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Interfacial state and characteristics of cold-sprayed copper coatings on aluminum substrate

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Abstract. Cold spray is a new process for bonding copper and aluminum. In this paper, the interfacial states of cold-sprayed copper on aluminum substrate were evaluated and compared with those resulting from hot extruding. No critical intermetallic layers were observed at the cold-sprayed interfaces and good electrical and mechanical properties were achieved. The effect of annealing on the characteristics of cold-sprayed bonding material was also examined. By annealing at appropriate temperature, a continuous layer of an intermetallic compound was formed and the electrical conductivity and bending strength were improved. This indicates that a good bonding state is obtained by using cold-spray technology in combination with subsequent thermal processing.

1. Introduction

Copper-aluminum bonding material is expected to be used for conductive parts, such as electrodes and connectors for electric vehicles and heavy electric machineries, and several other applications. The hot extruding method has been predominantly used as the process for bonding copper and aluminum. However, due to the formation of intermetallic compounds through the high-temperature process, it is difficult to obtain high mechanical strength and good electrical conductivity.

Recently, cold-spray technology has emerged as an intermetallic bonding method. In this process, compressed gas is injected into a Laval-barrel nozzle to produce a supersonic jet. At the same time, metal particles are fed by a turntable with nitrogen gas. Then, the metal particles are injected into the supersonic jet of compressed gas and accelerated to a high velocity of nearly Mach-2 (300–1,200 m/s) [1,2]. The accelerated particles are sprayed toward the substrate and plastic deformation occurs when colliding with the substrate. A bonding mechanism is inferred when the metal particles collide with a metal substrate. The metal bonding occurs in addition to the anchor effect caused by plastic deformation [3]. Since bonding occurs below the melting point, the bonding materials have the following characteristics, among others: no phase transformation, little oxidization, small thermal stress.

Previous studies reported that the particle deposition depends on the impact velocity. It is also suggested that the impact velocity must be higher than a certain critical velocity to obtain good bonding conditions. Below the critical velocity, colliding particles would cause only erosion of the substrate [4,5]. It is also proposed that the critical velocity depends on the combination of the particle and substrate materials [6,7]. The critical velocity is approximately 600 m/s in case of depositing copper particles on aluminum substrate. Spraying of copper particles on aluminum substrate is a relatively easy deposition process.

In this paper, the focus is on the interface states generated by cold spraying and hot extruding, and a comparison of the resulting mechanical and electrical properties is provided.

2. Experimental procedure

A commercially available cold-spray system (PCS-305, Plasma Giken Co. Ltd., Japan), as shown in Figure 1, was employed to make copper coatings by using nitrogen gas. The cold spray parameters are listed in Table 1.

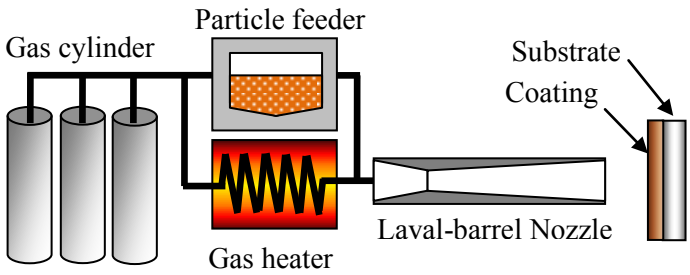


Figure 1. Schematics of the cold-spray system.

Table 1. Cold spray conditions.

Gas type	N2
Main gas temperature [K]	873
Main gas pressure [MPa]	3
Spray distance [mm]	25
Particle feed rate [g/min]	300

In this study, a pure aluminum block (100 × 100 × 50 mm^3) was used as substrate material. Non-spherical copper particles with an average particle size of 35 μm and 99.9% purity, as seen in Figure 2, were used to deposit the coatings up to 50 mm thick onto the substrate.

