Magnesium Technology 2013
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Wrought Materials III

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EFFECT OF PRECIPITATION ON DYNAMIC RECRYSTALLIZED GRAIN SIZE IN A MAGNESIUM ALLOY

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Abstract

Magnesium, the lightest structural metal has specific tensile strength and rigidity superior to iron and aluminum showing a rising demand for use in the automotive industries to lighten the structure. But, magnesium and its alloys have limited ductility and poor formability at room temperature due to an insufficient number of operative slip and twinning systems associated with its hexagonal close packed structure. A fine grain structure and random texture can improve the ductility or formability. The aim of this work is to investigate the effect of precipitates on microstructure evolution during hot deformation of Mg-Al-Sn alloy. Mg-Al-Sn alloy has been designed using thermodynamic modeling software, FactSage, based on forming precipitates at deformation temperature of 300 °C. The formation of precipitates, mostly Mg_2Sn, during the dynamic recrystallization process may slow down the grain growth resulting in finer grain size and grain size homogenization.

Introduction

Magnesium is the highest specific strength structural metal and is the most potential candidate for the automotive industry to reduce the weight of the vehicle. It is 36 % lighter than aluminum and 78 % lighter than steel [1]. In comparison to aluminum and steel, very limited wrought magnesium alloys are commercially available [2]. One big reason behind this is, the low formability of Mg alloys at lower temperature (<200 °C) due to its hexagonal close pack (HCP) structure [3] and a preferred crystallographic orientation (texture) that develops in wrought alloys [4, 5]. This leads to a fabrication process with high temperature to increase the formability resulting in, increase in price of magnesium alloy products. Numerous investigations are ongoing in searching for suitable Mg alloy systems with superior properties.

It is well established that mechanical properties of polycrystalline metals at room temperature can be improved by grain refinement according to the Hall-Petch relationship [6]. Also, it has been reported that, the Hall-Petch relationship is four times more significant in magnesium mechanical properties than in aluminum, indicating the importance of grain refinement in magnesium alloys [7]. Grain refinement in hot worked magnesium alloys are usually done through a recrystallization process. Recrystallization is a restoration and usually thermally activated process by which relatively strain free grains are formed within the deformed structure consuming the deformed grains, resulting in, refined grain size [8]. Recrystallization during the hot deformation process is termed as dynamic recrystallization (DRX). Dynamic recrystallization is attributed to magnesium alloy because of its low stacking fault energy (~ 78 mJ m^{-2}) and limited slip systems when a critical deformation condition is reached [9]. DRX generally starts at the parent or old grain boundaries. New grains are subsequently nucleated at the old grain boundaries and make a "necklace structure" by the band of recrystallized grains and eventually the material becomes fully recrystallized. When the time (strain rate) and temperature is not sufficient enough for the complete recrystallization process, partial dynamic recrystallization is observed [10].

Precipitation strengthening is defined by the precipitation of a second phase, which is different in composition than the matrix [10]. In Mg–Sn binary phase diagram, the maximum solid solubility of tin in magnesium is relatively high with 14.85 wt% at the eutectic transformation temperature of 561 °C, which drops off sharply with the temperature. At 200 °C the solubility limit is 0.45 wt% and almost zero at room temperature [11]. This fast decrease in solubility with decrease in temperature makes tin suitable for precipitation in the form of Mg_2Sn. The Mg_2Sn phase precipitates mainly at grain boundaries and may take part in the strengthening effect (increase of the flow stress) by restraining the dislocation movement [12]. More uniform and fine precipitates are formed with plastic deformation, which can generate high energy sites like dislocations, twins and deformation bands. These defects accelerate the diffusion of solute atoms enhancing the subsequent aging kinetics and are the preferential location for the solute atoms to gather due to the minimum of energy needed [13].

Jonas and Weiss [14] reported for micro-alloyed steel that strain induced precipitates prevents the further motion of grain boundary by pinning effect and retards the dynamic recrystallization resulting in a fine grain structure. They also mentioned that the size of the precipitate is an important factor. Smaller precipitates create stronger retardation to dynamic recrystallization. Retardation continues as far as the precipitates size is no larger than the recrystallized nuclei. In general, for the hot working process, the dynamic recrystallization grain size depends on the deformation amount (strain), strain rate, and deformation temperature [15].

