Diffusion and Formation of Intermetallic Compounds during Accumulative Roll-Bonding of Al/Mg Alloys

Ming-Che Chen¹, Che-Wei Kuo¹, Chia-Ming Chang¹, Chih-Chun Hsieh¹, Yin-Yu Chang² and Weite Wu¹

¹Department of Materials Engineering, National Chang Hsing University, 250 Kuo-Kuang Rd., Taichung 402, Taiwan, R. O. China
²Institute of Materials and System Engineering, MingDao University, 369 Wen-Hua Rd., Peetow, Changhua 52345, Taiwan, R. O. China

In this study, the accumulative roll-bonding (ARB) process is used to reduplicate Al (1100)/Mg (AZ31) alloy and then made thinner and longer by multi-rolling. Stacks with 24 layers were created and annealed at 300°C for 1 h. The microstructures were observed by using optical microscopy. The diffusion couples between Al and Mg were investigated to study the composition of the Al-Mg system. The layers of the intermetallic compounds Al₅Mg₁₂ and Al₁₇Mg₁₂ were observed by a field emission scanning electron microscope. The composition-depth curves of the diffusion zone were obtained by employing electron microprobe analysis after annealing at 300°C for 1 h.

Keywords: accumulative roll-bonding (ARB), aluminum/magnesium alloy, intermetallic compound, diffusion zone

1. Introduction

Aluminium and magnesium alloys are used in aerospace, electronic industries, and structural applications due to their unique characteristics. In recent years, lightweight materials composed of magnesium have been studied and used in many developed countries. The application of and research on magnesium and its alloys have been extended from naval and military applications to expensive civilian applications such as automobiles, computers, and communication equipment. Hence, the welding of Al and Mg to form a compound structure is necessary to obtain a low-weight and low-cost component. However, the formation of the refractory oxide film of Mg and Al results in inclusions of the weld metal. Moreover, since Mg possesses thermal brittleness, the fusion welding of Al and Mg, which are dissimilar materials, is difficult. This hinders the use of Al and Mg in the welding field. Using accumulative roll-bonding (ARB) process, the thermal brittleness and segregation resulting from fusion welding can be avoided.

ARB is a severe plastic deformation (SPD) process invented by the authors in order to fabricate ultrafine-grained metallic materials. ARB is the only SPD process that is applicable for the continuous production of bulky materials. The rolling performed in ARB in order to obtain a one-body solid material is not only a deformation process but also a bonding process (roll-bonding). By repeating this procedure, SPD of bulky materials can be realized. In the present study, the ARB process is applied to an aluminum alloy and a magnesium alloy, and the objective is to investigate the applicability of ARB process to Al/Mg alloy compounds and to clarify the production of the intermetallic compound. The microstructures at the interface of Al and Mg are analyzed by an optical microscope (OM) and a scanning electron microscope (SEM). The phase constitution in the Al/Mg alloy compound was analyzed by X-ray diffraction (XRD). This is helpful for the study of the ARB process and improves the microstructure performance of the ARB process; it also provides new ways to use Al/Mg compounds.

2. Experimental Procedure

The materials used in this study were pure aluminum (Al1100) and magnesium alloy (AZ31), whose chemical compositions are given in Table 1. The dimension of Al and Mg plates were 1 mm thick, 20 mm wide, and 200 mm long. Figure 1 illustrates the principle of the ARB process. Three pieces of sheets were stacked after degreasing them by acetone and sandblasting surface treatment; they were then roll-bonded by 50% reduction in thickness by one pass without lubricant at 300°C. The degreasing process and spray sand were required in order to obtain good bonding so as to prepare one-body solid material. The roll-bonded sheet was then cut into two sheets having dimensions identical to the initial dimensions. The samples were stacked and then rolled again.

The optical microscope (OM) observations and X-ray analysis were performed for longitudinal cross sections normal to the transverse direction (TD) plane. The OM was observed along the transverse direction of the rolled samples. The X-ray analysis was performed by employing a Mac MXIII X-ray diffractometer. The elements and intermetallic compounds distributed near the interface were examined by a JEOL JXA-8800M electron probe X-ray microanalyzer (EPMA) and JEOL JSM-6700F field emission scanning electron microscope (FE-SEM) and backscattered electron (BSE).

