Enhanced superplasticity in an extruded high strength Mg–Gd–Y–Zr alloy with Ag addition

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Abstract

The effect of 2 wt% Ag addition on the superplastic behavior of an extruded Mg–8.5Gd–2.5Y–0.5Zr (wt%) alloy was investigated by impression testing in the temperature range of 523–580 K. The average sizes of the dynamically recrystallized grains of the Ag-free and Ag-containing alloys were about 8 and 3 µm, respectively. Analysis of electron backscattered diffraction (EBSD) data confirmed the higher fractions of high-angle grain boundaries (HAGBs) in the Ag-containing alloy. The deformation response of this alloy in proper temperature range conforms to regions I, II and III, typical of superplastic deformation behavior. The addition of Ag to the base alloys led to enhanced superplasticity in region II by increasing the strain rate sensitivity (SRS) indices (m-values) from 0.25 to 0.51 and 0.36 to 0.46 at 573 and 598 K, respectively. These high m-values together with the activation energy of 181 kJ/mol suggest that the major mechanism involved in superplastic deformation is grain boundary sliding (GBS) accommodated by lattice diffusion at temperatures above 573 K.

1. Introduction

Magnesium alloys, as the lightest structural materials, have a great potential to replace denser materials due to many advantages, such as low density and high specific strength [1]. In spite of these advantages, Mg alloys exhibit low ductility due to the lack of slip systems near room temperature [2]. Therefore, attempts have been made to enhance their formability through superplastic deformation in order to fabricate light structural components with complex shapes [3,4]. The basic requirements for superplasticity are equiaxed small grain sizes typically smaller than 10 µm, high angle grain boundary (HAGB), high temperature deformation, and controlled strain rate [5–7]. Although severe plastic deformation techniques have been employed to fulfill such requirements, they are often time consuming and costly. Accordingly, it is quite desirable to achieve fine-grained microstructures, through the use of proper alloying elements and conventional deformation methods such as hot extrusion.

Among different Mg alloys, those based on the Mg–Gd system are very promising, due to their high strength and thermal stability [8,9]. Their properties can even be further improved by enhancing their ductility through incorporating superplastic behavior. Superplastic characteristics of an extruded Mg–9Gd–4Y–0.4Zr alloy, with an initial grain size of 10 µm, have been studied by carrying out tensile tests at various temperatures and strain rates [10]. The strain rate sensitivity (SRS) index of m = 0.54 and high ductility of 410%, obtained at the temperature of 723 K and strain rate of 2 × 10^{-4} s^{-1}, was attributed to grain boundary sliding accommodated by dislocation motion assisted by lattice diffusion. It was concluded that MgGd and MgGd,Y or β-phase particles had significant effect on grain boundary pinning, and that the strain was transformed from the matrix to deformable irregular-shape β-phase particles. The superplasticity of the same material with an average grain size of about 6.5 µm has also been investigated by the shear punch testing technique, in which an m-value of 0.4 has been recorded at 723 K [11]. The superplastic behavior of the extruded Mg–7Y–4Gd–1Zn alloy investigated by tensile test has yielded m values of about 0.55 and maximum elongations of 700% at 743 K [12]. The observed behavior was attributed to the disappearance of long period stacking ordered phase and the formation of a cubic phase.

Although tensile test has long been used as a common means for the assessment of superplasticity by recording high elongation values, other testing methods which are capable of measuring SRS index or m-value have also found application in this field. Various localized testing techniques such as indentation [6,13], impression [14–17], and shear punch testing [18] have been recently...
employed for such a purpose. Among all of these methods, impression test is of special interest since it uses a flat-ended cylindrical punch, which provides a constant stress state during the test. The measured $m$-values and activation energies can be used to assess the superplastic behavior and the operating mechanisms. The present work examines the effect of 2 wt% Ag addition on the superplasticity of an extruded Mg–8.5Gd–2.5Y–0.5Zr alloy via the impression testing technique in the temperature range of 523–598 K. The addition of Ag to the Mg–Gd alloy system has been found to significantly enhance room-temperature mechanical properties [19,20].