The specimens for cross-sectional observations of the surface were prepared by electrical discharge wire processing. Shape and size of the cross-sectional samples are shown in Figure 3 (A). Next, the specimens were examined by scanning electron microscopy (SEM, Carl Zeiss, Sigma, Germany) after being filled with resin and subsequent optical polishing. Furthermore, the samples were observed by transmission electron microscopy (TEM, JEOL, AMR-200F, Japan). In order to examine the possible generation of intermetallic compounds at the interface, sample (A) was also heat treated in vacuum. Annealing was conducted under various conditions, at temperatures ranging from 523 K to 773 K and holding times from 4 h to 300 h.

The specimens for the investigation of the mechanical and electrical properties were made with corresponding sizes and shapes as shown in Figure 3 (B) and (C). Samples of type (B) were examined by three-point bending tests after conductivity measurements using specimens of size 2 × 2 × 80 mm^3. Measurements of the electrical conductivity were carried out using the four-terminal method shown in Figure 4. The measurement distance was set at 35 mm when measuring copper conductivity. In case of measuring the conductivity of copper-aluminum including the bonding surface, the distance was set at 66 mm. As shown in Figure 5, the three-point bending tests were conducted with a displacement rate of 1 mm/min and fulcrum-gap distance of 30 mm. Samples of shape (C) were used for tensile tests, as shown in Figure 6.

The hot-extruded samples serving as comparison material were prepared from commercially available materials.

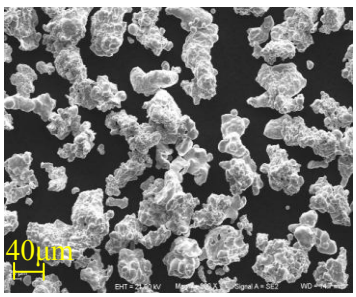


Figure 2. Morphology of copper particles.

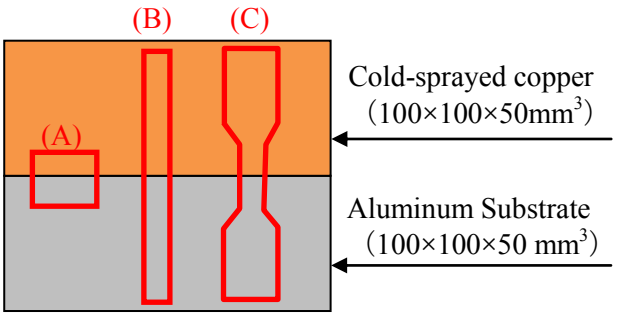


Figure 3. Shape and size of the specimens.

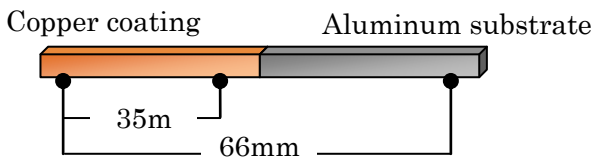


Figure 4. Layout of the conductivity measurements.

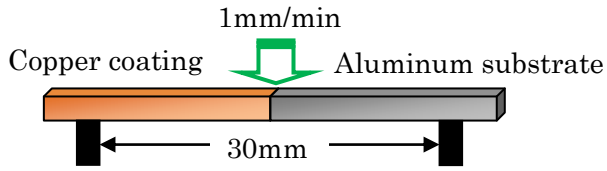


Figure 5. Schematic diagram of the bending test.

