Grain Boundary sliding mechanism during high temperature deformation of AZ31 Magnesium alloy

Peiman Shahbeigi Roodposhti^{*1,2}, Apu Sarkar¹, Korukonda Linga Murty¹, Harold Brody², Ronald Scattergood¹

¹North Carolina State University, ²University of Connecticut

Abstract

High temperature tensile creep tests were conducted on AZ31 Magnesium alloy at low stress range of 1 ~ 13 MPa to clarify the existence of grain boundary sliding (GBS) mechanism during creep deformation. Experimental data within the GBS regime shows the stress exponent is ~2 and the activation energy value is close to that for grain boundary diffusion. Analyses of the fracture surface of the sample revealed that the GBS provides many stress concentrated sites for diffusional cavities formation and leads to premature failure. Scanning electron microscopy images show the appearances of both ductile and brittle type fracture mechanism. X-ray diffraction line profile analysis (based on Williamson-Hall technique) shows a reduction in dislocation density due to dynamic recovery (DRV). A correlation between experimental data and Langdon's model for GBS was also demonstrated.

Keywords: AZ31, creep, grain boundary sliding, cavity formation

Introduction

Magnesium alloys due to their high specific strength and low density is considered as the next generation structural materials for a wide range of industries like automobile, aerospace and telecommunication [1]–[4]. They can even be replaced with some polymers due to their similar mechanical properties, more recyclability and cheaper prices [5], [6]. However like other hexagonal close packed (hcp) structure metals [7] they have relatively lower numbers of slip systems at room temperature, leading to their low ductility. At elevated temperatures, however, their prismatic and pyramidal slip systems can get activated and improve their ductility. It is well known that addition of some alloying element to Mg can improve its mechanical properties by forming a wide range of precipitations. Most of the precipitations have higher mechanical properties and can act as a barrier against grain boundary migration and diffusion. However some of them have low melting point and lose their mechanical properties at elevated temperature. Al and Zn are very common alloying elements which can be added to Mg alloys to make well-known Mg-Al-Zn alloys like AZ31. $Mg_{17}Al_{12}$ (β) is the predominant intermetallic phase in AZ31 Magnesium alloys. Because this alloy and the β phase have relatively low melting point (~600°C and ~450°C respectively [8]) creep is one of the main degradation mechanisms at engineering application. During GBS these particles can act as a stress concentration source and cause cavity formation and premature failure [9], [10].

* Corresponding author. Tel.: +1 919 757 7030, Fax.: +1 919 515 5115

Mail address: Mater. Sci. Eng. Department; 911 Partner Way, 3035 EB1, Raleigh, NC 27606-7907 USA

E-mail address: pshahbe@ncsu.edu

During last two decades several researches have been carried out to investigate the high temperature behavior of these alloys to characterize their creep mechanisms and fracture morphologies [2], [9]–[24]. At elevated temperature GBS can occurs via well-known Rachinger [25] or Lifshitz [26] mechanisms. Lifshitz (1963) explained grain boundary sliding via diffusion of the voids through grain boundary or lattice which leads to grains elongation in the tensile direction. While Rachinger (1952) provided a model based on grain displacement without any changes in the grains shape. In Lifshitz mechanism the number of surface grains remain constant, whereas they increases via Rachinger mechanism [11], [27], [28]. Due to grain deformation during Lifshitz creep mechanism, even large grain size samples can keep their coherencies during creep deformation and lead to a large plastic deformation. Whereas Rachinger mechanism can lead to high plastic deformation only for small grain size (~ around a few microns). In samples with large grain size the Rachinger sliding can cause cavity formation leading to premature failure. Both Lifshitz and Rachinger sliding can make similar offsets in marker lines [28].

During high temperature deformation of Mg alloys the minimum [28] strain rate can be related to the stress, temperature and grain size via well-known Bird, Mukherjee and Dorn power-law equation

$$\dot{\epsilon} = \frac{ADGb}{kT} \left(\frac{b}{d}\right)^p \left(\frac{\sigma}{G}\right)^n \quad (1)$$

where A is a material constant, G is the shear modulus, n is the stress exponent, p is the inverse grain size exponent, D is diffusion coefficient, k is the Boltzmann's constant, b is the burgers vector, d is the grain size and T is the absolute testing temperature. The diffusion coefficient can be related to activation energy via $D=D_0 \exp(-Q/RT)$ where the Q is activation energy for specific creep mechanism, D_0 is a pre-exponential term and R is the gas constant.