2. Experimental procedures

The materials used were Mg–8.5Gd–2.5Y–0.5Zr alloy with an actual chemical composition of Mg–8.2 wt% Gd–2.44 wt% Y–0.48 wt% Zr, and the same alloy with 2 wt% Ag addition, determined by inductively coupled plasma (ICP) analysis. The details of melting, alloying, and casting processes have been described elsewhere [21]. The cast billets of 42 mm diameter and 120 mm length were homogenized at 733 K for 6 h and extruded to 13 mm bars at 673 K. The metallographic samples were etched with a solution of 5 g picric acid, 10 cc acetic acid, and 95 cc ethanol. The microstructure of the cross sections perpendicular to the pressing direction of the extruded billets was examined by optical and electron backscattered diffraction (EBSD), from which the distribution of grain boundaries misorientation angle was determined. The specimen preparation procedure for EBSD involved grinding with 1200 grit SiC paper, polishing with 6, 3, and 1 μm oil-based diamond suspension, followed by a colloidal silica slurry, which provided a high quality surface finish for EBSD. The specimen was etched with a solution of 60% ethanol, 20% distilled water, 15% acetic acid and 5% nitric acid.

![Fig. 1. Impression creep curves of (a) Ag-free Mg–8.5Gd–2.5Y–0.5Zr (wt%) and (b) Ag-containing Mg–8.5Gd–2.5Y–0.5Zr–2Ag (wt%) alloy at 573 K under different stress levels.](image1)

![Fig. 2. Applied punch stress as a function of impression velocity for (a) Ag-free and (b) Ag-containing alloy at various temperatures.](image2)

The 4-mm slices cut from the extruded bars were tested in an impression tester. Constant-load impression tests were carried out in the air atmosphere using a split furnace mounted on a universal tensile testing machine. The details of the testing arrangements have been explained elsewhere [22]. Impression creep measurements were made on each sample in the temperature range 523–598 K and under punch stresses in the range 40–1000 MPa for dwell times up to 4000 s. The machine computer system acquired the creep data by the continuous recording of the impression depth as a function of dwell time.

3. Results and discussion

In order to characterize the superplasticity of the tested materials, impression tests were carried out at different temperatures and under different punch stresses for both alloys. Typical impression creep curves obtained under various constant stress levels are shown in Fig. 1. These curves illustrate the impression depth as a function of time for the tested alloys at 573 K. The slopes of the curves indicate that higher creep rates are achieved as the applied stress increases. For both materials there is a rather short primary creep stage, after which they exhibit a relatively linear relationship between the impression depth and dwell time. Further comparison of the slopes of the curves show that for a given stress level, the
Ag-free material possesses a lower penetration rate than the Ag-containing alloy.

It is generally accepted that the high-temperature flow stress ($\sigma$) of superplastic materials can be related to the strain rate ($\dot{\varepsilon}$) by a power-law relationship:

$$\dot{\varepsilon} = A \sigma^m \exp\left(-\frac{Q}{RT}\right)$$

(1)

where $A$ is a material constant, $T$ is the temperature, $R$ is the universal gas constant, $Q$ is the activation energy, and $m$ denotes the strain rate sensitivity (SRS) index. The superplastic deformation behavior of materials is often manifested by their high $m$-values [23].

Assuming an analogy between impression velocity and creep rate [24], one can obtain the following relationship between impression velocity and applied punch stress:

$$V_{imp} = B\left(\sigma_{imp}\right)^m \exp\left(-\frac{Q}{RT}\right)$$

(2)

where $B$ is a material parameter. The SRS index, $m$, can thus be determined from:

$$m = \left(\frac{\partial \ln(\sigma_{imp})}{\partial \ln(V_{imp})}\right)_T$$

(3)

This equation implies that $m$-value can be obtained from the slope of the ln($\sigma_{imp}$) against ln($V_{imp}$) plots. The activation energy, $Q$, can be obtained from:

$$Q = -B \left(\frac{\partial \ln(V_{imp})}{\partial (1/T)}\right)_{\sigma,m}$$

(4)

Therefore, the activation energy can be obtained from the slope of a semi-logarithmic plot of $V_{imp}$ against $1/T$.

