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Solid-state bonding between Al and Cu by vacuum hot pressing

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Abstract: Diffusion bonding between aluminum and copper was performed by vacuum hot pressing at temperatures between 623 and 923 K through two thermal processes: hot compression under the deformation rate of 0.2 mm/min for 10 min at pre-set temperatures, and additional pressing at 0.2 mm/min for 20 min during furnace cooling. After analyzing interface, the feasible diffusion bonding temperature was suggested as 823 K. The three major intermetallic layers generated during diffusion bonding process were identified as Al_2Cu, $AlCu+Al_3Cu_4$ and Al_4Cu_9. Furthermore, local hardness values of Al_2Cu, $AlCu+Al_3Cu_4$ and Al_4Cu_9 layers average at (4.97±0.05), (6.33±0.00) and (6.06±0.18) GPa, respectively.

Key words: vacuum hot pressing; diffusion bonding; Al−Cu intermetallic compound; composite interface; interface microstructures; nanoindentation

1 Introduction

The joining between dissimilar materials has attracted great attention indebted to their unique advantage for the viewpoint of multi-functionality. For example, Al−Cu bimetallic joint materials have their advantages due to their excellent properties combining low density, lightweight, corrosion resistance of aluminum, with extraordinarily high level of electrical and thermal conductivity of copper [1−8]. The major problem in the joining processes between Al and Cu is, however, the generation of complex Al−Cu intermetallic compounds (IMCs) at the interface at elevated temperatures around the liquidus of Al, which cause deleterious influence on both mechanical and electrical properties of the entire hybrid bimetallic composites. For the mechanical point of view, thermally induced brittle IMCs at interface between aluminum and copper reduce the overall ductility, though these IMCs give additional hardening to the interface [9]. Furthermore, it is well known that the interatomic bonds within IMCs between aluminum and copper reduce the number of available free electrons, resulting in higher electrical resistivities [10].

There were several studies on IMC generation during high temperature diffusion bonding between bulky copper and aluminum. It is well known that five equilibrium IMCs such as Al_2Cu (θ), AlCu (η_2), Al_3Cu_4 (ζ_2), Al_2Cu_3 (δ) and Al_4Cu_9 (γ_2) phases could be generated by diffusion bonding between Al and Cu at temperature near 773 K [11,12]. Recently, CHEN and HWANG [13] explained the diffusion mechanism indicating the stages in formation of interface structure in terms of formation energy of each IMC and diffusivity. At temperature intervals between 573 and 773 K, the sequence of diffusion mechanism can be described as follows [13]: 1) Saturated solid solution formed in Al-side at interface between Al and Cu; 2) Since the diffusion rate of aluminum into copper side is faster than that of copper into aluminum side, together with lower formation energy of Al_2Cu (0.78 eV) compared with that of Al_4Cu_9 (0.83 eV), Al_2Cu was presumed to form first followed by the generation of Al_4Cu_9; 3) Additional IMCs like AlCu and Al_3Cu_4 were formed after generation of previous two phases such as Al_2Cu and Al_4Cu_9.

However, there were several reports that aforementioned sequence of diffusion mechanism was varied with process temperature and time. LEE et al [10] found that there are only two IMC phases such as Al_2Cu and AlCu at the interface of the friction-welded Al−Cu

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joints for relatively short process time presumably due to the low diffusion rate of copper into aluminum. XUE et al [14] also verified that friction stir-welded joint between aluminum and copper under the fast moving speed of 100 mm/min was composed of Al_2Cu, Al_4Cu_9 and few AlCu particles [14]. In addition, it was found that joining between molten aluminum and solid copper wire by means of compound casting at 1003 K generated Al_2Cu, AlCu and Al_2Cu_3 IMCs at interface [15].

The aim of this work is to focus on the possibility of joining between aluminum and cooper by vacuum hot pressing at temperatures ranging from 623 to 923 K. Detailed microstructural investigation on the soundly diffusion bonded interfaces is then carried out in order to investigate the influence of process conditions on local mechanical properties.