In this study The AZ31 Mg alloy has been deformed at three different temperatures of 230°C, 270°C and 350°C and constant stresses of 1 to 13 MPa. The role of precipitates in creep mechanism and their stability at various temperatures have been analyzed. The values of n, p & Q in equation (1) along with microstructural characterization provide information on the dominant creep mechanism. The experimental data have been compared with previous models to correlate them with conventional models. Our previous works show that dynamic recovery (DRV) and slightly decrease in dislocation density is characteristic of this region [9], [10].

Experimental Procedure

A rod form of AZ31 Mg alloys with chemical composition of Mg-2.8Al-1.1Zn (in wt. %) has been used in this study. The primary dimensions of that were $1" \times 1" \times 12"$. Standard dog bone shape tensile specimens with gage length of 25 mm were extracted from as-received rods. To provide homogeneous microstructure and mechanical properties in all directions, samples were annealed at 450°C for two hours at protective atmosphere of argon gas then furnace cooled to room temperature. Optical microscopy analysis along with three dimensions microhardness tests prove their homogeneity in all directions. Tensile deformations at constant stresses of 1~13 MPa and three different temperatures of

230°C, 270°C and 350°C were conducted. The tensile axis was along the rolling direction. An extensometer and a Linear Variable Differential Transducer (LVDT) were installed in the experimental setup to measure the axial strain. Elevated temperatures were applied to the sample by a three zone split furnace. Because the experimental temperatures were below the oxidation temperature of AZ31 Mg alloy, tests have been done in unprotected atmosphere. Further characterizations also showed no significant amount of Oxide in the surface. Before each test, samples were held for 30 minutes to equilibrate at the testing temperature. Creep tests were continued to reach to the minimum strain rate then the specimens were cooled under load to room temperature to preserve the steady state creep characteristics. The well-known equations of $\epsilon = \ln(1 + \Delta L / L_0)$ and $\sigma = (F / A_0)(1 + \Delta L / L_0)$ can relate the total displacement of the extensometer (ΔL) and the initial dimensions of sample (A_0 and L_0) to the true strain (ϵ) and true stress (σ). To reveal grain size distribution the specimens were polished with 600 SiC abrasive papers followed by 9 μm , 3 μm and 1 μm glycol-based polycrystalline diamond suspensions and non-woven textile polishing clothes. Final polishing was done by 0.04 μm colloidal silica suspension on napless polyurethane cloth. A solution of 5ml Picric acid, 10ml acetic acid, 10ml H₂O and 70 ml ethanol were used to reveal the microstructure. To show the grains sliding over each other, a Focused Ion Beam (FIB) machine was used to draw some line pattern on the fully polished samples. Lines with dimensions of 600 $\times$ 0.1 $\times$ 0.3 μm^3 were drawn using Ga ion beam accelerated at 30 kV and 1 nA. Then the samples were creep tested to reach to the minimum strain rate at different stresses. Crept sample polished with 0.04 μm colloidal silica suspensions and etched to see grain sliding and possible line offsets. A Hitachi-3200 Scanning Electron Microscope (SEM) were used to reveal some precipitates in the matrix. To find the dislocation density of the samples based on Williamson-Hall technique, XRD analyses on the fresh and deformed samples were carried out using the analytical empyrean X-Ray diffractometer equipped with advanced PIXcel 1D detector. The thermal stability of the precipitations were evaluated via high temperature XRD analysis. Samples were placed inside a furnace and heated from room temperature to 450°C at a heating rate of 5 °C/min.

