Diffusion Bonding of Cu–Cu with Al–Ni Nano Multilayers

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The paper examines the joinability, microstructures and thermal behaviors of the Al–Ni nano multilayers. The results reveal the transitions and possible compositions of the nano multilayers when heating. Nanoscale Al–Ni multilayers could be used in Cu–Cu diffusion joining though the process and preparation method need to be optimized. [doi:10.2320/materials.MD201221]

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1. Introduction

Nano multilayers have excellent properties, such as super hardness, magnetic property, high thermal conductivity and low friction coefficient etc.¹ The Al–Ni nano multilayers have been investigated as a reactive nano material which could intensively release heat after an ignited self-propagation reaction. Therefore it has the potential to be used in micro-joining.² To joining copper and copper materials, the choice should be solder or brazing materials to adapt to the needs of the components where heating furnaces were commonly used. If using nano multilayers films as heating source or joining material, the joining process could be simplified and some low heat resistant component can be protected in the joining process. The Indium Company has developed a system to do joining with nanofoils, however the rate of running such a reaction in the system is difficult to control because of the large size of its components, poor contact or presence of oxide films on their surface, hence the technology has not been widely applied. This paper investigates the probability of joining copper with Al/Ni nano multilayer films and shows several critical points which are worth to be concern in future study.

2. Experiments

Nickel (Purity: 99.95%) and aluminum (Purity: 99.95%) were used as targets and to fabricate the Al/Ni multilayers film on copper base with high vacuum magnetron sputtering method (Type: JGP-560b, ShenYang scientific instruments company, The Chinese Academy of Sciences). After being successively cleaned by the ultrasonic with acetone and alcohol for 10 min respectively, the targets were mounted in the vacuum chamber and vacuum to 1 × 10⁻⁵ Pa. Then the atmosphere was filled with 24 sccm and 0.7 Pa argon gas. The Al and Ni were alternately sputtered to two copper bases for 3 and 10 cycles. The prepared samples were cut to different pieces for microstructure observation (SEM, Nova Nano 430, FEI, USA) and thermal analysis (STA409 PC, NETZSCH). For joining test the prepared sample with 3 alternations, without being stripped, is joined to a copper plate (20 × 20 × 1.5 mm³) in a furnace (500°C, 10 min) with gas protection under 10 MPa pressure. After that the joint was cut and measured by the SEM to analyze the microstructure and composition.

3. Results and Discussion

3.1 Microstructure observations

Figure 1 shows the micromorphology of the prepared sample. It can be seen that the surface of the nano-scaled film on the copper base is inclusion and void free (Fig. 1(a)), although the copper base surface is rough (Fig. 1(b)). It’s very important since the voids or other defects (crack, wrinkle and swell etc.) on the surface have hereditary in the reaction of the joining process.

Apparently the film has multilayer structure (Fig. 1(c)), where the bright fringes represent the nickel layer (purity 100% as measured). There were happened reactions between nickel and aluminum during the sputtering process, which forms the intermixed layers between the Ni and Al layers. It’s measured and calculated that the mean bilayer thickness (one alternation of Al and Ni) value of the film is 260 nm, and the mean thickness of the nickel layer and aluminum layers are 185 and 60 nm respectively. It is known that the atomic fraction of the reaction Ni–Al multilayers should be half by half to get pure AlNi intermetallic with high melting temperature (1638°C), consequently, the prepared nano multilayers with more nickel deposition will get a variety of intermetallics with lower melting temperature, such as Al₃Ni₁₅ (transit to AlNi₃ at 700°C) and AlNi₃ (1362°C). To join Cu–Cu interface,³ obviously the reaction products with melting temperature close to that of the copper (1084°C) is better.

3.2 Thermal analysis and reaction heat

Figure 2 shows the thermal analysis results by the differential scanning calorimeter (DSC). Under the 5°C/min heating ramp rate, an exothermic peak appears (Fig. 2(a)) and a weight growth (Fig. 2(c)) between 435.1 and 480.8°C where the self propagation reaction happens. The reaction temperature is lower than the lowest eutectic temperature (639.9°C) of the Al–Ni phase system. This result is similar to Hee Y. Kim’s observation⁴ who studied the synthesis of NiAl microfoils. This temperature range also supplies the temperature condition in joining copper with thermal igniting

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method. The weight growth is because of the oxidation or nitridation in the severe reaction. From Fig. 2(b), it can be seen that other 2 endothermic peaks (P1 and P2) appear between 631.7 and 700°C. P1 is probably due to the transition of Al3Ni5 \( \rightarrow \) Al3Ni5 (AlNi) + Al3Ni5 (AlNi3) solid solutions. It can be concluded that P2 represents the peritectoid transition of the P1 transition products to AlNi3 (AlNi) solid solution.

As known that J. Wang5) has developed a formula (1) to calculate the reaction heat of equal atomic ratio Al–Ni nano multilayers in self-propagating process.

\[
\Delta H = \Delta H_0 \left(1 - \frac{2w}{\lambda}\right). \tag{1}
\]

The \( \Delta H_0 \) is the maximum possible amount of heat that can be released, \( \lambda \) is the bilayer period, \( w \) represent the thickness of the intermixed layers. If inserting a parameter \( t \) into formula (1) to get formula (2) which includes the influence effect of the thickness of excessive Ni or Al, the reaction heat of the nickel/aluminum rich nano multilayers could also be measured and calculated as the following formula (2):

\[
\Delta H = \Delta H_0 \left(1 - \frac{2w + t}{\lambda}\right). \tag{2}
\]

By the formula (2) and the thermal analysis, the Ni–Al nano multilayers with controllable reaction heat for diffusion bonding could be designed, and some key conditions (such as the reaction temperature, the size of the filler multilayers and the needed load etc.) in the process of Cu–Cu diffusion bonding with the Ni–Al foils can be determined.

3.3 Nano joining process

Figure 3 reveals the cross section state of the micro joint. It indicates that the micro fusion zone successfully forms by the self-propagation reaction though the morphologies of the joint are not uniform (Figs. 3(a), 3(b)). The main elements in the bond are Al, Ni, Cu and O as measured by the EDX method (Fig. 3(c)). The mean bond width is 5.80 µm which is much thicker than the thickness of the Al–Ni nano multilayers (750 nm, see Fig. 1(c)); this convinces that the heat released from the reaction of the multilayers can join the Cu plates together. From Fig. 3(a) it can also be seen that a few parts of the bond don’t thoroughly react. The phenomenon could be attributed to the orientation result by the various contact condition of the copper plates. Probably the coated Cu plate has been protected by the pre-oxides of aluminum layer before the self-propagation happens. Some nano-scale Ni particles (by EDX measurement) could be found in Fig. 3(c) either, which shows the excess of Ni element in the reaction. This implies that the nickel layer should be a little bit thinner and more works need to get better multilayer for copper joining.
4. Conclusions

Al–Ni nano multilayer (the thickness of Al is thinner than that of Ni layer) can join the Cu–Cu through the reaction heat of self-propagation. The reaction temperature is between 435.1 and 480.8°C, the peritectoid transitions happen because of the atomic fraction of the aluminum is far lower than that of the nickel. The reaction heat of the nickel rich Al–Ni nano multilayers could be calculated and controlled to match the bonding process. The thickness of the Cu–Cu fusion zone is 7.7 times thicker than the Al–Ni nano multilayers though a few parts of the bond do not react. That is the reason of the residual nickel particles and demonstrates the room for improvement.

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REFERENCES


Fig. 3 Micrographs by SEM. (a) Bond with unreacted strips. (b) Fusion zone. (c) Residual nickel particles with EDX.