2 Experimental

Materials used in this study were Al 1050 and oxygen-free high conductivity copper (OFHC, C101 in ASTM). Aluminum alloy was first cut as a cylindrical type. Copper alloy was then prepared as shell material for canning cylindrical aluminum. The exact dimensions are shown in Fig. 1(a). The vacuum hot pressing (VHP) was performed for diffusion bonding and the schematics is shown in Fig. 1(b). Initial stage of hot compression was done under a deformation rate of 0.2 mm/min at pre-set isothermal temperatures ranging between 623 and 923 K for 10 min within the atmosphere evacuated up to around 133.322×10^{-2} Pa followed by purging with purified argon gas. Then additional pressing was carried out under the same deformation rate of 0.2 mm/min after turning off the power of resistant furnace for 20 min.

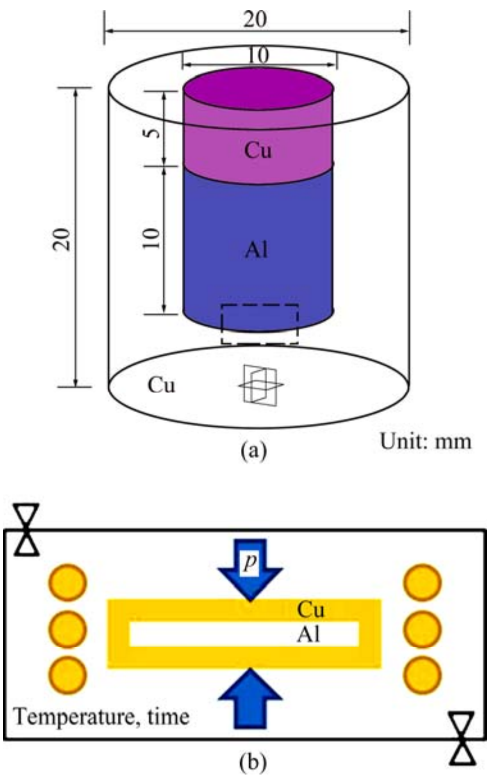

Fig. 1 Schematic diagram of sample dimension of aluminum/copper couple (a) and vacuum hot pressing used for solid-state diffusion bonding (b)

Joint samples were cut from the lower side of joint (marked as dot rectangular part in Fig. 1(a)) in order to characterize the interface microstructures. Optical microscope (OM), field-emission scanning electron microscope (FE-SEM, model TESKAN MIRA II) attached with Oxford energy dispersive X-ray spectrometer (EDS) and transmission electron microscope (TEM, model JEOL JEM−2100F) at 200 kV were applied to investigating microstructures at the Al−Cu interface. Nanoindentation measurement was also carried out on joint surfaces of the representative samples embedded in resin and polished to a mirror finish by utilizing an MTS nanoindenter XP equipped with a triangular Berkovich diamond indenter.

3 Results and discussion

Macroscopic diffusion bonding features at various vacuum hot pressing temperatures are shown in Fig. 2. It is apparent that low-temperature diffusion bonding below 773 K under the constant deformation rate results in insufficient joining between aluminum and copper. When the process temperature was increased above 873 K, no distinct lateral delamination exhibited at the interface between aluminum and copper. The thickness of diffusion-induced interfacial layers apparently increases with further increasing process temperature. It is interesting to note that the interface near the aluminum side is not smooth as the process temperature is above 898 K, presumably attributed to the drastic decrease of viscosity at semi-solid aluminum side.

For the sample 4 diffusion bonded at 823 K in Fig. 2, EDS analysis was carried out to investigate the exact composition of each IMC layer. Figure 3 exhibits the cross-sectional backscatter electron (BSE) image for sample 4 obtained by FE-SEM, where several IMCs can be distinguished by the color. Results of EDS analysis conducted on points *A*, *B*, *C*, *D* and *F* in Fig. 3 are then summarized in Table 1. The newly generated layers *B*, *C*, and *D* could be identified as Al_2Cu, AlCu, and Al_4Cu_9, respectively. The IMC phases were exactly consistent with other previous reports [12]. The averaged thicknesses of layers *B*, *C* and *D* were also evaluated as (6.32±0.31), (1.92±0.39) and (6.82±0.24) μm, respectively. On the other hand, the reason why other IMCs such as Al_2Cu_3 and Al_3Cu_4 could not be detected has to be

