 1991.
[11] R.S. Mishra, M.W. Mahoney, S.X. McFadden, N.A. Mara, A.K. Mukherjee, Scripta Mater. 42 (2000) 163.
[12] Z.Y. Ma, R.S. Mishra, M.W. Mahoney, Acta Mater. 50 (2002) 4419.
[13] Z.Y. Ma, R.S. Mishra, Scripta Mater. 53 (2005) 75.
[14] Y.A. Filatov, V.I. Yelagin, V.V. Zakharov, Mater. Sci. Eng. A 280 (2000) 97.
[15] S. Ota, H. Akamatsu, K. Neishi, M. Furukawa, Z. Horita, T.G. Langdon, Mater. Trans. 43 (2002) 2364.
[16] F.C. Liu, Z.Y. Ma, Scripta Mater. 59 (2008) 882.
[17] R.W. Fonda, J.F. Bingert, K.J. Colligan, Scripta Mater. 51 (2004) 243.
[18] J.Q. Su, T.W. Nelson, C.J. Sterling, Mater. Sci. Eng. A 405 (2005) 277.
[19] C.G. Rhodes, M.W. Mahoney, W.H. Bingel, M. Calabrese, Scripta Mater. 48 (2003) 1451.
[20] J.K. Mackenzie, Biometrika 45 (1958) 229.
[21] K.h.A.A. Hassan, A.F. Norman, D.A. Price, P.B. Prangnell, Acta Mater. 51 (2003) 1923.
[22] M. Eddahbi, T.R. Mcmelley, O.A. Ruano, Metall. Mater. Trans. 32A (2001) 1093.
[23] R. Kaibyshev, E. Avtokratova, A. Apollonov, R. Davies, Scripta Mater. 54 (2006) 2119.
[24] R.H. Bricknell, J.W. Edington, Met. Trans. A 7 (1976) 153.
[25] M.M. Attallah, H.G. Salem, Mater. Sci. Eng. A 391 (2005) 51.
[26] H.G. Salem, Scripta Mater. 49 (2003) 1103.
[27] I. Charit, R.S. Mishra, Scripta Mater. 58 (2008) 367.
[28] K.N. Krishnan, J. Mater. Sci. 37 (2002) 473.
[29] F. Musin, R. Kaibyshev, Y. Motohashi, G. Itoh, Scripta Mater. 50 (2004) 511.
[30] G.R. Cui, Z.Y. Ma, S.X. Li, Scripta Mater. 58 (2008) 1082.
[31] P.R. Rios, Scripta Mater. 39 (1998) 1725.
[32] P.R. Rios, M.E. Glicksman, Acta Mater. 54 (2006) 5313.