The Evolution of Microstructure and Mechanical Properties of a 5052 Aluminium Alloy by the Application of Cryogenic Rolling and Warm Rolling

Ui Gu Gang*, Sang Hun Lee* and Won Jong Nam

School of Advanced Material Engineering, Kookmin University, Seoul 136-702, Korea

The microstructural evolution and the corresponding mechanical properties of a 5052 Al alloy processed by cryogenic rolling followed by warm rolling were investigated. The application of cryogenic rolling combined with warm rolling at 448 K showed a significant improvement of tensile strength up to 452 MPa. The kinetics and the microstructural evolution occurred during warm rolling were investigated using differential scanning calorimeter and transmission electron microscope. This notable increase of tensile strength was achieved by the formation of fine precipitates during warm rolling. It was found that the cryogenic rolling combined with warm rolling would be effective in improving mechanical properties. [doi:10.2320/matertrans.MD200801]

(Received March 31, 2008; Accepted July 10, 2008; Published December 25, 2008)

Keywords: cryogenic rolling, warm rolling, severe plastic deformation, precipitation

1. Introduction

The recent development of ultra-fine grained (UFG) aluminium alloys for structural applications has attracted much attention, because they are expected to provide extremely high strength at ambient temperature without degradation of toughness. Earlier works have shown that severe plastic deformation (SPD) processes, such as equal channel angular pressing (ECAP),¹⁻⁵⁾ accumulative roll bonding (ARB)⁶⁻⁸⁾ and severe torsional straining (STS),^{9,10)} etc. are effective in improving strength of metallic materials through the grain refinement.

Among these SPD processes, an ARB process is known to have a potential for continuous production of materials in bulk. In the process, a repeated procedure of cutting, stacking and roll-bonding of sheets at warm rolling temperatures introduces high plastic strain without any geometrical change. Thus, the microstructural evolution occurred during an ARB process, including work hardening, the subdivision of grains and dynamic recovery, has yielded ultrafine grains and high strength after several repetition cycles. However, an ARB process also would not be appropriate for practical application to produce bulk materials, since they require extremely large amount of plastic deformation and special procedures. Meanwhile, the deformation at cryogenic temperature is also recognized as an effective process to produce UFG materials. The suppression of dynamic recovery during deformation at extremely low temperatures is expected to preserve a high density of defects generated by deformation. Consequently, the larger driving force for recrystallization, due to the larger density of accumulated dislocations, would lower recrystallization temperature or accelerate recrystallization during annealing. Thus, the deformation at cryogenic temperature would require less plastic deformation for achieving ultrafine grains than other SPD processes at ambient or elevated temperatures.¹¹⁻¹⁷⁾ Thus, it is anticipated that the large stored energy accumulated during the deformation at cryogenic temperature would lead to the larger driving force for the occurrence of dynamic recovery and,

then, possibly reduce the required number of cycles in an ARB process to achieve ultrafine grains and high strength. Accordingly, the combination of cryogenic rolling with warm rolling, whose temperature range is similar to that of an ARB process, would become a good candidate for the formation of ultrafine grains with the less amount of the required deformation.

In view of the aforementioned, the present work was carried out to investigate the microstructural evolution and the corresponding mechanical properties of a 5052 Al alloy processed by the application of cryogenic rolling followed by warm rolling.

2. Experimental Procedure

A commercial 5052 Al alloy used for structural applications was chosen for the present work. The chemical compositions of Al alloy are 2.50 mass% Mg, 0.40 mass% Fe, 0.25 mass% Si, 0.25 mass% Cr, 0.10 mass% Cu, 0.01 mass% Zn and 0.10 mass% Mn. The material was annealed at 613 K for 2 hours and then quenched in water resulting in a grain size of 65 μm . The combination of cryogenic rolling with warm rolling consisted of two steps. Cryogenic rolling with 55% reduction was followed by warm rolling with 56% reduction (total reduction of 80% in thickness). As the first step, the plates, 8 mm in thickness, were rolled with the reduction of 55% at cryogenic temperature. Cryogenic rolling was performed by dipping plates into liquid nitrogen for at least 15 min before each rolling pass. The second step was carried out in the temperature range from cryogenic temperature to 573 K. Before each rolling pass of warm rolling, the sheets were heated at the temperature range of 448~573 K for 5 min to homogenize the temperature.