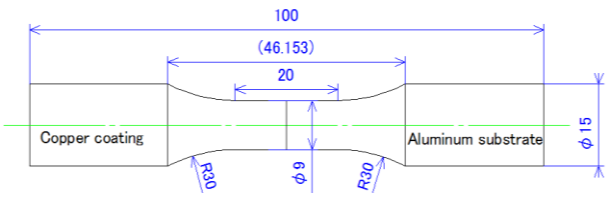
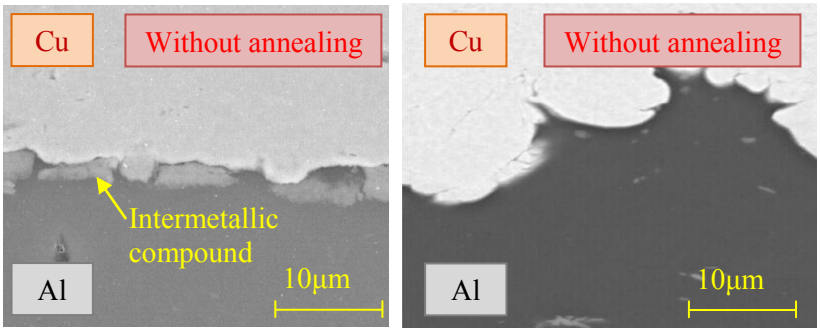


Figure 6. Shape of the tensile-test specimen.

3. Results and discussion

3.1. Cross-sectional observations

Figure 7 shows SEM images of the hot-extruded interface (A) and cold-sprayed interface (B). After the hot extruding process, discontinuous intermetallic compounds were observed at the bonding interface. On the other hand, employing the cold spray process, an intermetallic compound was not clearly observed in the SEM images. Therefore, the cold-sprayed interface was additionally examined by TEM and a continuous intermetallic layer of 200 nm in thickness was observed, as shown in Figure 8. Thus, these results clearly show that the formation of an intermetallic compound in the cold spray bonding process is suppressed compared with in the hot extruding process. Cold-sprayed interfaces were also observed after annealing, shown in Figure 9. As a result, the intermetallic layer gradually grew by high-temperature annealing in a uniform and continuous manner. Therefore, a stable bonding between aluminum and copper was not created in the hot extruding process, whereas it was confirmed that the cold sprayed interface was in a stable bonding state. Figure 10 shows the growth curve of the intermetallic compound at 523 K. According to this graph, the intermetallic compound is expected to grow up to a thickness of 15 μm after 2,000 h.



(A) Hot extruding interface (B) Cold sprayed interface

Figure 7. SEM images of bonding interface.

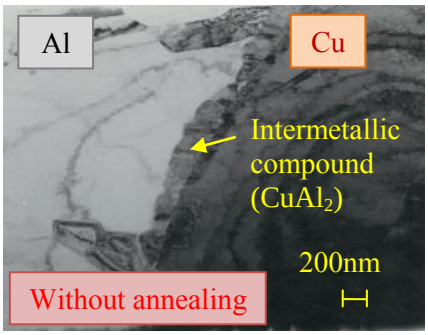


Figure 8. TEM image of cold-sprayed interface.

