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High-strength and high-conductive Cu/Ag multilayer produced by ARB

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ABSTRACT

In this research high-strength and high-conductive multilayered Cu/Ag composites were produced by accumulative roll bonding (ARB) process for the first time using Cu and Ag strips up to nine cycles. Electrical resistivity changes were measured by the four-point-probe method and tensile tests were performed to investigate the mechanical behavior of ARB products. Finally scanning and transmission electron microscopy (SEM and TEM) were used to study the microstructure of the layers.

1. Introduction

Cu/Ag microcomposites have attracted interest because of their advantageous combination of excellent mechanical strength and relatively high electrical conductivity [1]. The microcomposites, have technological importance in windings in high-field magnet design electronic devices [2], advanced lead frames in large-scale integrated circuits [3], high magnetic field research, electric railways, heat transfer components, plasma-facing components [4], contacts and interconnect layers in semiconductor industry [5] and applications that require functional properties in the form of a non-equilibrium system [6].

To date, various methods including cold drawing combined with intermediate heat treatments (thermomechanical processing) for Cu/Ag wires [7,8] roll-bonding and subsequent diffusion annealing of Cu and Ag strips [9] thin film deposition [10,11], non-equilibrium processes including rapid quenching, plasma processes [12,13] and severe plastic deformation methods like mechanical alloying [14,15] have been developed for the fabrication of Cu–Ag microcomposites.

Thin film deposition with complicated processing has limited the potential applications for commercial use. Conventional thermomechanical processing methods are not suitable for manufacturing ultrafine grained (UFG) materials, since the minimum grain sizes that can be achieved are of the order of a few microns. Severe plastic deformation (SPD), which is a production method that consists of applying very large strains, often to a bulk sample, has proven to be a very effective way of fabricating UFG materials. UFG bulk metals and composites are considered very attractive structural materials since they are significantly stronger than their coarse-grained counterparts and retain good ductility. Additionally, UFG metals have improved corrosion resistance and potential for superplasticity at high strain rates and low temperatures [16,17]. Nevertheless, there has been few works on the production of Cu–Ag microcomposites with UFG microstructure by SPD. Recently Koa et al. [18] have used a combination of equal channel angular pressing and subsequent cold rolling to investigate the mechanical and electrical responses of nanostructured Cu–Ag alloy as a function of the imposed strain. By combining forced mixing and decomposition in ball-milled Cu50Ag50 powders, Zghal et al. [15] reported the formation of Cu/Ag nanocomposite with a very high hardness. Accumulative roll bonding (ARB) is one of the severe plastic deformation (SPD) processes which can produce bulk ultrafine grained (UFG) metallic materials [19,20]. Thus far, the ARB process has been applied to fabricate various ultrafine grained metallic sheets and multilayered composites owing to the relatively simple processing [21–24]. Ohsaki et al. [25] operated the ARB process on Cu–Ag eutectic alloy as attempts of bulk mechanical alloying.

In this study, the ARB process was used to produce Cu/Ag multilayered nanocomposites for the first time by choosing Cu and Ag strips as sandwich sheet layers. By this method we produced the microcomposite directly without any further processing. The gradual change of the microstructures as well as electrical and

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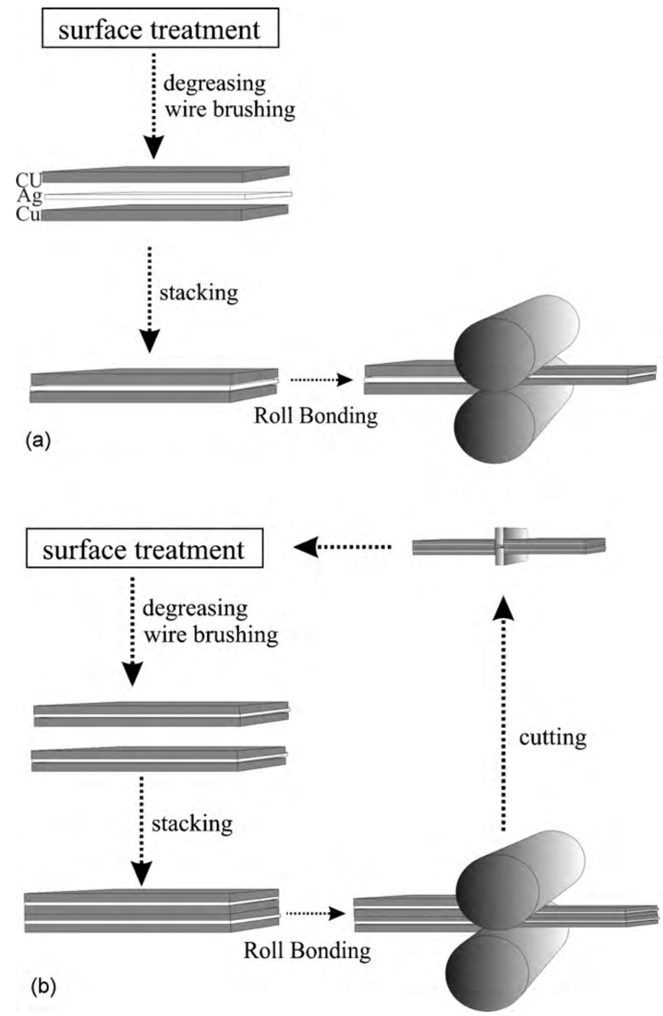

Fig. 1. Schematic illustration showing the principle of the accumulative roll bonding (ARB) process: (a) sandwich preparation cycle and (b) ARB cycles.

mechanical properties of the nanocomposite products were examined and discussed.