For tensile tests, the sheets rolled at various temperatures were machined into the ASTM subsize specimen of the 25 mm gauge length. Uniaxial tensile tests were conducted with the initial strain rate of $3 \times 10^{-3}/\text{s}$ on an INSTRON machine operating at a constant crosshead speed. For a detailed understanding of the microstructural evolution, a transmission electron microscope (TEM) was used. Thin foils

*Graduate Student, Kookmin University

parallel to the transverse cross section of the sheets were prepared by utilizing a conventional jet polishing technique in a mixture of 75% Methanol and 25% HNO_3 at the temperature of -30°C . Thermal analysis was performed under a flowing Ar gas in a NETZSC-DSC 200 F3 (differential scanning calorimeter, DSC) with the temperature calibrated using pure In, Zn, Bi and Sn standards. As the DSC peak position depends on the heating rate,¹⁸⁾ the measured data of the peak positions for different heating rates of 1 ~ 32 K/min in a flowing Ar atmosphere were used. When obtaining the activation energy, both the Kissinger¹⁹⁾ and Chen and Spaepen²⁰⁾ methods were applied in this work.

3. Results and Discussion

3.1 Microstructures and mechanical properties

Figure 1 shows the engineering stress-strain curves of 5052 Al alloy deformed at cryogenic temperature with 55% reduction and subsequently deformed at various temperatures with 56% reduction. The stress-strain curves, reflecting the characteristics of microstructures evolved during the deformation, show the different response to rolling temperatures of the second step. The further deformation at cryogenic

Fig. 1 The stress-strain curves of 5052 Al alloys deformed at cryogenic temperature with 55% reduction and subsequently deformed at various temperatures with 56% reduction (total reduction of 80% in thickness).

temperature led to the increase of tensile strength (TS) by 9.4% (30 MPa), compared with 320 MPa for cryo-rolling with 55% reduction. This indicates that cross-slip or climb of dislocations associated with dynamic recovery would be effectively suppressed during cryo-rolling and therefore the dislocation density remained high. The microstructure cryo-rolled with 55% reduction (Fig. 2(a)) consisted of parallel bands of severely elongated substructures along the rolling direction, 0.15~0.25 μm in width, while the narrower width of elongated substructures, 0.05~0.15 μm , containing higher density of dislocations, was observed in the sheet cryo-rolled with 80% reduction (Fig. 2(b)). The stress-strain curves in Fig. 1, except for a rolling temperature of 448 K, reflects the general trend that the increase of rolling temperatures in the second step accelerates softening due to dynamic recovery rather than work hardening. Meanwhile, it is interesting to note that TS increases from 320 to 452 MPa (nearly 40% increase) after warm rolling at 448 K. It is worth mentioning that the cryogenic temperature rolling with the same reduction results in TS of 350 MPa (9.4% increase), which is lower than TS, 452 MPa, of the sheet warm rolled at 448 K. To understand this unusual increase of TS, the microstructure of the sheet deformed at 448 K (Fig. 3(a), (b)) was examined, compared with the microstructure deformed at cryogenic temperature (Fig. 2(b)). The microstructure deformed at 448 K shows the similar characteristics to that deformed at cryogenic temperature, consisting of parallel bands of elongated substructures, 0.05~0.15 μm in width and containing a high density of dislocations. The only difference was the presence of fine precipitates, about 50~100 nm in diameter (indicated as arrows in Fig. 3(a), (b)). Thus, it can be expected that the remarkable increase of TS during warm rolling at 448 K would be closely related with the formation of fine precipitates. To investigate the effect of warm rolling on the formation of precipitates, hardness of samples, cryo-rolled and subsequently annealed for 5 min., is compared with that cryo-rolled and warm-rolled in Fig. 4. Static annealing at 448 K increases hardness from 103 Hv (cryo-rolled) to 111 Hv. This increase of hardness would be attributed to the formation of precipitates. However, high hardness of 127 Hv in samples cryo-rolled and warm rolled at 448 K cannot be explained with the contributions of the precipitation during static annealing and work hardening

Fig. 2 TEM micrographs of a 5052 Al alloys, (a) deformed at cryogenic temperature with 55% reduction and (b) deformed at cryogenic temperature with 80% reduction.