The steady-state or minimum impression rates were determined by differentiating the creep data with respect to time, using a five-point cubic spline numerical differentiation computer program. The logarithmic variation of punch stress with minimum impression velocity is plotted on a double-logarithmic scale at test temperatures of 523, 548, 573, and 598 K, as shown in Fig. 2a and b for the Ag-free and Ag-containing alloys, respectively. The SRS index, $m$, can be determined from the slope of the curves, according to Eq. (3). It is evident that the measured SRS indices and their variation with temperature show two different patterns for the Ag-free and Ag-containing alloys. It can be inferred from Fig. 2a that the rate-dependence of stress for the Ag-free material is well expressed by linear relationships having different slopes over the whole stress and temperature ranges studied. It is observed that the $m$-values increase monotonically with temperature from 0.13 at 523 K to 0.32 at 598 K. This behavior is in contrast to that of the Ag-containing alloy, shown in Fig. 2b. The punch stress-impact velocity curves show a sigmoidal shape with three regions I, II, and III, which become more distinct as the test temperature increases. In the intermediate strain rate region II, $m$-value showed the highest values, increasing from 0.30 at 523 K to 0.51 at 573 K and then decreasing to 0.46 at 598 K. This drop in $m$-value might have been caused by the slight grain coarsening that occurs at high temperatures. Lower $m$-values of about 0.25 are obtained in the lower and higher strain rate regions I and III. Similar $m$-values found for the fine-grained Sn–5Sb alloy have been attributed to the domination of other deformation mechanisms [17].

Our findings are comparable to the results of impression tests on a superplastic Zn–22Al alloy with the $m$-values increasing from...
0.27 to 0.51 as the temperature increased from 351 to 545 K [14]. Other similar results obtained in the localized shear punch testing of Mg–12Li–1Zn [18], Mg–9Gd–4Y–0.4Zr [11], and Sn–5Sb solder [25] are indicative of m-values of 0.44 at 548 K, 0.40 at 723 K, and 0.57 at room temperature, respectively. The superplasticity of an Ag-free Mg–9Gd–4Y–0.4Zr, studied by the conventional tensile tests, has shown that SRS indices as high as 0.54 can be obtained at 723 K [10]. It seems that the average grain size of 10 μm has increased the optimum superplastic temperature to 723 K, as opposed to our fine-grained material that shows the lower optimum temperature of 573 K. The values of activation energies can be deduced from the slope of the fitted lines in Fig. 3. For the Ag-free alloy, the activation energy changes from 152 to 272 kJ/mol, as the stress decreases (Fig. 3a). The activation energy of 181 kJ/mol obtained for the Ag-containing alloy in stage II of the sigmoidal curve is almost independent of stress, as shown in Fig. 3b. Higher activation energies of 216–460 kJ/mol, which are much greater than the activation energy for lattice diffusion (135 kJ/mol), have been reported for the superplastic deformation of a similar Mg–9Gd–4Y–0.4Zr alloy [10].