Results and discussion

Solute atoms in popular Mg-Al alloys, like AZ31, can suppress grain boundary migration. These atoms can make some precipitates in the matrix. The most popular intermetallic phase which can make a eutectic in the Mg-Al phase diagram is the Mg₁₇Al₁₂ known as β phase. Figure 1(a) shows optical micrograph of the precipitations dispersed randomly in the matrix. An analysis using SEM equipped with X-Ray energy dispersive spectroscopy (EDS) reveals the existence of Mg₁₇Al₁₂ (Figure 1(b)). Figure 1(c-d) show XRD plots for AZ31 Mg alloys before creep test. Along with some peaks for matrix (Mg), around 2 θ of 37° a peak for Mg₁₇Al₁₂ were indexed. Figure 2 shows the optical micrograph with an average linear intercept grain size (ALIGS) of ~22 μm and three dimensions microhardness test results. The analyses revealed that the grains are equiaxed and mechanically homogeneous.

Mg-Al phase diagram shows that at elevated temperatures β phase dissolves in the matrix. Figure 3 shows a study on the high temperature XRD patterns of up to 450°C carried out at 50°C intervals. It is clear that the intensity of the peak of precipitates is a function of temperature. By increasing the

temperature the relative intensity decreases which is related to the dissolving of precipitates in the matrix.

(a)

(b)

(c)

(d)

Figure 1 appearance of the β phase in the matrix (a) Optical Microscope (b) SEM – EDS (c) XRD $30 < 2\theta < 100$ (d) $30 < 2\theta < 40$

(a)

(b)

(c)

Figure 2 Microstructure of AZ31 after 2hr annealing at 450 °C (a) grain size distribution (b) and microhardness results in three dimensions (c).

Figure 3 High temperature XRD pattern of AZ31 Mg alloy

Figure 4a shows the creep curve conducted at 350°C and 1 MPa. Most of the tests continued to reach to the steady state region. Then the stresses were increased to a higher value. However, stress change experiments did not lead to remarkable transients, so that it reaches to the steady state condition very quickly. Experiments show that creep rates of single stress tests are similar to stress change test. Figure 4b shows the strain rate vs. time plot for test conducted at 350°C and 1 MPa. It is clear that the strain rate continuously decreases to reach to a minimum value. Steady state or minimum creep rate values have been determined for different deformation conditions. A double log plot of steady state (minimum) creep-rate vs. normalized stress (σ/E , ($E=2.6(1.92(10^4-8.6T))$ in MPa (T in K)) [2]) illustrated in Figure 5(a). Straight line fits to the data show the trend of increasing minimum strain rate with stress for all temperatures. A plot of minimum strain rate vs inverse of temperature shows the temperature dependency of creep deformation. The slop of this curve is related to the activation energy, which is around 87 kJ/mole and is very close to the activation energy of the diffusion along the grain boundary of Mg alloys. Figure 5(a) can be converted to temperature compensated creep curve using the calculated activation energy in equation 1. Results illustrated in Figure 5(c).

Figure 4 (a) creep curve at 350°C, 1 MPa and (b) strain rate vs time

Figure 5 (a) Double log plot of minimum strain rate vs. stress at T=230°C, 270°C and 350°C, (b) Corresponding activation energy and (c) temperature compensated creep-rate versus normalized stress.

Fitting the data in Figure 5c with the equation (1) yields the stress exponent of $n \sim 2$. Previous investigations show that this stress exponent along with an activation energy close the grain boundary diffusion is indicative of grain boundary sliding mechanism during creep deformation. Equation (2) shows a model developed by Langdon for GBS creep mechanism. Although the value for n and p is different for small and large grain size, because of the intermediate grain size of the material of this study, they have characteristics of both large and small grain size. Figure 6 shows the correlation between experimental data with the model:

$$\dot{\epsilon} = \frac{AD_{gb}Gb}{kT} \left(\frac{b}{d}\right)^2 \left(\frac{\sigma}{G}\right)^2 \quad (2)$$

Where D_{gb} is grain boundary diffusion and A is a constant ~ 10 . [11]