In AZ31, the leading alloy commercially available as wrought products, the precipitate forms at lower temperature (~200 °C) than the industrial deformation temperature range of 250-400 °C. So, there is no or little effect of second phase particles on the recrystallized grain size as they are mostly dissolved in the matrix during high temperature deformation [16]. But it is possible to design alloys where high temperature stable precipitates such as Mg_2Sn forms during the hot deformation temperature range of 250-350 °C. It is expected that this stable precipitates will retard the dynamic recrystallization process by the grain boundary pinning [17] or particle simulated nucleation

(PSN) process [10] resulting in reduced recrystallized grain size, which literally means the increase of ductility and strength at room temperature.

The goal of this research is to design alloy where it is possible to form precipitates during high temperature deformation and to investigate the possibility of recrystallized grain size reduction and homogenization by the pinning effect of the precipitates at the grain boundary.

Experimental Procedures

Thermodynamics software FactSage [18] was used to design a candidate Mg alloy with the alloying elements Al and Sn. Tin has been added in the magnesium-aluminum alloy to form thermally stable Mg_2Sn precipitate at deformation temperature. The alloy was then produced with commercially pure Mg, Al and Sn melted using an induction furnace (20 kW, 5 kHz) in a graphite crucible and cast in a copper mold preheated to 250 °C. The alloy addition and casting were done at 690 °C and 730 °C, respectively in a protective environment created by the mixture of CO_2 (99.5 %) and SF_6 (0.5 %) gases. The cast ingots were homogenized for 24 hours at 440 °C to fully dissolve the precipitates. The temperature for homogenization was chosen using the FactSage generated equilibrium diagrams.

Compression samples of 11.4 mm in height and 7.6 mm in diameter (h/d ratio of 1.5) were machined from the homogenized bar. The uniaxial compression was performed using a computer controlled servo-hydraulic materials testing system (MTS) with a 100 kN capacity, coupled with a radiant furnace. Isothermal compression testing was performed at 300 °C with strain rates of 1.0, 0.1, 0.01, and $0.001 s^{-1}$. In this study the deformation amount was fixed to 90 % (strain of 0.9). The specimens were held at the deformation temperature for 10 minutes prior to deformation to ensure a uniform distribution of temperature throughout the sample and water quenched immediately after the deformation to preserve the dynamically formed microstructure.

Specimens for metallurgical examination were cut along the compression axis. Polished specimens were etched in a fresh solution of 10 ml acetic acid, 4.2 gm picric acid, 10 ml water and 70 ml alcohol for 3-8 seconds. Microstructural examination was carried out using Nikon Epiphot 200 optical microscope and Clemex image analysis software, with grain size measured by linear intercept method. A Philips XL30 FEG SEM with energy dispersive X-ray spectroscopy (EDS) was used to examine the microstructure and to characterize the second phase particles. Microindentation hardness was measured by Clark CM-100AT machine with a load of 50 gram and a dwell time of 10 seconds.

Results and Discussion

Mg-3 wt% Al-3 wt% Sn (AT33) alloy has been designed using thermodynamic modeling software FactSage in the target of forming precipitate at the deformation temperature of 300 °C. Figure 1 shows the equilibrium phase fraction obtained from FactSage indicating the types and amount of precipitates formed during the equilibrium cooling. Second phase particles Mg_2Sn and $Mg_{17}Al_{12}$ are starting to precipitate out from the Mg matrix just below 380 °C and 210 °C, respectively. Certain amount (80 °C in this study) of undercooling was well thought-out in the alloy design to generate the necessary driving force for the nucleation of second phase particle Mg_2Sn.

Figure 2-a shows the as-cast optical microstructures of the alloy. The microstructure consists of primary α-Mg, and secondary particles between dendrite arms. The average grain size was measured as ~120 μm. The precipitates are coarse having the size around 2-5 μm. EDS analysis confirms that the precipitates are mainly composed of Mg, Al, and Sn.

