Abnormal grain growth during annealing in the Al/Al₂O₃ composite produced by accumulative roll bonding

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Abstract

The mechanical and physical properties of composites are related to the matrix microstructure and dispersion of reinforcements. Microstructural evolution during annealing can change the properties of the composites. Consequently, in this paper, the microstructural evolution of Al/Al₂O₃ composite during annealing and the interpretation of metallurgical phenomena were investigated. Accumulative roll bonding technique was used for manufacturing the Al/Al₂O₃ composite. The Al/Al₂O₃ composite specimens were further annealed at 375 °C for 15, 30 and 60 min without any protected atmosphere. The grain orientation (texture) and the grain size of the composites were investigated. Results showed that by increasing the annealing time to 60 min, abnormal grain growth started. Moreover, the frequency of low angle boundaries (1.5–5) increased, while the frequency of $\Sigma^3$ Coincident Site Lattice boundaries decreased. In fact, grain boundaries with low Coincident Site Lattice were less prone to reinforcement pinning. Then low Coincident Site Lattice boundaries were more susceptible to formation and growth of abnormal grains.

Introduction

Special properties of metal matrix composites (MMC) contribute to their use in various industries. Aluminum composites are an important class of MMCs which have gained considerable popularity in the last three decades owing to their versatile properties [1, 2]. High abrasion resistance [3], good strength at high temperatures, low density, and high oxidation resistance of Al₂O₃ as a reinforcement have developed a special class of advanced materials, i.e. Al–Al₂O₃ composites [4–6].

The physical and mechanical properties of composites are dependent on the matrix microstructure and dispersion of reinforcements [7, 8]. Some specifications depend on microstructural evolution during annealing since it is very important and can change the mechanical properties. A driving force during annealing causes grain growth. This driving force is caused by severe deformation processes like rolling and friction stir processing [9]. Stored energy reduces during annealing because grain boundaries area decreases. Two classes of grain growth during heat treatment of metals are Normal Grain Growth (NGG) and Abnormal Grain Growth (AGG) [10, 11].

Normal grain growth increases the mean size of uniform grains of a material by migration of grain boundary and consumption of smaller grains [12]. Therefore, a monomodal grain-size distribution can be obtained. AGG, also referred to as the overstated or secondary growth is a kind of annealing phenomena. During annealing and after initial recrystallization, certain crystallites grow rapidly. As a result, bimodal grain-size distribution appears in the microstructure. Apparently, secondary recrystallization occurs when NGG is stopped [11].

Many factors can cause the AGG. The large pre-existing grains are one of them. In other words, AGG initiates from large pre-existing grains [11].

It is found [13] that the grains have the ability to abnormally grow with their size twice as large as the surrounding grains. The second most influential factor is Zenner Pinning phenomenon. A distribution of
second-phase particles, or reinforcements in the alloys and composites limit the migrating boundaries by applying a pinning pressure and delayed growth [14]. In the following, the dissolution and coarsening of the second-phase may occur, reducing the pinning pressure in some regions [11].

The third factor is misoriented boundary properties. Grain boundary character distribution (GBCD) may provide extensive data on the structure-property [15]. There are two types of boundaries, including low-angle (low mobility) boundaries and high-angle (high mobility) boundaries. This difference in the mobility of boundaries caused by the abnormal growth of some grains during annealing [16].

The fourth factor is Coincident Site Lattice (CSL) boundaries. At certain misorientations, one can get the perfect overlap of the lattice sites in the two crystals. The overlapping lattice sites create a new lattice called coincidence site lattice (CSL). CSL is characterized by \( \Sigma \) that is defined as the volume ratio of the unit cell of the CSL to that of the original crystal lattice [17, 18]. Special properties of boundaries are achieved by the high density of CSL lattice (low \( \Sigma \) value) [19].

The CSL boundaries have higher mobility and lower energy compared to the general grain boundaries [20]. Therefore, Coincident Site Lattice boundaries have less capability compared to Zenner Pinning [21].

The purpose of this research is to assess the microstructural evolution of Al/Al\(_2\)O\(_3\) composite during annealing, interpret the metallurgical phenomenon and develop a practical map of grain growth for this material.

**Materials and methods**

Accumulative roll bonding technique is used for manufacturing the Al/Al\(_2\)O\(_3\) composite samples. Commercially pure aluminum sheets (as the matrix) having a chemical composition as reported in table 1 were cut into specimens of dimensions 50 × 100 × 2 mm. Al\(_2\)O\(_3\) particles (<50 \( \mu \)m) were used as reinforcements. Figure 1 shows the initial grain structure of aluminum matrix. In order to fabricate Al/3vol% Al\(_2\)O\(_3\) composites, Al\(_2\)O\(_3\) particles were uniformly dispersed among aluminum sheets.
The strips were then roll-pressed together. During each rolling step, 50% reduction was obtained. During rolling, using laboratory rolling mill (at 25 °C), no lubricants were used. Rolling process was repeated up to eight cycles. Between each cycle, the strips were washed with acetone and then wire-brushed. This caused the interstices between the strips to be well bonded. The Al/Al2O3 composite specimens were further annealed at 375 °C for 15, 30 and 60 min. This temperature is within the recommended range for heat treatment. At lower temperatures due to prolonged heat treatment time, the surface oxidation of the samples increases. At higher temperatures, due to increased grain growth rate, the time variable is not easily controlled and the recrystallization process is performed rapidly and unpredictably due to severe deformation of the material.