2. Experimental procedures

Commercially available copper of 1.0 mm thickness and pure silver of 0.1 and 0.2 mm thicknesses were cut into 3 mm × 150 mm. After degreasing and wire-brushing of the sheet surfaces, they were stacked in the manner of two copper layers with one core silver layer. The stacked sheets were first roll-bonded with 57% reduction in thickness, corresponding to the Von Mises equivalent strain of 0.99 (Fig. 1(a)).

Data regarding the sandwich preparation cycle including the entry thickness, the exit thickness, the reduction per cycle and the qualitative observations concerning the bonds are given in Table 1. These data showed that in the zero cycle, the bonds were generally well-formed as long as the reduction in the strip thickness was above a certain limit. This reduction was estimated to be approximately 56%. When the reduction was below this level, the bonding appeared weak. Therefore, the obtained roll-bonded sheets had two sandwich structure groups: Group1-Ag100 with a 100 μm thick Ag sheet in the core, Group2-Ag200 with a 200 μm thick Ag core.

Table 1
Examination of the sandwich preparation cycle.

Samples' number	$h_{in\ (mm)}$	$h_{out\ (mm)}$	Reduction %	Bonding quality
1	2.1	0.92	56	High
2	2.1	0.8	62	High
3	2.1	0.91	57	High
4	2.1	1.1	48	Low
5	2.1	1.4	33	Low

Examination results for the sandwich preparation cycle.

In the next stage, the roll-bonded sandwich was cut into two halves. The contacting surfaces of the halves were degreased, scratch-brushed and after being stacked over each other, roll-bonded with a fixed percentage of 50% reduction (the Von Mises equivalent strain of 0.8). This step, Fig. 1(b) of the process, was repeated up to nine cycles. After each cycle, the strips were prepared to be stacked and roll-bonded again. Therefore, 10 roll-bonding cycles were performed in total; one cycle in the first stage (denoted by zeroth cycle) and nine cycles in the second stage by the ARB process.

The roll-bonding experiments were carried out at room temperature without any lubricant using a laboratory rolling mill with a loading capacity of 20 ton. The roll diameter was 170 mm, and the rolling speed (ω) was 12 rpm. The tensile test samples with the tensile axes parallel to the rolling direction were 8 and 3 mm in gage length and width, respectively, which correspond to one-fifth of the JIS-5 standard dimensions [21]. These samples were machined from the products of the different cycles of the ARB process. Tensile tests were conducted at 25 °C and the initial strain rate of 10^{-4} s (using an Instron machine). The microstructures of Cu/Ag composites were studied using a scanning electron microscope (Vega©Tescan) in the back scattering mode before which the cross-section of the samples were ground, polished, etched in a solution of 100 ml C_2H_5OH, 5 ml HCl and 2 g $FeCl_3$ and repolished. The cross-sections of the samples were also examined using a transmission electron microscopy (200 kV, JEM-2100 LaB6). For this purpose, a thin slice from ND–RD plane was cut out from the rolled samples. The thickness of the sample was reduced to 50 μm by using sand paper followed by a dimple grinder. The thin slice was carefully glued to a copper grid and subjected to 12 h of precision ion milling with a radiation angle of 5°.

The X-ray diffraction patterns were recorded on the RD and TD plane surfaces of the ARB-processed sheets. 40 kV 30 mA Philip X'pert pro with negligible instrumental broadening using Cu Kα_1 radiation (λ = 0.15406 nm) in the range 2θ = 20–100° using a step size of 0.02° and a counting time of 0.4 s per step. The classic Williamson–Hall method [26] was used for measuring the crystallite size.

Electrical resistivity was measured at ambient temperature via the four-point-probe direct current technique from the surface of the samples. The resistivity of each sample was measured ten times and the mean value was reported.

3. Results and discussion

3.1. Critical reductions

In this investigation the amount of critical reduction to achieve sufficient copper-to-silver bonding strength in the zeroth cycle was (56%). Yang et al. [27] have recently investigated the bond strength of various metal multilayers produced by cold rolling of metal foils with different thermal conductivity. Results have indicated that the

Table 2
Variation in the thickness and number of layers by increasing ARB cycles.

ARB cycle (n)	Number of Cu layer (2×2^n)	Number of Ag layer (2^n)	Total layer number (3×2^n)	Equivalent strain (ε) ($0.98 + 0.8n$)	Final thickness of Cu (μm)	Final thickness of Ag (μm)
0	2	1	3	0.98	430	42.8
1	4	2	6	1.78	215	12.46265
2	8	4	12	2.58	107.5	7.15791
3	16	8	24	3.38	53.75	4.11114
4	32	16	48	4.18	26.875	2.36123
5	64	32	96	4.98	13.4375	1.35617
6	128	64	192	5.78	6.71875	0.77892
7	256	128	384	6.58	3.35938	0.44737
8	512	256	768	7.38	1.67969	0.25695
9	1024	512	1536	8.18	0.83984	0.14758