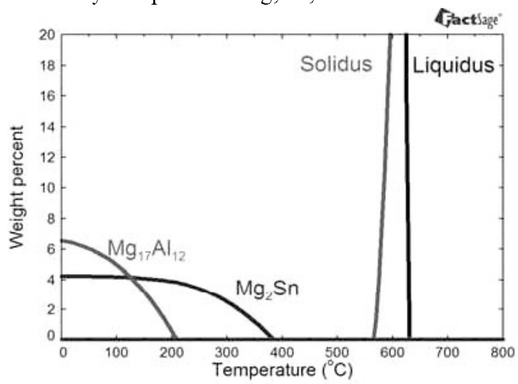
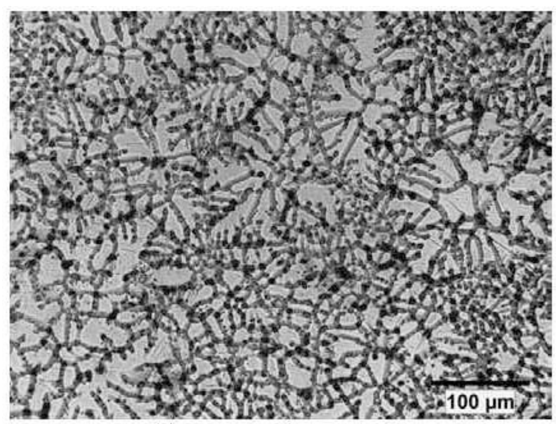
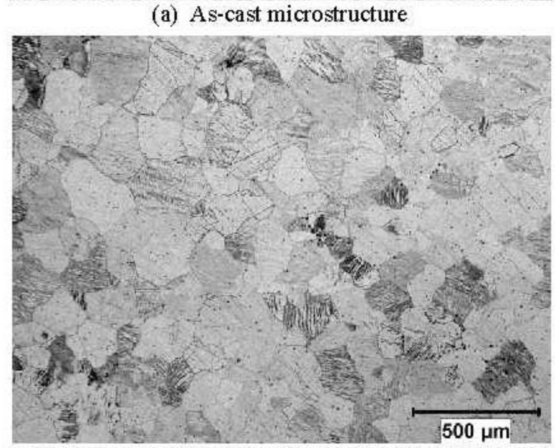


Figure 1: Equilibrium phase fraction of Mg-3Al-3Sn alloy predicted by FactSage.

No precipitate has been found after homogenization for 24 h at 440 °C, as shown by the microstructure in Figure 2-b. The average grain size of the homogenized sample displayed almost no grain growth during the homogenization treatment.



(a) As-cast microstructure



(b) Homogenized at 440 °C for 24 hours to dissolve precipitates

Figure 2: As cast and homogenized microstructures for alloy AT33

Figure 3 shows the variation of flow behavior at 300 °C for different strain rates. The typical true stress and true strain curve obtained at a strain of 0.9 exhibits work hardening, a single peak at low strain (0.1-0.3), after which the flow stresses decreases consistently followed by flow softening. This softening behavior is attributed to dynamic recrystallization phenomena [10]. The effect of strain-rate on the flow behaviour is significant and is shown in figure 4. Peak stress, flow stress, and yield stress (0.2% proof) decrease with decrease in strain rate. This is reasonable with regard to the time dependence of DRX. The decrease in flow stress with decreasing strain rate is due to the increasing ease of dynamic recrystallization and in part to the activation of additional slip systems. Lower strain rates provide more time for boundary migration and accelerate the nucleation and growth processes of dynamic recrystallization [19].