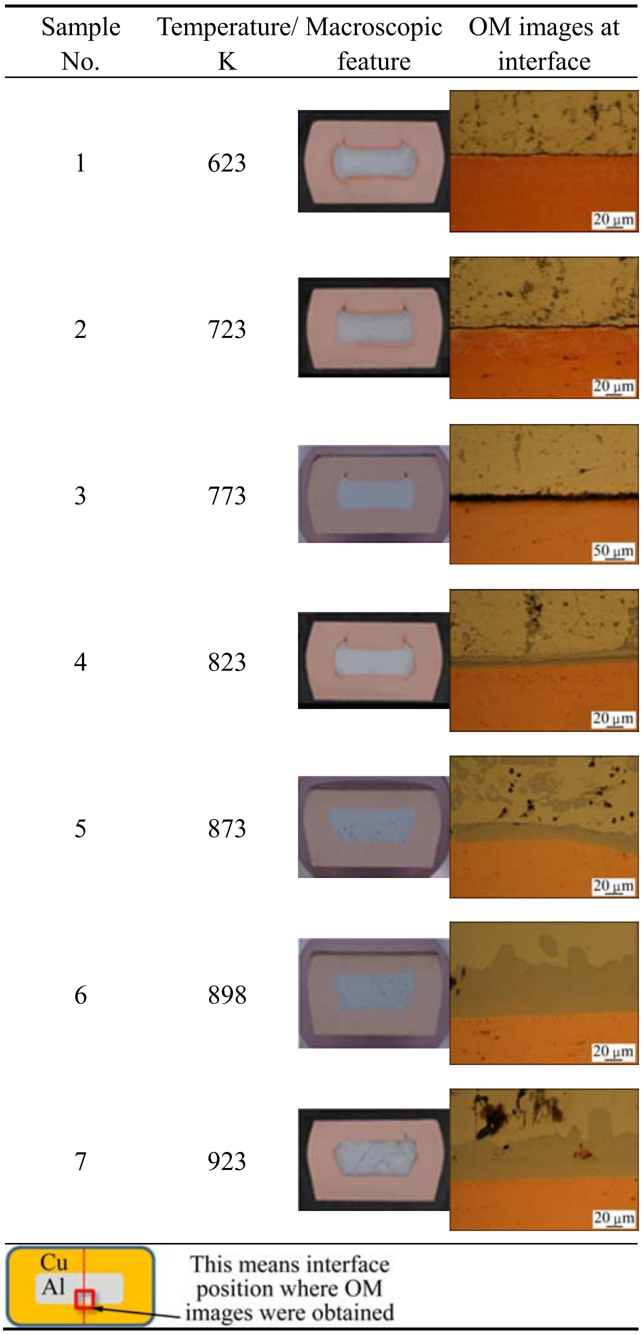

Fig. 2 Macroscopic features and optical micrographs of interfacial microstructures observed from diffusion bonded samples at different temperatures ranging from 623 to 923 K

further mentioned. Though one can assume that it is not possible for Al_2Cu_3 and Al_3Cu_4 IMCs to be generated for relatively short diffusion time at adopted temperature, it is also considered that the layer *C* is too thin to precisely resolve the composition at boundaries between newly generated IMC phases by means of typical EDS analysis.

After cutting sample at the interface between aluminum and copper joined at 823 K from dot rectangular region in Fig. 3(b) by means of focused ion beam, bright-field image was obtained by TEM. As shown in Fig. 4(b), interfacial layers are composed of three different colors implying distinctive IMC phases. It

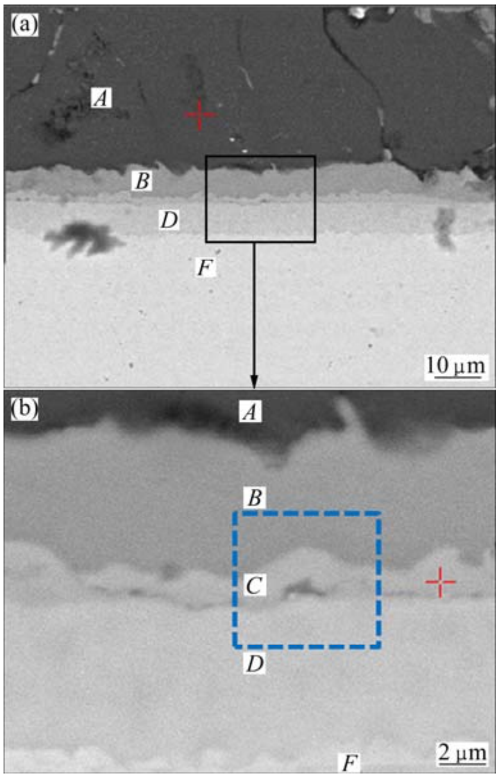

Fig. 3 FE-SEM images of interface obtained from Al−Cu joint of sample 4 in Fig. 2