Fig. 3 TEM micrographs of 5052 Al alloys deformed at cryogenic temperature with 55% reduction and subsequently deformed with 56% reduction at (a) 448 K (arrows indicate precipitates), (b) at 448 K and SAD pattern (arrows indicate precipitates), (c) 523 K and (d) 623 K.

Fig. 4 The variations of hardness with temperature in 5052 Al alloys, received cryo-rolling and annealing, and cryo-rolling and warm-rolling.

during warm rolling. From the above discussion, it seems most probable that this notable increase of tensile strength would be caused by the finer precipitates produced during warm rolling at 448 K than during static annealing.

Warm rolling at 523 K, leads to the formation of subgrains and the rearrangement of dislocations including some loss of dislocations, as recovery proceeds. Many of elongated substructure boundaries recovered into subgrain boundaries exhibiting the distinct contrast (Fig. 3(c)). Simultaneously, the slightly increased subgrain width results in the reduction of the subgrain aspect ratio. The ultrafine subgrains with the

small aspect ratio would inhibit the formation of dislocation cells during tensile deformation.²¹⁾ Without the cell formation as well as the dramatic decrease of dislocation density, the slight increase of dislocation mean free path associated with the slight increase of the subgrain width does not cause any significant change in tensile properties in Fig. 1. That is, the small decrease of TS from 350 MPa (cryo-rolling) to 318 MPa (warm rolling at 523 K) would be attributed to little change of the dimension of dislocation substructures during warm rolling. It is worth mentioning that the warm deformation at 523 K produces the similarly shaped stress-strain curve to the deformation at cryogenic temperature with 55% reduction. This implies that during warm rolling at 523 K the degree of work hardening due to plastic deformation would be balanced with the degree of softening due to dynamic recovery. As rolling temperature increases up to 623 K, nearly equiaxed grains, whose diameters range from 100~200 nm, as well as elongated subgrains, become the dominant microstructure. It is expected that the occurrence of grain subdivision by deformation-induced boundaries observed by Tsuji, *et al.*⁸⁾ during an ARB process, would contribute to the formation of nearly equiaxed grains during the warm deformation at 623 K. Thus, the dominant effect of dynamic recovery caused a drop in tensile strength to 258 MPa. The increased width of subgrains and the low density of dislocations observed in Fig. 3(d) provide an evidence of dynamic recovery.

Figure 5 shows the relationship between TS and total elongation in severely plastic deformed 5 series Al alloys. Tensile stress of nano or ultra-fine grain structured 5series alloys is inversely proportional to total elongation. Such a

Fig. 5 The relationship between TS and tensile elongation for severe plastic deformed 5 series Al alloys. Number next to symbols denotes the pressing number. (* means work done in this study)

Fig. 6 DSC curves of 5052 Al alloys, deformed with 80% reduction at cryogenic temperature, at the heating rates of 2 and 16 K/min.

trend of strengthening accompanied by a loss of ductility is general for Al and other metals processed in various ways. However, the combination of cryogenic rolling with warm rolling moves from the region of tensile properties representing severely plastic deformed 5052 Al alloys to the region for severely plastic deformed 5083 Al alloys in Fig. 5 (indicated by an arrow). This implies that the combination of cryogenic rolling with warm rolling would become a useful method to achieve a superior combination of mechanical properties. Another important thing to note is that this achievement is obtained only rolling process without the application of any other severe plastic deformation process.