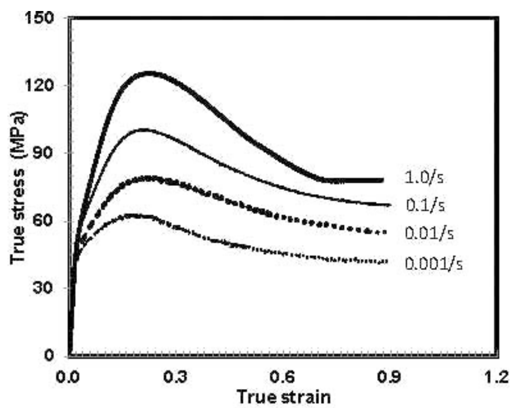


Figure 3: Flow behavior at 300 °C for different strain rates

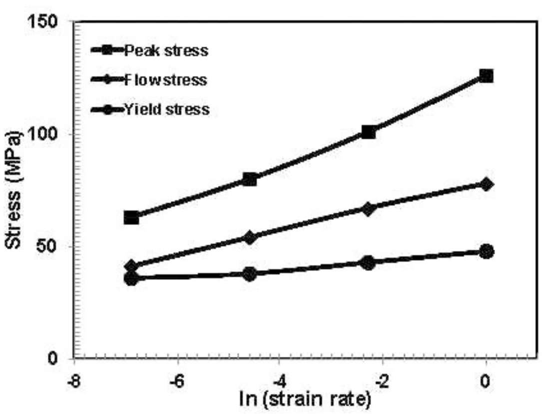


Figure 4: Effect of strain rate on peak stress, flow stress, and yield stress

Figure 5 shows the microstructures of the alloy deformed at 300 °C and a strain of 0.9 subjected to various strain rates. A necklace type structure was observed with recrystallized grains distributed along pre-existing boundaries. The microstructures are characterized by dynamic recrystallization and grains are significantly refined. The recrystallized grains evolved dynamically as the static recrystallization was prevented by quenching the samples immediately after deformation.

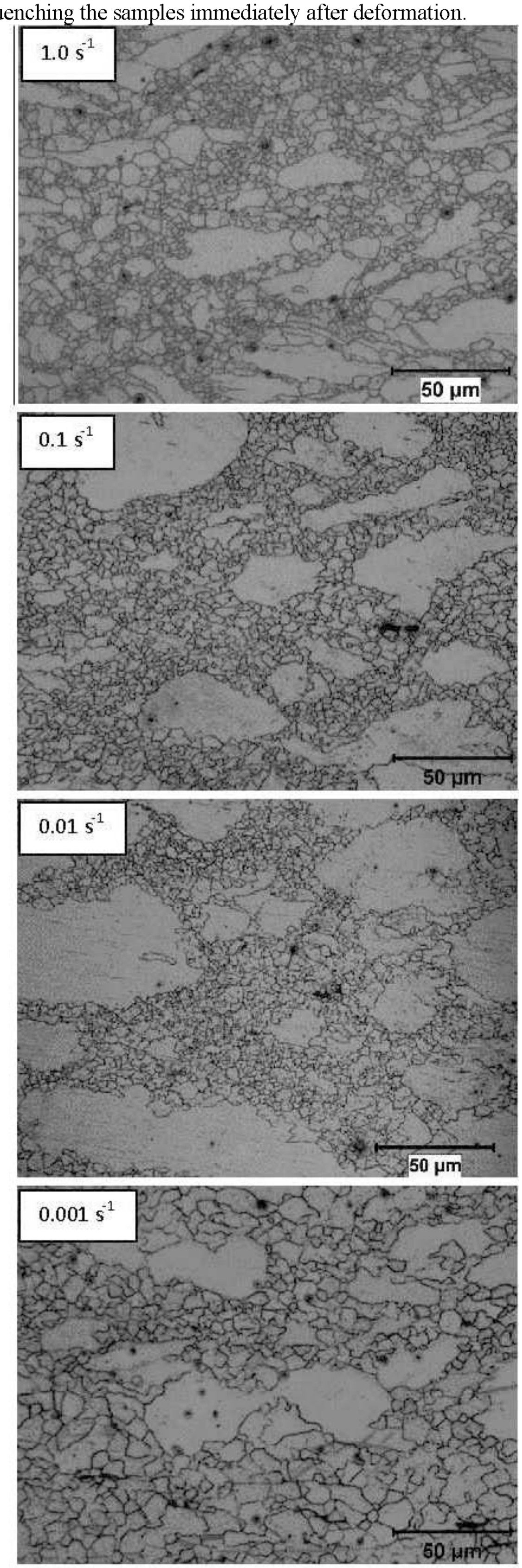


Figure 5: Deformed microstructure for different strain rates

Figure 6 shows the effect of strain rate on DRX grain size. The DRX grain size dependency on strain rate is related to the available time, with a lower strain rate (0.001s^{-1}) equating to a longer time of deformation and more grain growth during the deformation. The time difference between the maximum and minimum strain rate is 15 minutes for a strain of 0.9 in this study. DRX grain size increases about 2 µm in total for the lowest strain rate. This increase can be reasoned by the time availability for grain growth in lowest strain rate as well as the size of the precipitates which is discussed in the next section.