Figure 6 Comparison of the present creep data and the model

As far as grains geometrics are concerns, their deformation and relative movement over each other are two most conventional plastic deformations mechanisms of polycrystalline materials at elevated temperatures and low strain rates. Offset in the marker lines shows grains geometry after GBS. A deformation on the line in the grains along with line offset between grains could be sign of Lifshitz sliding while lines in Rachinger mechanism are fully straight in the grains and just have some offset when they pass adjacent grains. Figure 7(a) shows the line patterns placed on polished starting sample by Focused Ion Beam (FIB) machine. Then the samples were crept at 350°C, 3 MPa . Figure 7(b) shows the microstructure of the surface of the deformed sample. The tensile axis is perpendicular to the line patterns. It is clear that the original lines are shifted in some grain boundaries while they are straight within the grains. This can be related to the Rachinger GBS mechanism. Because the amount of shifting is related to the grain boundaries structure, the displacements are not appearing in all grain boundaries.

Figure 7 Focused ion beam (FIB) images of starting sample, (b) SEM microstructure of crept sample at 350°C and 3 MPa showing offset line (c) higher magnification

In relatively large grain size materials, Rachinger GBS can make some cavities and lead to premature failure [28]. Microstructures of crept samples reveals the appearance of a large number of cavities (Figure 8a). Cavity formation, like other nucleation and growth process, is easier in heterogeneous sites. Grain boundaries, triple points and precipitates are nucleation sites for cavity formation in AZ31 Mg alloy. During GBS, the stress concentration in heterogeneous sites reaches to a critical value. If the void establishment is faster than stress relaxation, diffusional cavities form. Our previous study shows that this alloy is very susceptible to cavity formation in precipitates and grain boundaries [9] (Figure 8b). The fracture mechanism is a combination of ductile and brittle type fracture. Figure 9 shows the fracture surface of crept samples in the GBS region. The fractographs show the existence of cleavage line along with serpentine sliding. The former is related to Trans-granular fracture due to void coalescence and cavity formation and the latter indicates the appearance of dimple shape fracture.

(a)

(b)

Figure 8 cavity formation (a) in the precipitate, crept at 350°C, 4 MPa (b) in the free surface close to fracture surface crept at 350°C, 9 MPa

(a)

(b)

(c)

Figure 9 Fractured surface of the crept sample at (a) 270°C, 7 MPa (b, c) 350°C, 4 MPa.

Although Mg alloys, due to their low stacking fault energy and high grain boundary diffusion velocity, are very susceptible to dynamic recrystallization at high stress thermoplastic deformation [29]–[34], our previous study shows that in low stress regions dynamic recovery (DRV) is dominant mechanism [10]. This is mainly due to the lack of energy to complete recrystallization. Dislocation density (ρ) calculation based on Williamson-Hall (WH) technique shows a reduction of ρ during GBS mechanism. This is mainly because of the annihilation of dislocations over each other due to DRV at low stress and high temperatures [10].

Conclusion:

Grain boundary sliding mechanism of AZ31 Mg alloy during high temperature, low stress creep was investigated. Experimental data revealed a stress exponent of $n \sim 2$, inverse grain size exponent of $p \sim 2$ and an activation energy value of ~ 87 kJ/mol which is close to that for grain boundary diffusion. Line

patterns developed by Ga ion beam spattering show offsets in some grain boundaries and no deformation through the grains which is characteristics of Rachinger GBS mechanism. The experimental data demonstrated a good correlation with Langdon's GBS model. SEM images show that the fracture surfaces have characteristics of both ductile and brittle type fractures. Cavity formation and growth is responsible for premature failure of AZ31 alloys during GBS mechanism.

Acknowledgement:

This research is supported by the National Science Foundation grant 0968825 and Department of Energy grant DE-NE-0000538.