The structural micrograph (figure 2a) showed the uniform distribution of Al2O3 in the aluminum matrix. Also, the strong bond between reinforcements and matrix achieved by the ARB process. The chemical composition of Al2O3 particles was also confirmed by EDS analysis (figure 2b).

The micro-texture and micro-structure of the composites were investigated by Hitachi SU6600 SEM equipped with EDS and EBSD. An accelerating voltage of 20 kV at a working distance of 15 mm was used. Because of using a step size of 50 nm, the EBSD could not detect the grains smaller than 50 nm. To perform EBSD data visualization and post-processing, the HKL CHANNEL5 software was used. The particle size, their distribution and grain size were carried out according to E112-96 ASTM standard on the surfaces using the CLEMEX image analysis software. X-ray diffraction (XRD) analyses were carried out using a Bruker 2D system, with Cu Kα radiation (λ = 1.54056 Å) to determine the texture of the deposits. The incomplete experimental pole figures data were used to obtain inverse pole figures (IPF) using TexTools software (RestMat Co.).

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Results and discussion

Isothermal grain growth

The rolling direction (RD)-inverse pole figure (IPF) and pole figure (PF) map of four specimens are shown in figure 3, (a) as-eight cycles of accumulative roll bonding without annealing and the other specimens annealed for (b) 15 min, (c) 30 min and (d) 60 min X-ray diffraction (XRD) analyses were carried out to determine the texture of the deposits (IPF part of figure 3).

It is quite obvious that these specimens had different overcoming textures and grain sizes. In the ARB specimen without annealing, the grains were quite straight along the RD. The texture components intensity for the ARB specimen without annealing was stronger than the annealed sample. The crystallographic orientation along the rolling direction was non-uniformly dispersed with a small peak near $<001>$ (figure 3a). After annealing the mixture for 15 min at 375 °C, no significant change was observed in the microstructure, while the mean grain size was about 23 μm. This means that recrystallization did not occur after 15 min, and possibly only recovery occurred. After 30 min annealing, recrystallization started and the grain size decreased to 3.3 μm. IPF map showed that the crystallographic orientation along RD was relatively uniformly distributed in this stage.

With increasing the annealing time to 60 min, abnormal grain growth started. Specimen with abnormal grain growth (AGG) was consisted of both unusually large grains (about 100 microns) and a matrix with uniform grains (about 5–10 microns). In fact, this specimen contained bimodal grain size dispersion.

Pole figures in figure 3 show that the dominant texture in all samples is (100). The intensity of this texture decreases by increasing annealing time.

Compared with the microstructural evolution of the samples annealed at different temperatures showed very similar grain growth behaviors. However, the grain growth became slower and took a longer time for the stable microstructure to be obtained [22].

AGG from the particle distribution view point

As mentioned before, reinforcing particles can limit the grain size by Zenner Pinning effect. Based on Zenner force, volume fractions of small particles for limiting grain size is expected to be $(4r/3F_v)$ and at large volume fractions it will be proportional to $(\beta r/\sqrt{F_v})$, where $r$ is the mean reinforcement radius, $F_v$ is the particle volume fraction in the structure and $\beta$ is a small geometric constant. The critical volume fraction at which there is a transition is not known precisely, but may be $\sim 0.05$ [23]. In this research, the circular equivalent diameter or area-equivalent diameter was used to calculate the mean radius of reinforcements which is defined as the diameter of a circle with the same area as the particle. Once the area of the particle, $A$, was measured, the area-equivalent diameter ($X_A$) can be calculated from equation $X_A = (4A/\pi)^{0.5}$ [24]. The mean radius of reinforcements was calculated to be about 15 μm. Also, the area fraction of the particle ($\text{Al}_2\text{O}_3$) was 0.026. A
study of quantitative stereology shows that the apparent area fraction determined on cut surfaces can be statistically represented by the actual volume fraction [25]. Therefore, by using the above relationship, the grain size should be about 6.7 $\mu$m (However, the measured grain size in the structure was about 7–8 $\mu$m).

According to [26] after normal grain growth and reaching the critical size, the second step of growth (AGG) is only possible after dissolution/coarsening of the reinforcement. However, the probability of this phenomenon, due to the low temperature, is low.

Indeed, at short annealing times, no significant change occurred in particle distribution. By prolonged heating, the inter particle spacing of the fine reinforcement increased [26]. According to the above, particles and reinforcements were considered to be mainly responsible for the grain boundary pinning (Zenner Pinning). Therefore, decreases or increases in particle spacing might state the next observations [26].

Some grain boundaries may not be limited by pinning. Also, reinforcement dissolution and reinforcement coarsening reduce the pinning force at the boundaries [11]. At this stage, the neighboring grains are merged together and AGG occurs (grain A in the figure 3d).

