



# Application of accumulative roll bonding and anodizing process to produce Al–Cu–Al<sub>2</sub>O<sub>3</sub> composite



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## ABSTRACT

In the present investigation, production of Al–Cu–Al<sub>2</sub>O<sub>3</sub> composite by means of Accumulative Roll Bonding (ARB) coupled with the anodizing process was studied. For this purpose, the alumina was grown on Al sheets by electrolyte technique and then the coated Al was laid between two Cu sheets followed by roll bonding to a specific reduction. This process was repeated up to seven times in order to achieve a bulk composite. The microstructure was characterized by SEM and optical microscopy while the mechanical properties were measured by microhardness, triple point bending and tensile testing. Microstructural evolution of the produced composite revealed that alumina was fractured in the primary sandwich and distributed non-uniformly throughout the composite. However, the alumina distribution was improved as the ARB cycles proceeded. It was also found that the tensile strength was improved up to the third cycle, after which it was decreased for the fourth and fifth cycles and again, it was increased for the last cycles. The bend strength showed the same trend as the tensile strength, while the elongation represented weak values for almost all cycles. Moreover, it was observed that as strain was increased (more ARB cycles), the microhardness for both Al and Cu layers was increased by two different trends. Additionally, failure analysis revealed that the mode of fracture was governed by two mechanisms: micro crack initiation between the metallic layers and formation of micro voids mainly around the alumina particles followed by their coalescence.

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## 1. Introduction

Metallic Multilayered Composites (MMCs) as advanced materials have a great potential to be employed for applications such as aerospace and automobile [1]. For producing such composites among the candidate metals, Al, due to its special properties, has received great attention [2].

Among methods of producing these composites, Accumulative Roll Bonding (ARB) technique has been successfully employed in the recent decades [3]. In this respect, different variables such as layer material [4–6], reinforcement particles [7,8] and using supplementary processes such as heat treatment [9,10] have been optimized in order to enhance the mechanical properties of the composites.

A new method using anodizing process coupled with ARB process was recently introduced to achieve a uniform particulate Al matrix composite [11]. It was reported that the main advantage of utilizing anodized layer was producing a highly uniform

composite. In this regard, other researchers combined electroplating and ARB processes to produce a multilayered Al–Cu–Ni composite in which Cu fragmentations and Ni particles acted as reinforcement agents in the Al matrix. It was reported that as strain was increased (i.e., proceeding ARB cycles), Ni layers were firstly fractured and then Cu layers yielded necking and fracturing, resulting in a uniform distribution of all metals. Besides, mechanical properties such as tensile and bend strength were increased after 11 cycles [12].

In this investigation, the potential of ARB process to produce Al–Cu–Al<sub>2</sub>O<sub>3</sub> composite was evaluated using coatings produced with the anodizing process. For this purpose, as the first step, a range of different parameters were optimized to achieve the best conditions for Al<sub>2</sub>O<sub>3</sub> coatings and reductions during cold roll bonding [13]. As the second step, the effects of different amounts of Al–Cu composite and also, the subsequent annealing treatment on Al–Cu composites were investigated [14]. In the present study, as the third step, an Al–Cu–Al<sub>2</sub>O<sub>3</sub> composite were produced following the previous studies. The mechanical properties were determined through microhardness and tensile testing while microstructural development was characterized by metallography and fracture analysis.

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## 2. Experimental procedure

### 2.1. Materials and surface preparation

Commercially pure Al and Cu strips with specifications given in Table 1 were employed. The Cu and Al strips (200 mm long, 50 mm wide) were cut parallel to the original rolling direction from cold rolled initial strips. In order to achieve composites with different amounts of Cu and Al, different thicknesses of Al and Cu (150, 300, 500 and 1000  $\mu\text{m}$ ) were used. The Al and Cu strips were first annealed at 370  $^{\circ}\text{C}$  and 480  $^{\circ}\text{C}$  for 1 h, respectively. This was followed by air cooling to room temperature. The strips were then de-greased with acetone to remove surface contaminations. Contaminated layers including oxides, adsorbed ions (ions of sulfur, phosphor and oxygen), greases, moisture and dust particles could impair the formation of a strong joint during cold rolling. Preparation process was followed by scratch brushing on the side of the surfaces at the peripheral speed of 2000 rpm for at least 60 s. The brushing was performed parallel to rolling direction using a stainless steel circumferential brush made of wires 0.25 mm in diameter. Handling of Al and Cu strips after preparation and stacking was done carefully to avoid renewed contamination, in addition to immediate cold roll bonding process. In addition, the time between surface preparation and rolling was kept less than 60 s [12].