The microstructural features responsible for the observed differences between the behaviors of the two tested materials are shown in Fig. 4. It is clear that in both materials, recrystallization has occurred, though the grain sizes are significantly different. The microstructure of the base alloy, depicted in Fig. 4a, consists of many fine equiaxed grains less than 10 μm in size, and a few unrecrystallized grains larger than 10 μm, the average grain size being about 8 μm. The introduction of Ag into the base alloy has resulted in a fully recrystallized fine-grained microstructure with an average grain size of 3 μm, as shown in Fig. 4b. Another evident difference between the microstructures of the two tested alloys is the volume fraction of the second phase particles. It can be deduced that Ag addition results in a considerable increase in the volume fraction of intermetallic particles from 2.5 to about 13% that are mainly formed as a network in the matrix. Furthermore, the particles in the base alloy were identified as Mg5(Gd,Y), while in the Ag-containing alloy, this phase was replaced by the new Mg16Gd2YAg compound. It is, therefore, concluded that the higher density number of the second phase particles should have contributed to the achievement of the very fine recrystallized structure of the Ag-bearing alloy after hot deformation. These particles can effectively improve the dynamic recrystallization (DRX) process by pinning the grain boundaries during extrusion and hindering the growth of the recrystallized grains.

Grain boundary sliding (GBS) is known as the most widely accepted mechanism for superplastic deformation of materials in proper temperature and strain rate ranges [26]. In addition to the very fine grain structure, this mechanism necessitates high-angle grain boundaries. Fig. 5 exhibits the EBSD orientation maps and the corresponding distributions of grain boundary misorientation angles of both alloys. As can be seen in Fig. 5a, the base alloy contains some coarse un-DRX grains, which are surrounded by the much smaller equiaxed recrystallized grains. The results also indicate that the coarse un-DRX grains have the non-basal (1010) orientation. The Ag-containing alloy possesses a very uniform equiaxed recrystallized grain structure, as shown in Fig. 5c. According to the EBSD orientation map, the majority of the DRX grains are non-basal (2110) planes in this case. The respective distributions

![Fig. 5. The EBSD orientation maps and the corresponding distribution of grain misorientation angle of (a and b) Ag-free and (c and d) Ag-containing alloy.](image-url)
of grain boundary misorientation angles for the base and Ag-containing alloys are shown in Fig. 5b and d. It can be deduced that addition of Ag increases the percentage of HAGBs (θ ≥ 15°) from 73% to 88%. The obtained high fraction of HAGBs together with the very finer grain structure have resulted in the higher m-values and enhanced superplasticity in the Ag-bearing alloy.

As a support for the assessment of the deformation mechanism, a combination of strain rate sensitivity indices and activation energies can be considered [27]. Grain boundary sliding has been correlated to m-values of about 0.5 and activation energy values equal to that of grain boundary diffusion [28]. In the Ag-containing alloy the activation energy of 181 kJ/mol, which is nearly constant over the whole temperature range, is higher than the activation energy for grain boundary diffusion and even lattice diffusion (Q_a = 135 kJ/mol). Similar high values of superplastic activation energy for Mg–Gd–Y based alloys have been reported in the literature [11,12,29]. In the Ag-containing alloy, GBS accommodated by lattice diffusion is thus suggested as superplastic deformation mechanism. The abnormally high activation energy in Mg–Gd–Y alloys has been attributed to the low diffusion coefficients of Gd and Y elements in the Mg matrix [30]. The low diffusion coefficients would result in high values of activation energies according to the Arrhenius-type equation, which suggests an inverse relationship between the lattice self-diffusion coefficient (D_{sd}) and activation energy (Q) at constant temperature.

4. Conclusions

The effect of 2 wt% Ag addition on the microstructure and superplastic behavior of an extruded Mg–8.5Gd–2.5Y–0.5Zr (wt%) was studied. It was demonstrated that Ag refined the grain structure down to an average grain size of 3 μm and increased the fraction of high angle grain boundaries as well as the volume fraction of intermetallic particles. The occurrence of these events was reflected in the achievement of the high SRS index of about 0.5, in the intermediate region of sigmoidal stress-impression rate curves, which is typical of superplastic deformation behavior. This high m-value together with the activation energy of 181 kJ/mol, suggest that grain boundary sliding accommodated by lattice diffusion is the dominant superplastic deformation mechanism in the Ag-containing alloy. The Ag-free base material with a coarser grain size of 8 μm and a lower fraction of high-angle grain boundaries could not develop m-values higher than 0.32.

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References