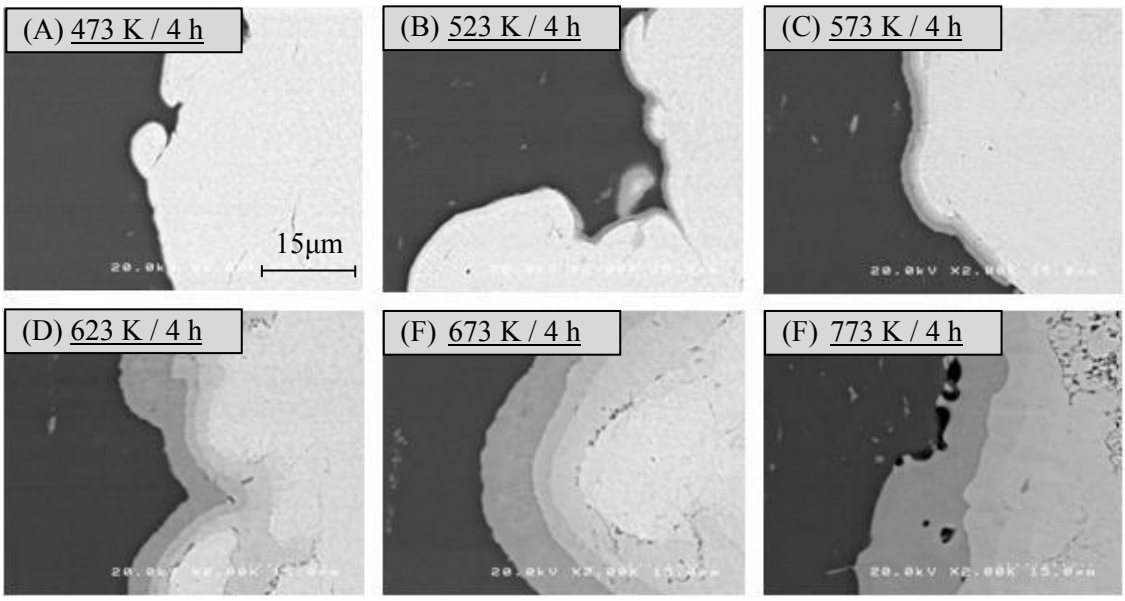


Figure 9. Growth process of the intermetallic compound at the cold-sprayed interface.

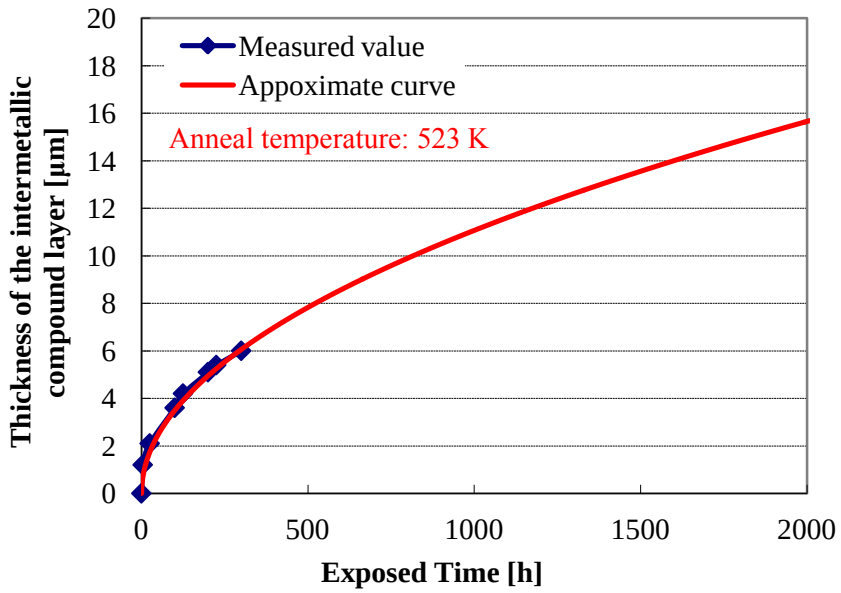


Figure 10. Growth curve of the intermetallic compounds at the cold-sprayed interface.

3.2. Mechanical properties

Figure 11 shows results of the three-point bending test of cold-sprayed copper on aluminum material. It was found that the deposited cold spray material was ruptured in the copper coating. However, if annealed at 623 K for 4 h, this material deformed plastically without fracturing. In addition, if annealed at 673 K for 4 h, the material ruptured at the copper-to-aluminum interface. It is conjectured that brittle fracture occurred by dislocations in the copper coatings without annealing. In the annealed materials, ductility was generated in the copper coatings by dislocation recovery at appropriate lower annealing temperatures, whereas interfacial fracture occurred due to the growth of intermetallic compounds by high temperature annealing.

In order to obtain further insight into the mechanical behavior, the cold-sprayed bonding material that was annealed at 623 K for 4 h was evaluated by tensile tests. The bonding interface was not ruptured and the breaking point was within the aluminum substrate, as shown in Figure 12. A mechanical strength of about 80 MPa was achieved. This breaking strength is different from the result shown in Figure 11 because of the different geometry between the bending and the tensile test.