### 2.2. Anodizing

In order to produce Al–Cu composite with  $\text{Al}_2\text{O}_3$  particles, alumina was grown on the Al strips using the anodizing process. Al strips were cleaned in NaOH and anodized in 15 wt.% sulfuric acid under an applied voltage of 15 V. In order to obtain a uniform oxide thickness, strips were held in electrolyte for a soaking time of 30 min. To ensure a constant and homogeneous temperature (i.e., 10  $^{\circ}\text{C}$ ) throughout the solution, the forced convection was provided by electrolyte stirring. The thickness of the alumina layer was determined using standard metallographic procedures with 20 measurements for a given sample. The thickness of  $\text{Al}_2\text{O}_3$  was determined to be  $20 \pm 0.2 \mu\text{m}$ .

### 2.3. ARB process

This process, as schematically shown in Fig. 1, consisted of two steps. In the first step, an anodized Al strip was laid between two Cu strips and fastened at both ends using steel wire. Accordingly, the above mentioned couple was roll-bonded to 50% reduction. In the second step, the roll-bonded strips were cut in half and then the two intimate Cu surfaces were scratch-brushed, stacked on each other and fastened at both ends to be roll-bonded (50%). The latter was repeated up to 7 cycles. The alignment of the strip edges prior to rolling had to be strictly considered. The ARB process was carried out with no lubrication, using a laboratory rolling mill with a rollers diameter of 125 mm, a rolling speed of 2 m/min and a loading capacity of 20 tons. To investigate the effect of different thicknesses of strips on the microstructure evolution of the composites, two different compositions of composites named Al–75.5%Cu–3% $\text{Al}_2\text{O}_3$  and Al–21.5%Cu–3% $\text{Al}_2\text{O}_3$  were manufactured by altering the initial metallic layers. For this purpose, anodized

Al strips with the thickness of 300 and 1000  $\mu\text{m}$  were laid between two Cu strips with the thickness of 500 and 150  $\mu\text{m}$ , respectively.

### 2.4. Microstructural studies and mechanical properties

The microstructures of composite strips were investigated using optical microscopy (OM) at different directions on (Rolling Direction) RD, (Normal Direction) ND, ND-(Transverse Direction) TD and RD-TD planes. Also, the fractured surface of composites was studied by SEM. The tensile test specimens were wire cut from the ARBed strips according to the ASTM E8M standard along the rolling direction. Tests were conducted at room temperature using a Hounsfield H50KS testing machine at an initial strain rate of  $1.67 \times 10^{-4} \text{ s}^{-1}$ . Microhardness test was performed under 50 g load on RD-TD plane of the composite layers. Triple point bending test was conducted on composites at different ARB cycles. The bend strengths of the specimens were calculated from Eq. (1):

$$\sigma = \frac{3FL}{2Wt^2} \text{ (MPa)} \quad (1)$$

where  $F$  is the bending load (N),  $L$  is the distance between the two supports (mm),  $W$  is the samples width (mm) and  $t$  is the thickness (mm) [15].

## 3. Results and discussion

### 3.1. Microstructure

Fig. 2 shows the primary sandwich of the roll-bonded Al–Cu strips. As can be seen, during plastic deformation, alumina layer was fractured within which bonding occurred in limited spaces in the middle of alumina fragments. Also, Fig. 3 indicates the microstructure evolution of Al–75.5%Cu–3% $\text{Al}_2\text{O}_3$  composite at different planes, including RD-ND, RD-TD and TD-ND, during ARB cycles. As can be seen, alumina particles fractured during the primary cycles were unevenly distributed among the layers. Moreover, Cu layers started necking and fracturing in some areas within the third ARB cycle as shown by arrows in Fig. 3(a). It is noteworthy to mention that Cu layers kept their continuity and did not fracture in most areas at the third cycle as can be seen in Fig. 3(a) (RD-ND and ND-TD planes). From the fifth cycles onward, a composite with continuous Al layers and fractured Cu distributed in the matrix were achieved, Fig. 3(b and c). Despite the fact that at the higher stages of ARB cycles, a better distribution of alumina fragments was observed in the matrix of the composite, there were still particle free zones and agglomerations of particles (agglomeration of the particles is denoted by ellipses in Fig. 3(c) on different planes after the last ARB cycle.

During plastic deformation of a dissimilar multilayered composite, some instabilities including necking and fracturing of harder layers were reported by a number of studies [5,16]. In most cases, differences between the mechanical properties of metallic layers were found to contribute to instabilities. In the present study, the differences between the mechanical properties of Al, Cu and alumina layers resulted in fracturing in the harder layers, i.e., alumina and Cu as shown in Figs. 2 and 3. It is important to note that other possible reasons for such a behavior can be summarized as follows [12,16,17]:

**Table 1**  
Specifications of initial Al and Cu strips.

Material	Chemical composition (wt.%)	Condition	Tensile strength (MPa)	Elongation (%)	Hardness (HV)
Al	99.64Al, 0.24Fe, 0.04Si, 0.02Ti and other element were balanced	Annealed	80.1	25.0	21.4
Cu	99.86Cu, 0.06Fe, 0.02Al, 0.01Sn and other element were balanced	Annealed	189	40.4	53.2

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