Table 1 Chemical composition of the Al (1100) and Mg (AZ31) alloys (in mass%).

<table>
<thead>
<tr>
<th></th>
<th>Si</th>
<th>Al</th>
<th>Cu</th>
<th>Mn</th>
<th>Zn</th>
<th>Ti</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al1100</td>
<td>1.0</td>
<td>98.75</td>
<td>0.05</td>
<td>0.05</td>
<td>0.1</td>
<td>0.05</td>
</tr>
<tr>
<td>AZ31</td>
<td>0.01</td>
<td>3.14</td>
<td>0.02</td>
<td>0.33</td>
<td>0.55</td>
<td>—</td>
</tr>
</tbody>
</table>
3. Results and Discussion

3.1 Microstructure analysis

The microstructures of the Al/Mg alloy compound after 1–3 cycles of the ARB process were displayed by Chen et al.;11) the authors showed that the microstructure changes and the interdiffusion found with Al and Mg atoms during the ARB process. In the present study, there were up to four cycles of the ARB process and the number of layers was increased to 24, as shown by the micrographs in Fig. 2. From this result, it can be observed that the Al alloy appeared in a continuous phase and the Mg alloy was covered by Al in the Al/Mg alloy compound. The structure of the fabricated layers was considerably dense for four cycles as compared to that for three cycles.11) According to the Al-Mg phase diagram (shown in Fig. 3), a new phase between the phases of Al and Mg—Al-Mg intermetallic compound—was produced. Since the rolling temperature was 300°C, the Al-Mg intermetallic compound was rapidly formed at the interface of Al and Mg. In Fig. 4(a), the bright region representing the intermetallic compound was found at the interface of Al and Mg after four cycles of the ARB process. The thickness of the intermetallic compound was approximately 4.5 µm. In order to observe the intermetallic compound clearly, the sample was heated at 300°C for 1 h after 4 cycles of the ARB process (shown in Fig. 4(b)). The thickness of the intermetallic compound was increased to approximately 25 µm after annealing.

3.2 Diffusion

The Al and Mg atoms can diffuse into the Mg and Al alloys, respectively, during the ARB process. Figure 4 shows the mapping image of Al/Mg alloy compounds (measured by SEM) for different ARB cycles and annealing temperatures. The image shows little diffusion between Al and Mg for one cycle of the ARB process with low strain (Figure 4(a)). In this cycle, the main bonding mode is mechanical bonding and the interface of the Al/Mg alloy compound clearly appears as a gap.11) Figures 4(b) and (c) show the diffusion region between the Al and Mg alloys. As the strain increases, the deformation process can be improved by applying a large quantity of heat to enhance the diffusion. When the sample is heated at 300°C for 1 h, the diffusion capability increases. Therefore, the thickness of the diffused region increases with the diffusion capability.

3.3 Phase identification

The XRD analysis was carried out using copper target for the following conditions: the working voltage was 40 kV and
the working current was 30 mA. The XRD results for the phase constituents of a cross section of the Al/Mg alloy compound heated at different annealing temperatures for 1 h after four cycles of the ARB process are shown in Fig. 5. The results were compared with the data obtained from the Joint Committee on Powder Diffraction Standards (JCPDS).

In this case, the diffusion region would contain an Al-Mg intermetallic compound that was near the interface of the Al and Mg alloys. According to the XRD results, the phase constituents of the cross section of the Al/Mg alloy compound were Al, Mg, Al<sub>3</sub>Mg<sub>2</sub>, and Al<sub>12</sub>Mg<sub>17</sub>. The first cycle of the ARB process did not produce any intermetallic compounds. The result was compared to Fig. 5(a) where there was no diffusion at the interface of the Al and Mg alloys, thereby leading to the development of the intermetallic compounds. Al<sub>3</sub>Mg<sub>2</sub> and Al<sub>12</sub>Mg<sub>17</sub> were discovered after four cycles of the ARB process and after four cycles of the process subsequent to the annealing at 300°C. It turned into intermetallic compounds of Al<sub>3</sub>Mg<sub>2</sub> and Al<sub>12</sub>Mg<sub>17</sub> when the diffusion zone formed.