Table 1 Results of EDS analysis on sample 4 diffusion bonded at 823 K

Point	x(Al)/%	x(Cu)/%	n(Al):n(Cu)
A	100	0	−
B	67.84	32.16	~ 2:1
C	52.34	47.66	~ 1:1
D	30.61	69.39	~ 4:9
F	0	100	−

is also apparent that VHP induced bonding at 823 K yields a continuous layer *C* with irregular thickness around 2 µm. Also elemental mapping of this Al−Cu macrocomposite shows that layers *B* and *D* are rich in aluminum and copper, respectively, as shown in Figs. 4(c) and (d).

In order to further investigate the characterization of narrow diffusion band, high resolution TEM observation was also carried out and the results are shown in Fig. 5. It is clear that the $Al_2Cu(\theta)$ phase was precipitated within aluminum-rich matrix on layer *B* shown in Fig. 5(a). From HRTEM image at the interface between layers *B* and *C* in Fig. 5(b), it was seen that layer *B* could be indexed as $Al_2Cu(\theta)$ phase which exhibited a tetragonal crystal structure along [011] direction determined by the fast Fourier transformed (FTT) diffraction pattern. On the other hand, the

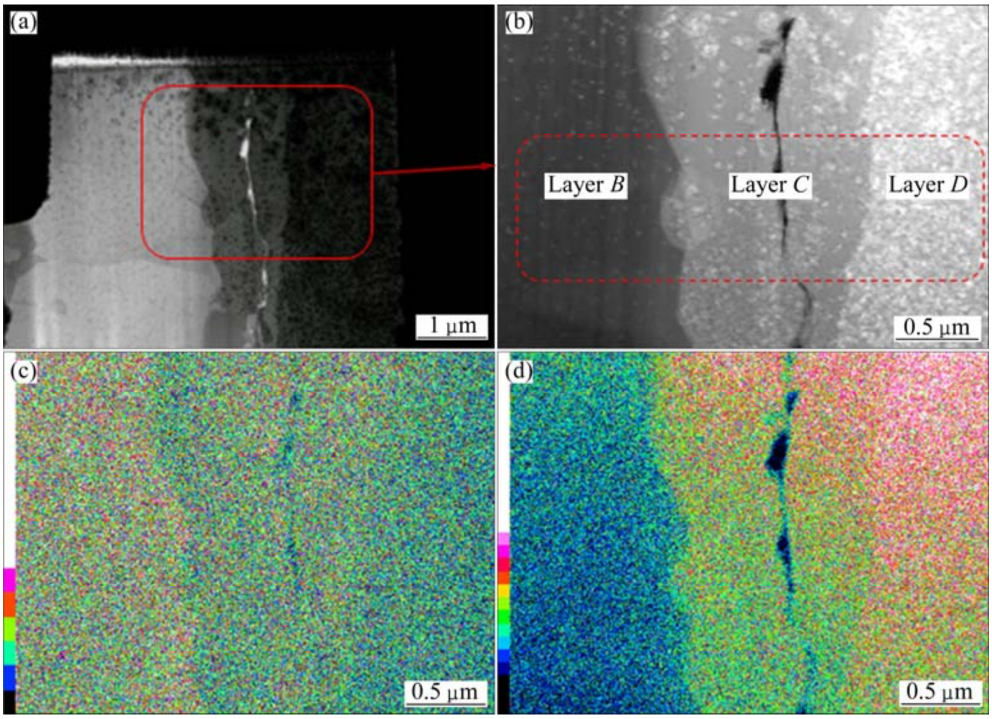

Fig. 4 SEM images of sample 4 made by focused ion beam (FIB) exhibiting three distinguished intermetallic compound layers (a), cross-sectional bright-field TEM image (b) obtained from rectangular box in (a), elemental mapping results of aluminum (c) and copper (d)