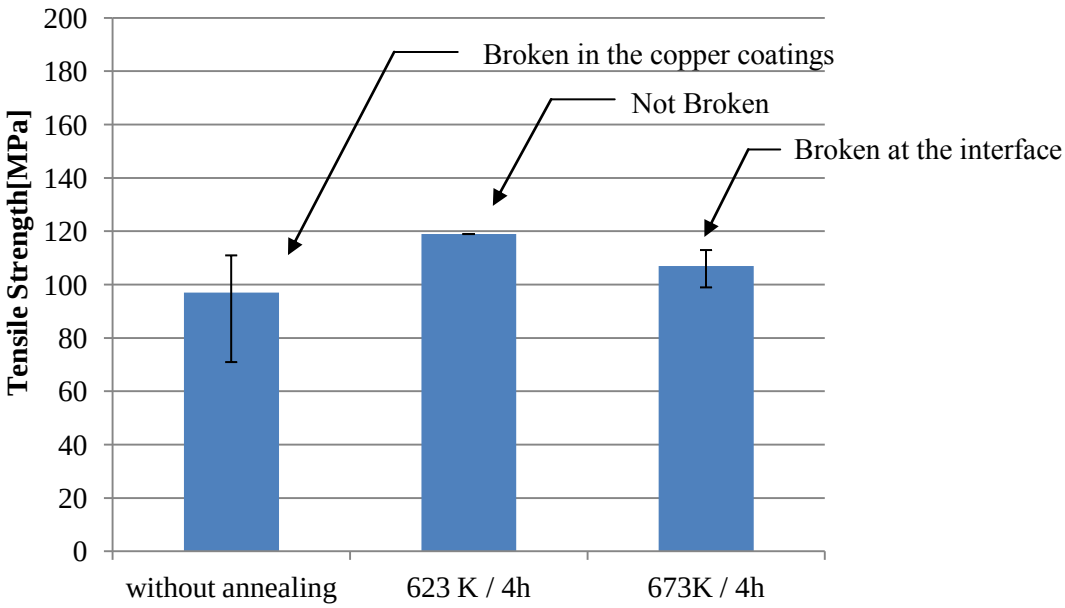
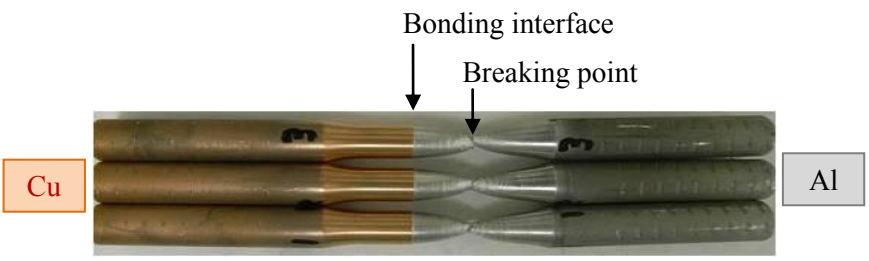


Figure 11. Test results of three-point bending tests.



Annealing condition: 623 K/4 h

Figure 12. Photo of a specimen after tensile test.

3.3. Electrical properties

The evaluation of the electric conductivity was carried out by using the four-terminal method and the result is shown in Figure 13. The cold-sprayed copper-aluminum materials have excellent electrical conductivity, close to theoretical bonding limit. This result evidences that the thin intermetallic layer at the bonding interface has no critical effect on electrical conductivity.

In addition, the effect of the various annealing conditions on the electrical characteristics of cold-sprayed copper-aluminum bonding materials was evaluated and the result is shown in Figure 14. The figure illustrates the conductivity changes due to annealing of the copper coatings and copper-aluminum bonding material. The conductivity without annealing was fixed at 1. Consequently, the conductivity increased on higher annealing temperature in the copper coatings until 673 K, but not further at 723K. This phenomenon can be explained by the recrystallization of copper. In addition, the

conductivity of the copper and aluminum bonding increased with higher annealing temperature. However, the increase rate is slightly smaller in the copper-aluminum junction than in the pure copper coatings because of the intermetallic layer grown at the interface between copper and aluminum. Note that the conductivity increased in the copper coatings by annealing.

