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# Effect of annealing on the interface microstructure and mechanical properties of a STS–Al–Mg 3-ply clad sheet

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ABSTRACT

In this study we investigated the influence of post-rolling heat treatment upon the microstructure evolution at interface and subsequent uniaxial tensile properties of roll-bonded ferritic stainless steel (STS430)–aluminum (Al3004)–magnesium (AZ31) 3-ply clad metal. By utilizing optical, scanning electron and transmission electron microscopes, the generation and growth of interlayer consists of $\gamma$ ($Mg_{17}Al_{12}$) and $\beta$ ($Mg_2Al_3$) phases was verified between the constituent layers in the Mg/Al with total thicknesses of 4.56 and 11.21 μm during annealing at 300 °C for 1 and 3 h, respectively. Although as-rolled clad metal was somewhat joined by mechanical locking at the interface, annealing-induced generation of thin diffusion layer between AZ31 and Al3004 by annealing at 300 °C for 1 h resulted in the enhancement of uniaxial tensile properties in terms of elongation. However, further annealing for 3 h lead to weakening the interface bonding properties due to the significant generation of brittle intermetallic phases. The mechanism for retarding interface delamination along the direction of tensile axis was confirmed by the observation from the side surfaces of ex-situ stepped tensile-test specimens.


## 1. Introduction

Recently huge effort has been dedicated to fabricate clad metallic materials, a kind of typical laminated composites composed of more than two dissimilar metallic alloys joined at interface, which have been utilized in a wide range of industrial fields such as automotive, cookware, electronics, aerospace, food processing, and so on. Among the fabricating processes to achieve bonding-based clad metallic materials [1], solid-state bonding methods such as roll bonding [2–5], explosive bonding [6–8], friction stir welding [9], and diffusion bonding [10] have been mostly applied to generate multi-layered clad sheets. Taking both easy processes ability and cost reduction into account, roll bonding technique was the most widely used process to continuously fabricate sheet-type clad metals [11].

Typical metallic materials used for cladding matrix were stainless steel, carbon steel, titanium, copper and so on. In recent years, various studies for worldwide environmental protection have been underway, especially for reducing carbon dioxide emission. In order to achieve this goal, finding application fields of lightweight materials such as magnesium and aluminum alloys has been actively progressed [12–14]. For example, in order to broaden the application of magnesium alloys, stainless steel/magnesium (STS/Mg) combination can be drawn a growing interest as an excellent macro-scale clad metal combining the advantages of STS's high strength and excellent corrosion resistance with Mg's extraordinary lightweight and high specific strength properties. However, direct bonding between STS and Mg alloys is considerably restricted at present [15]. Therefore, roll bonding between STS and Mg alloy by inserting aluminum alloy is one way to produce lightweight 3-ply laminated composite. For utilizing unique multi-functional properties of the STS–Al–Mg 3-ply laminated composite, solid-state joining techniques have also been typically applied [16] because interface cracks can easily be generated by conventional fusion welding indebted to the higher level of brittleness of Mg at elevated temperature [17]. Another essential factor is that melting-based bonding easily induces the generation of detrimental intermetallic precipitates at the interface between dissimilar metals, which can be retarded or suppressed for the considerable process windows in terms of time and temperature in the solid-state bonding. Since the performance of laminated composites is governed by the formation of a strong metallurgical bond at interface between dissimilar metals, a good understanding of the generation of inter-diffusion layer is prerequisite for improving overall mechanical properties and formability of the laminated composites. For instance, the relatively higher interfacial hardness between magnesium and aluminum alloys is mainly attributed to strain localization during

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thermomechanically-induced bonding process [18], the increase of stress due to the difference in coefficient of thermal expansion [19], and the generation of solid-solution without noticeable formation of brittle intermetallic compounds [20,21]. However, the mechanism of retardation of catastrophic delamination resulting in the improvement of interfacial bonding strength as well as the overall mechanical property of the laminated composites composed of aluminum and magnesium alloys has not been apparent yet upon post-roll bonding process condition such as suitable annealing.

We present here an exploration of the relationship among post-roll bonding process variables such as annealing time, evolution of interface microstructure and overall mechanical properties of the 3-ply laminated composites composed of STS, Al and Mg in order. The effect of annealing conditions upon the evolution of interface microstructure is first analyzed by means of optical microscope (OM), scanning electron microscope (SEM), transmission electron microscope (TEM). Taking the composition and thickness of each intermetallic compound layer between Al and Mg into account, results devoted to the interfacial microstructure and overall mechanical properties of the 3-ply STS–Al–Mg are also given. The mechanism of improvement of interfacial bonding is also verified by means of ex-situ observation from the surface of stepped tensile-test specimens subjected to various true strain values. Finally the competitions between two contradictory effects, viz., interface strengthening by the generation of IMCs and the corresponding loose of overall ductility due to the embrittlement is discussed.