References

- [1] a. Bussiba, a. Ben Artzy, a. Shtechman, S. Ifergan, and M. Kupiec, "Grain refinement of AZ31 and ZK60 Mg alloys — towards superplasticity studies," *Mater. Sci. Eng. A*, vol. 302, no. 1, pp. 56–62, Apr. 2001.
- [2] S. W. Chung, H. Watanabe, W.-J. Kim, and K. Higashi, "Creep Deformation Mechanisms in Coarse-Grained Solid Solution Mg Alloys," *Mater. Trans.*, vol. 45, no. 4, pp. 1266–1271, 2004.
- [3] B. L. Mordike and T. Ebert, "Magnesium Properties — applications — potential," *Mater. Sci. Eng. A*, vol. 302, pp. 37–45, 2001.
- [4] W.-J. Kim, S. . Chung, C. . Chung, and D. Kum, "Superplasticity in thin magnesium alloy sheets and deformation mechanism maps for magnesium alloys at elevated temperatures," *Acta Mater.*, vol. 49, no. 16, pp. 3337–3345, Sep. 2001.
- [5] N. Farahbakhsh, P. ShahbeigiRoodposhti, A. S. Ayoub, R. A. Venditti, and J. S. Jur, "Melt Extrusion of Polyethylene Nanocomposites Reinforced with Nanofibrillated Cellulose from Cotton and Wood Sources," *J. Appl. Polym. Sci.*, vol. 132, no. 17, p. 10, 2014.
- [6] S. Ziaei and M. a. Zikry, "Modeling the Effects of Dislocation–Density Interaction, Generation, and Recovery on the Behavior of H.C.P. Materials," *Metall. Mater. Trans. A*, Nov. 2014.
- [7] P. Shahbeig Roodposhti, N. Farahbakhsh, A. Sarkar, and K. L. Murty, "A microstructural approach to the Equal Channel Angular Processing of Commercially Pure Titanium – a review," *Trans. Nonferrous Met. Soc. China*, vol. 25, pp. 1353–1366, 2015.

- [8] P. Shahbeigi Roodposhti, A. Sarkar, and K. L. Murty, "A review of the influence of production methods and intermetallic phase on the creep properties of AZ91," *Magnes. Technol.*, pp. 59–64, 2014.
- [9] P. Shahbeigi Roodposhti, A. Sarkar, and K. L. Murty, "Fracture behavior of AZ31 Magnesium alloy during low stress high temperature deformation," *Metallogr. Microstruct. Anal.*, vol. 4, pp. 91–101, 2015.
- [10] P. Shahbeigi Roodposhti, A. Sarkar, and K. L. Murty, "Microstructure development of high temperature deformed AZ31 magnesium alloys," *Mater. Sci. Eng. A*, vol. 626, pp. 195–202, 2015.
- [11] R. Korla and A. H. Chokshi, "A Constitutive Equation for Grain Boundary Sliding: An Experimental Approach," *Metall. Mater. Trans. A*, vol. 45, no. 2, pp. 698–708, Oct. 2013.
- [12] R. B. Figueiredo and T. G. Langdon, "Developing superplasticity in a magnesium AZ31 alloy by ECAP," *J. Mater. Sci.*, vol. 43, no. 23–24, pp. 7366–7371, Jul. 2008.
- [13] S. Spigarelli, M. El Mehtedi, D. Ciccarelli, and M. Regev, "Effect of grain size on high temperature deformation of AZ31 alloy," *Mater. Sci. Eng. A*, vol. 528, no. 22–23, pp. 6919–6926, Aug. 2011.
- [14] S. Spigarelli, M. El Mehtedi, M. Cabibbo, E. Evangelista, J. Kaneko, A. Jäger, and V. Gartnerova, "Analysis of high-temperature deformation and microstructure of an AZ31 magnesium alloy," *Mater. Sci. Eng. A*, vol. 462, no. 1–2, pp. 197–201, Jul. 2007.
- [15] H. Somekawa, K. Hirai, H. Watanabe, Y. Takigawa, and K. Higashi, "Dislocation creep behavior in Mg–Al–Zn alloys," *Mater. Sci. Eng. A*, vol. 407, no. 1–2, pp. 53–61, Oct. 2005.
- [16] K. Ishikawa, H. Watanabe, and T. Mukai, "High temperature compressive properties over a wide range of strain rates in an AZ31 magnesium alloy," *J. Mater. Sci.*, vol. 40, no. 7, pp. 1577–1582, Apr. 2005.
- [17] A. G. Beer and M. R. Barnett, "Influence of initial microstructure on the hot working flow stress of Mg–3Al–1Zn," *Mater. Sci. Eng. A*, vol. 423, no. 1–2, pp. 292–299, May 2006.
- [18] S.-H. Choi, J. K. Kim, B. J. Kim, and Y. B. Park, "The effect of grain size distribution on the shape of flow stress curves of Mg–3Al–1Zn under uniaxial compression," *Mater. Sci. Eng. A*, vol. 488, no. 1–2, pp. 458–467, Aug. 2008.
- [19] H.-K. Kim and W.-J. Kim, "Creep behavior of AZ31 magnesium alloy in low temperature range between 423 K and 473 K," *J. Mater. Sci.*, vol. 42, no. 15, pp. 6171–6176, Apr. 2007.