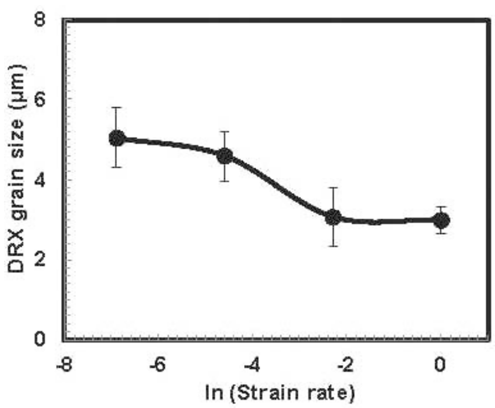


Figure 6: Effect of strain rates on DRX grain size at 300 °C

As mentioned earlier, the samples were held 10 minutes at the deformation temperature prior to deformation to achieve the uniform temperature throughout the whole sample. Quenching from this stage confirms the presence of Mg_2Sn precipitates in the parent grain boundaries. These precipitates are coarser compared to those forms during deformation.

Figure 7 shows the distribution of precipitates at different strain rates in the deformed alloy. Strain induced precipitates of 70-300 nm in size are observed mainly in the DRXed areas along or close to the DRX grain boundaries. It is distinguishable from the figure that both the size and the volume fraction of the precipitate increase with decreasing strain rate. For a strain rate of 1.0 s^{-1} (deformation time 0.9 sec), only the prior deformation precipitates in the parent grain boundaries are visible at lower magnification. But at higher magnification, very fine dispersion of precipitates (~70 nm) is visible close to those parent grain boundaries. Decrease in strain rate increases the time for more precipitates to nucleate and the existing one to grow. Thus, at the strain rate of 0.001 s^{-1}, where the total deformation time is 15 minutes, more volume fraction of precipitates are visible as large as ~300 nm in size. The fine dispersion of precipitates may restrain grain growth by pinning the boundaries and dislocations. The increasing size of the precipitates for low strain rates may not be as effective as the fine precipitates are, for the pinning effect, resulting in the increases of DRX grain size as shown in figure 6.