3.2 Kinetics of the precipitation

To understand the kinetics and the microstructural evolution occurred during warm rolling, thermal properties of cryo-rolled sheets with 80% reduction were examined using DSC. The DSC curves in Fig. 6 reveal three distinct exothermic peaks for the different heating rates of (a) 2 K/min and (b) 16 K/min. These peaks correspond to the heat evolved during the microstructural evolution, such as the formation of precipitates, recovery and recrystallization. For

Fig. 7 Calculated activation energies for the precipitation, recovery and recrystallization of the cryo-deformed 5052 Al alloy with 80% reduction.

the heating rate of 2 K/min, the temperature of the first broad peak was found as 459 K and that of the second peak was 489 K and that of the third peak was 598 K. Meanwhile, for the heating rate of 16 K/min, the temperatures of the observed peaks were 487, 518 and 629 K, respectively. The activation energies for the formation of precipitates, recovery and recrystallization could be determined with the data from the DSC measurements. The apparent activation energy of the DSC peaks obtained by analyzing a Kissinger plot¹⁹⁾ or a Chen and Spaepen plot²⁰⁾ is useful for understanding the mechanisms more precisely. According to the methods, the heating rate (B), the apparent activation energy of the process (Q), and the maximum temperature of the peak T_p are related by the eq. (1);

$$\ln(\alpha) = -\frac{Q}{k_B T_p} + C \quad (1)$$

where k_B is the Boltzmann's constant and C is an integration constant. Figure 7 shows plots of $\ln(\alpha)$ (where $\alpha = B/T_p^{19}$) and $\alpha = B/T_p^{20}$) against $1/k_B T_p$ and the activation energy Q calculated from each slope. The two methods show a reasonable agreement with each other, yielding Q values of 1.18~1.22 eV (113~117 kJ/mol) for the first peak, 1.40~1.44 eV (135~138 kJ/mol) for the second peak and 2.13~2.18 eV (205~210 kJ/mol) for the third peak. The apparent activation energy of the first peak, 1.18~1.22 eV, agrees with the measured activation energy for diffusion of Mg in Al in the dilute limit²²⁾ and is close to the calculated activation energy for Mg diffusion by the nearest neighbor mechanism (1.228 eV).²³⁾ This confirms that the mechanism for the first peak is the diffusion of Mg atoms in the matrix Al. Thus, it is expected that the unusual increased TS of the sheet deformed at 448 K would be caused by the formation of fine precipitates, which is controlled by the diffusion of Mg in the matrix Al. The Q value of the second peak, 135~138 kJ/mol, seems close to the activation energy for lattice self diffusion of aluminum (= 142 kJ/mol²⁴⁾), implying that the recovery would be mainly controlled by the self diffusion of Al atom. However, the measured activation energy of the third peak, 205~210 kJ/mol, seems considerably higher than

the reported Q values available for Al-Mg alloys. There has been no clear explanation regarding the mechanism of the third peak. To clarify the mechanism of the third peak, we compared the Q values of the third peak with the activation energy for recrystallization in 1050 Al alloy, 162~167 kJ/mol. And then it is expected that Mg in 5052 Al alloy would play an important role in increasing the activation energy required for the occurrence of recrystallization. Although the authors do not have physical insight at the moment, the pinning effect of boundary movements by fine precipitates^{5,25,26)} might become a likely candidate for the cause of the increase in the activation energy for recrystallization in a 5052 Al alloy deformed at cryogenic temperature.

4. Conclusions

The microstructural evolution and the corresponding mechanical properties of a 5052 Al alloy processed by cryogenic rolling followed by warm rolling, were investigated.

(1) The combination of cryogenic rolling with warm rolling was found more effective than a single cryogenic rolling process in improving mechanical properties of a 5052 Al alloy.

(2) Most stress-strain curves showed that the increase of rolling temperature effectively reduced tensile strength, due to the contribution of dynamic recovery. However, warm rolling at 448 K combined with cryogenic rolling was found to enhance tensile properties. This notable increase of tensile strength would be attributed to the formation of fine precipitates during warm rolling at 448 K.

(3) The formation of fine precipitates during warm rolling would be controlled by the diffusion of Mg in the matrix Al.

Acknowledgements

This work was supported by the 2007 research fund of Kookmin University in Korea.

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