### 3.4 Metallography and electron microprobe analysis of the diffusion zone

Figure 6 shows the interface of the Al/Mg alloy compound after the ARB process. Two intermetallic compound phases were found at the interfaces of Al and Mg, and the intermetallic compounds were Al<sub>3</sub>Mg<sub>2</sub> and Al<sub>12</sub>Mg<sub>17</sub>; these compounds were obtained after four cycles of the ARB process and annealing at 300°C for 1 h. From the phase diagram of the Al-Mg system<sup>12</sup> (shown in Fig. 8), the Al-rich edge and the Mg-rich edge were separated to yield Al<sub>3</sub>Mg<sub>2</sub> and Al<sub>12</sub>Mg<sub>17</sub>, respectively. From Figures 6(a) and (b), it can be observed that the phase constitution did not vary and the thickness of Al<sub>3</sub>Mg<sub>2</sub> and Al<sub>12</sub>Mg<sub>17</sub> increased after annealing. Since the annealing temperature was lower than 320°C, it was impossible for the other phases of the intermetallic compounds to form. The result conformed to the XRD data and confirmed that the intermetallic compounds developed after four cycles of the ARB process and grew rapidly during annealing at 300°C for 1 h.

The interfaces between all the four phases were more or less flat but showed a waviness of the order of a few micrometers. Figures 7(a) and (b) show the composition profile of the Al-Mg couple without annealing and with annealing at 300°C for 1 h, respectively, after four cycles of the ARB process. These images were obtained by electron X-ray microanalyzer that was performed for the diffusion direction. The composition measurement including the matrix of Al and Mg and the diffusion zone are shown in Fig. 6. Two solid solutions (Al(Mg) and Mg(Al)), two intermetallic compounds (Al<sub>3</sub>Mg<sub>2</sub> and Al<sub>12</sub>Mg<sub>17</sub>), and three interfaces (Al(Mg)/Al<sub>3</sub>Mg<sub>2</sub>, Al<sub>3</sub>Mg<sub>2</sub>/Al<sub>12</sub>Mg<sub>17</sub>, and Al<sub>12</sub>Mg<sub>17</sub>/Mg(Al)) could be identified. Al<sub>3</sub>Mg<sub>2</sub> and Al<sub>12</sub>Mg<sub>17</sub> near the interface of Al<sub>3</sub>Mg<sub>2</sub>/Al(Mg) and Al<sub>12</sub>Mg<sub>17</sub>/Mg(Al) comprised 40.26% and 62.38% of Al, respectively, after four cycles of the ARB process (shown in Fig. 7(a)). As shown in Fig. 7(b), Al<sub>3</sub>Mg<sub>2</sub> and Al<sub>12</sub>Mg<sub>17</sub> comprised 39.57% and 63.11% of Al, respectively, after annealing at 300°C for 1 h. The result showed that the composition range of the interface without annealing at 4 cycles closed to annealed situation. Similar results were also obtained for the other interfaces.

In addition, the composition ranges of the four dominating phases at the interface were practically independent of the annealing time.<sup>13</sup> In this experiment, the annealing time affected only the thickness of the intermetallic compound. The positions of the solid-solid equilibria are displayed in Fig. 8. The present phase fields for intermetallic compounds and for Mg(Al) were in fairly good agreement with those suggested by Ref. (12), whereas for the solubility limit of the primary Al(Mg) solution demonstrated slightly lower values.

### 4. Conclusions

In the present study, the diffusion reactions at the interface
of Al and Mg after four cycles of the ARB process with and without annealing at 300°C for 1 h were studied. The salient results are as follows:

1. An Al/Mg alloy compound with 32 layers was successfully obtained after four cycles of the ARB process.
2. The diffusion zone was obtained after four cycles of the ARB process without annealing and after annealing with thicknesses of 4.5 μm and 25 μm, respectively.
3. Two intermetallic phases—Al₃Mg₂ and Al₁₂Mg₁₇—were clearly identified, as expected from the phase diagram.
4. The interfaces of Al(Mg)/Al₃Mg₂, Al₃Mg₂/Al₁₂Mg₁₇, and Al₁₂Mg₁₇/Mg(Al) were obtained after four cycles of the ARB process without annealing and after annealing.
5. The compositions near the interface of the four dominating phases were measured closely after four cycles without annealing and after annealing.

Acknowledgement

This research was partially supported by the National Science Council, Taiwan, R.O.C., under project NSC 92-2216-E-005-005.

REFERENCES