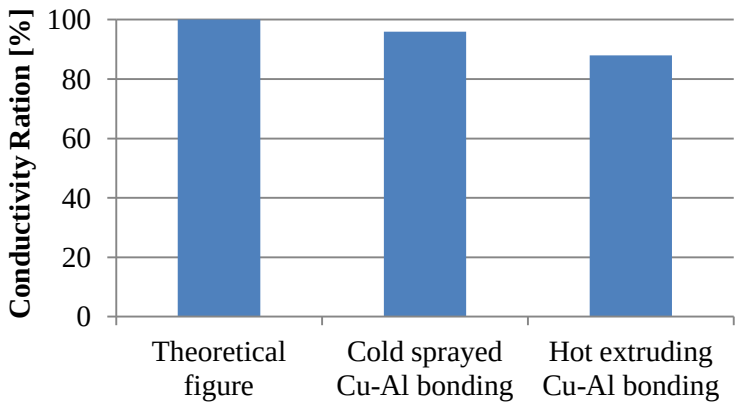


Figure 13. Comparison of the conductivities of cold-sprayed and hot-extruded Cu-Al to the theoretically expected conductivity.

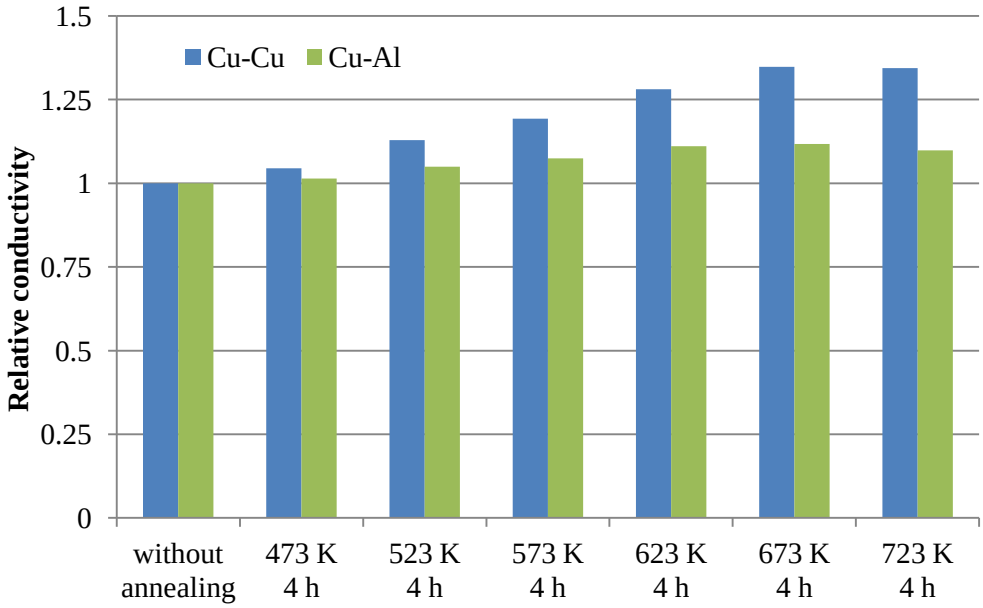


Figure 14. The conductivity changes induced by annealing.

4. Conclusions

In this study, the mechanical and electrical properties of cold-sprayed copper and aluminum bonding were evaluated with a focus on the interface and compared to those of the materials obtained by hot extruding. According to the experimental results obtained in this study, the following conclusions can be drawn:

- No critical intermetallic compound was observed in the cold-sprayed material, whereas a discontinuous intermetallic compound was observed at the hot-extruded interface.
- At the cold-sprayed interfaces, a continuous and uniform intermetallic layer was observed by TEM.
- It is estimated that the growth curve of the intermetallic layer is parabolic and that a good bonding interface is obtained.
- Using the cold spray process, superior mechanical and electrical properties can be achieved by appropriate annealing compared with the hot extrusion process.

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