Understanding the effects of increasing the grain size during annealing (especially when reinforcement coarsening/dissolution occurs) on the grain boundary character distribution is very difficult [26]. The area fraction of particles in the abnormal/normal grain boundaries was greater than the area fraction of particles in the other surface area of the matrix [11]. This means that because of aggregation of the particles (reinforcements) at boundaries and an increase in the pinning pressure, some grains could not merge to larger grains (Grains A and B in figure 3d). In fact, both types of boundaries had comparable reinforcement fractions. This demonstrated that pinning force alone could not truly cause the grains to remain unmerged [11].

Grain growth may be prevented by Zenner pinning and/or neighbor grains having the similar orientation. By increasing the heating time to 60 min at 375 °C, the Zenner Pinning effect decreased. However, even in the specimen without reinforcements, grains can be significantly pinned. Actually, low angle/low mobility boundaries grain growth may inhibit the grain growth [26].

**AGG from the misorientation and changes in GBCD viewpoint**

Figure 4 shows the histogram of the distribution of grain boundaries for different annealing times at 375 °C. In the as-ARb sample (figure 4a) Grain boundary misorientations ranged from 1.5 to 60.5°. These GBs are shown by red and green lines in figure 5 (green smaller than 10 degrees and red higher than 10 degree). This figure shows that increased annealing time at 375 °C enhanced the frequency of low angle (1.5–5°) boundaries. For example, the frequency of 1.5° boundaries causes an increase from 0.67 to 0.85. Also, in the 60-min annealed sample (figure 4c), GB ranged from 0.5 to 16.5°.

It was expected that the density of low energy boundaries (low angle boundaries) increases with grain growth [26], which was also proved experimentally here. However, it is proved that misorientation alone cannot affect the stability of grains.

One of the best locations for forming unmerged grain is low-mobility boundaries (CSLs). Researchers have shown that $\Sigma 3$ misorientation in the boundaries prevents the grain growth and forms stable grains near large grains [11].

Figure 6 plots the fraction number of CSL boundaries ($\Sigma 3$–$\Sigma 49$) before and after annealing. This figure shows that annealing at 375 °C for 60 min decreased the frequency of $\Sigma 3$ CSL boundaries. In fact, $\Sigma 3$ CSL boundaries were consumed during abnormal grain growth.

The mobility of boundaries during annealing changes the grain boundary character distribution; high-energy boundaries are unstable and boundary tensions cause the growth of grains and reduce the total amount of high-energy boundaries [23].

Experimental studies [27, 28] have shown that annealing increased the low energy boundaries and CSL. Our studies on Al/Al$_2$O$_3$ composite indicated that the low energy boundaries (low angle 1.5°–5°) extremely enhanced and high angle boundaries removed with annealing.

The number of low-angle boundaries increased at 60 min when dispersoids or reinforcements were unstable. The surface tension in the boundary causes changes in the grain boundary distribution and affects the pinned boundaries. Consequently, a combination of both determines the final size and shape of the grains.

During annealing, a few overgrown grains determine the final microstructure and texture. These overgrown grains form the abnormal grain growth phenomena. It is possible to determine the orientation selection for AGG. In fact, to achieve higher mobility and low-energy boundaries, it is necessary to have grains larger than the mean grain size of the matrix. Some initial grains may also be approximating oriented.

We can determine Zenner Pinning pressure as $\left\{ (3F_r\gamma)/(2r) \right\}$, where $r$ and $F_r$ are the mean radius of the reinforcement and the volume fraction of reinforcements; reinforcement- grain energy interface is marked with $\gamma$ [23]. For low CSL boundaries (low energy boundaries), $\gamma$ and relatively the Zenner Pinning pressure are...
Figure 4. Histogram of grain boundaries distribution, (a) as-eight cycles ARB processed without annealing, (b) 15 min annealed, (c) 60 min annealed.
expected to be less. This means that a low Coincident Site Lattice boundary may be less prone to reinforcement pinning [26]. Then, low CSL boundary is susceptible to growth and formation of abnormal grains. In fact, $\Sigma^3$ CSL boundaries have been consumed by abnormal growth grain.

**Conclusions**

The crystallographic orientation along the rolling direction was non-uniformly distributed with a small peak near (001). By increasing the annealing time to 60 min, abnormal grain growth started and a bimodal grain size distribution occurred.

By increasing the annealing time, the decomposition and coarsening of confined particles led to a localized reduction in the pinning force. Then, the neighboring grains are merged together and abnormal grain coarsening might occur. Increasing the annealing time at 375 °C enlarged the abundance of very low angle (1.5°–5°) boundaries.

Prolonged annealing at 375 °C decreased the frequency of $\Sigma^3$ CSL boundaries and $\Sigma^3$ CSL boundaries were consumed by abnormal growth grain. In fact, larger grain than mean grain sizes of the matrix and also some initial grains may be approvingly oriented to achieve higher mobility and low energy boundaries. This means that a low Coincident Site Lattice boundary could be less prone to reinforcement pinning. Then low Coincident Site Lattice boundary was prone to growth and formation of abnormal grains.
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Compliance with ethical standards
Conflict of interest
The authors declare that they have no conflict of interest.

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