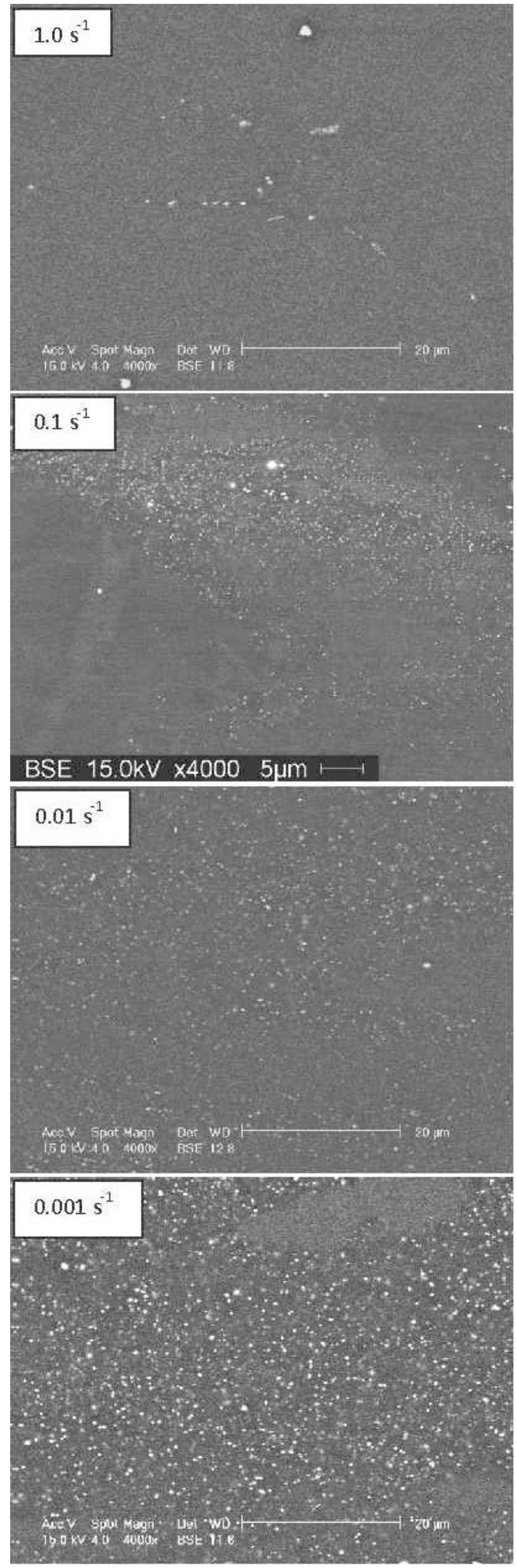


Figure 7: Distribution of Mg_2Sn particles with strain rates

The EDS study (Figure 8) confirms that the precipitates are mostly composed of Mg and Sn, which is consistent with the

Mg_2Sn precipitate predicted by the thermodynamic calculation results from FactSage at 300 °C. To study the room temperature strength, micro-indentation hardness experiment was performed. Figure 9 shows the effect of strain rate on the micro-indentation hardness at different strain rates. Hardness increases with decreasing the strain rate. Dotted line in the figure indicates the hardness of the homogenised sample. The increase in hardness in deformed samples than the homogenized one is reasoned by the presence of Mg_2Sn precipitates. Mg_2Sn is a hard (~119 HV) and brittle phase which contributes to strengthen the matrix [12]. The size and volume fraction of the Mg_2Sn phase increases with decreasing strain rate. This explains the continuous increase in micro-hardness of the alloy with decreasing strain rate.

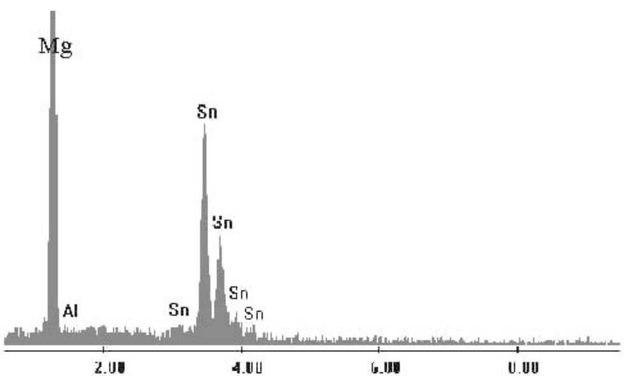


Figure 8: EDS qualitative analysis of the precipitate

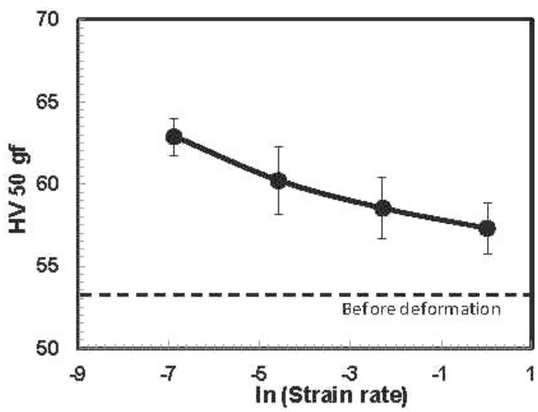


Figure 9: Effect of strain rate on micro-indentation hardness

Summary

The aim of this work was to investigate the grain boundary pinning effect by precipitates to reduce DRX grain size. So far, the result indicates that the precipitates are mostly along or close to the dynamically formed grain boundaries, which may be an indication of the precipitation pinning. The following results can be summarized:

1. The experimental result supports the FactSage calculation in terms of homogenization temperature and precipitation prediction.
2. Deformed microstructure is characterised by dynamic recrystallization and the grain size is significantly refined.
3. DRX grain size increases with decreasing strain rate.
4. Precipitation formation along the DRX grain may be associated with the grain boundary pinning.
5. Size of the precipitates may be an important factor in pinning effect.
6. Hardness increases with the decrease in strain rate due to the formation of more and larger Mg_2Sn precipitate.

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