- [20] J. A. D. E. L. Valle and O. A. Ruano, "Deformation Mechanisms Responsible for the High Ductility in a Mg AZ31 Alloy Analyzed by Electron Backscattered Diffraction," *Metall. Mater. Trans. A*, vol. 36, no. June, pp. 1427–1438, 2005.
- [21] S. W. Chung, C. S. Chung, and D. Kum, "Super plasticity in thin Magnesium alloy sheets and deformation mechanism maps for magnesium.," *Acta Mater.*, vol. 49, pp. 3337–3345, 2001.
- [22] K. Kitazono, E. Sato, and K. Kuribayashi, "Internal stress superplasticity in polycrystalline AZ31 magnesium alloy," *Scr. Mater.*, vol. 44, no. 12, pp. 2695–2702, Jun. 2001.
- [23] H. Somekawa and T. Mukai, "Molecular dynamics simulation of grain boundary plasticity in magnesium and solid-solution magnesium alloys," *Comput. Mater. Sci.*, vol. 77, pp. 424–429, Sep. 2013.
- [24] P. Shahbeigi Roodposhti, A. Sarkar, and K. L. Murty, "Creep deformation mechanisms and related microstructure development of AZ31 Magnesium alloy," *Magnes. Technol.*, pp. 29–34, 2015.
- [25] W.A. Rachinger, "relative grain translations in the plastic flow of aluminium.pdf." p. 33.
- [26] I.M. Lifshitz, "On the theory of diffusion-viscous flow of polycrystalline bodies," *Sov. Phys. JETP*, vol. 17, pp. 909–20, 1963.
- [27] W.R. Cannon, "The contribution of grain boundary sliding to axial strain during diffusion creep," *Philos. Mag*, vol. 25, pp. 1489–97, 1972.
- [28] T. G. Langdon, "A unified approach to grain boundary sliding in creep and superplasticity," *Acta Metall.*, vol. 42, no. 7, pp. 2437 – 2443, 1994.
- [29] M. Qian and Z. . Guo, "Cellular automata simulation of microstructural evolution during dynamic recrystallization of an HY-100 steel," *Mater. Sci. Eng. A*, vol. 365, no. 1–2, pp. 180–185, Jan. 2004.
- [30] S. Gourdet and F. Montheillet, "A model of continuous dynamic recrystallization," *Acta Mater.*, vol. 51, no. 9, pp. 2685–2699, May 2003.
- [31] S. Gourdet and F. Montheillet, "Effects of dynamic grain boundary migration during the hot compression of high stacking fault energy metals," *Acta Mater.*, vol. 50, no. 11, pp. 2801–2812, Jun. 2002.

- [32] N. Xiao, C. Zheng, D. Li, and Y. Li, "A simulation of dynamic recrystallization by coupling a cellular automaton method with a topology deformation technique," *Comput. Mater. Sci.*, vol. 41, no. 3, pp. 366–374, Jan. 2008.
- [33] G. Kugler and R. Turk, "Modeling the dynamic recrystallization under multi-stage hot deformation," *Acta Mater.*, vol. 52, no. 15, pp. 4659–4668, Sep. 2004.
- [34] X. LIU, L. LI, F. HE, J. ZHOU, B. ZHU, and L. ZHANG, "Simulation on dynamic recrystallization behavior of AZ31 magnesium alloy using cellular automaton method coupling Laasraoui–Jonas model," *Trans. Nonferrous Met. Soc. China*, vol. 23, no. 9, pp. 2692–2699, Sep. 2013.