## 2. Materials and experiments

The raw materials used to roll bonding were a ferritic stainless steel (STS 430), an aluminum alloy Al 3004 and AZ31 magnesium alloy, whose composition were given in Table 1. Metallurgical bonding between the various metals results from the continuous roll cladding followed by additional annealing at elevated temperature. Fig. 1 exhibits the schematics to fabricate STS–Al–Mg 3-ply clad sheet with stacking sequence of the STS430–Al3004–AZ31 in order [22]. Initial size of the base metallic alloys was about 300 mm × 400 mm in width × length. Also an aggregate thickness of the base materials was above 7.5 mm, while the final thickness of the 3-ply clad sheet was around 3.83 mm. Warm roll bonding immediately followed by isothermal annealing for several seconds was carried out at around 400 °C, which is higher than the dynamic recrystallization temperature of the AZ31 ($\sim 0.6T_m = 260$ °C). The initial thickness of each materials were measured by a toolmaker's microscope as $0.549 \pm 0.005$ mm for STS 430, $1.041 \pm 0.018$ mm for Al 3004 and $2.290 \pm 0.015$ mm for AZ31, respectively.

**Table 1**
Chemical compositions of raw materials used in this study.

| | | | | | |
|---|---|---|---|---|---|
| STS (STS430) | C<br>≤ 0.12 | Cr<br>16–18 | Mn<br>≤ 1.0 | Si<br>≤ 1.0 | Fe<br>Bal. |
| Al (Al3004) | Mn<br>1–1.5 | Mg<br>0.8–1.3 | Fe<br>≤ 0.7 | Si<br>≤ 0.3 | Al<br>Bal. |
| Mg (AZ31) | Al<br>2.5–3.5 | Zn<br>0.6–1.4 | Mn<br>0.2–1.0 | Si<br>≤ 0.1 | Mg<br>Bal. |

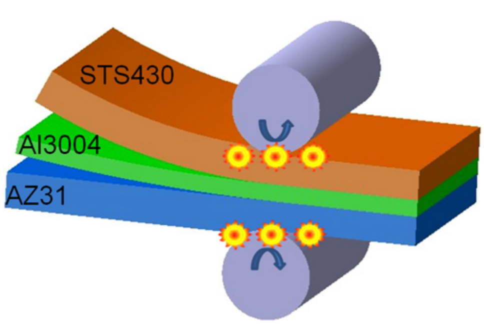


**Fig. 1.** Schematic diagram of roll bonding process.

**Table 2**
Conditions for stepped tensile tests.

| | |
|---|---|
| Samples | A0 (as-rolled)<br>A1 (annealed at 300 °C, 1 h)<br>A3 (annealed at 300 °C, 3 h) |
| Gage length (mm) | 20 (ASTM-E8M-01, sub-size) |
| Thickness (mm) | Multilayer sheet: 3.88<br>STS430: 0.55, Al3004: 1.04, AZ31: 2.29 |
| Temperature (°C) | Room temperature |
| Targeted elongation (%, true strain) | 3, 7, 11, 15, 19 (*in case it is available*),<br>100 (fracture) |
| Strain rate ($s^{-1}$) | $10^{-3}$ |

After roll bonding, the clad sheets were treated to reduce the residual stresses in a resistant furnace without any protective atmosphere for different post-annealing times of 1, 3, and 12 h at 300 °C, in order to form the metallurgical bonding at interfaces with different thickness of intermetallic compounds (IMCs). For simplicity, the 3-ply clad plates without post-annealing, annealed at 300 °C for 1, 3 and 12 h followed by furnace cooling are designated as A0, A1, A3 and A12, respectively. The cross-section of the samples were cut, mounted, ground and polished with a finishing of 1 μm diamond paste in order to characterize interface microstructures. Optical microscope (OM), field-emission scanning electron microscope (FE-SEM, model TESKAN MIRA II) attached with Oxford energy-dispersive X-ray spectroscopy (EDXS) and transmission electron microscope (TEM, model JEOL JEM-2100F) at 200 kV were applied to especially investigate evolution of the IMCs at the interface between Al 3004 and AZ31.

In order to investigate the influence of post-annealing process condition upon overall mechanical properties, dog-bone-type sub-sized tensile specimens, with gage length of 20 mm, were then cut from A0, A1 and A3 clad plates parallel to the rolling direction following ASTM standard E8M-01. Tensile tests were then run on a screw-driven UTM attached with a 100-kN load cell at a nominal strain rate of $10^{-3}$ $s^{-1}$. Longitudinal displacements were measured with a double-sided clip-on extensometer. Detailed tensile-test conditions are summarized in Table 2. In order to further investigate delamination mechanism, ex-situ images of laminated surface were observed after uni-axial, stepped tension with controlled true strain values of 3, 7, 11, 15 and 19% from the gage length of the tensile specimens.

## 3. Results and discussion

### 3.1. Microstructural evolution at interfacial diffusion layers

Figs. 2(a)–2(d) exhibit the OM micrographs of the cross-sectional interfaces from STS–Al–Mg 3-ply samples A0, A1, A3 and A12, respectively. Fig. 2(a) reveals initial roll bonding process adopted in this research yields relatively sound interface free of pores, cracks or lateral delamination at Mg/Al and Al/STS interfaces. It is apparent from Figs. 2(a)–2(d) that Al/STS interfacial transition zone seems to be similar regardless of further annealing at 300 °C for 12 h. On the other hand, the thickness of diffusion zone at interface between Mg and Al from sample A3 undergone post-annealing at 300 °C for 3 h is drastically increased as shown in Fig. 2(c). It is interesting to note from