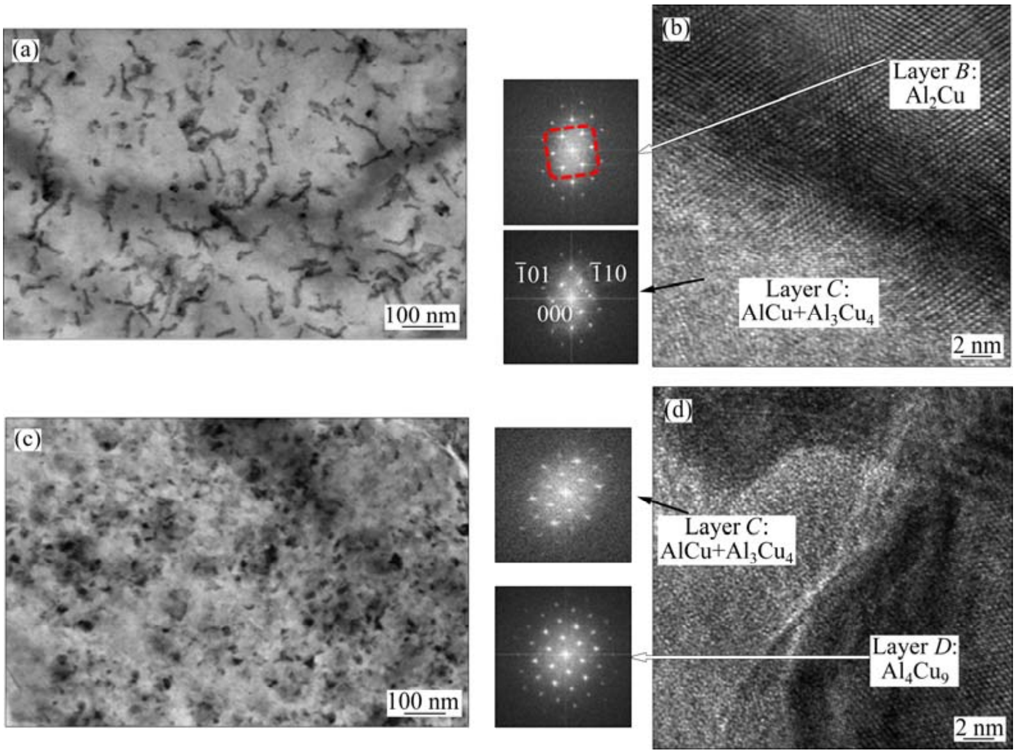

Fig. 5 TEM images of soundly diffusion bonded sample 4: (a) Bright field image at layer *B*; (b) HRTEM image at interface between layers *B* and *C*: (c) HRTEM image at interface between layers *C* and *D*; (d) Bright field TEM image at layer *D*

diffraction spots obtained from narrow region of thin layer *C* has a difficulty for identification. From the lower inlet of Fig. 5(b), main phase in layer *C* could be AlCu IMC since bright spot indicates typical orthorhombic crystal structure with *a*=1.004 nm, *b*=0.695 nm and *c*=0.416 nm along [111] direction. But other faint isolated diffraction spots were also seen in the FFT diffraction pattern from Fig. 5(b), which implied that another phase such as Al_3Cu_4 having orthorhombic structure with different lattice parameters was also generated during diffusion bonding. While there is a difficulty in indexing layer *C*, and it is easily confirmed from Figs. 5(c) and (d) that layer *D* consists of the precipitation of several nm-sized Al_4Cu_9 IMC phase within copper-rich solid solution. From FFT diffraction pattern, the Al_4Cu_9 phase could be identified as simple cubic structure along the [111] direction with the lattice parameters of a=b=c=0.87 nm.

Figure 6(a) illustrates a series of nanoindentation marks at the interface from sample 4 in Fig. 2, where nanoindentations with an array of 5×15 indents were performed under the displace-control mode with a depth limit of 150 nm. Furthermore the change of aluminum and copper contents along the arrow in Fig. 6(a) was overlapped onto the hardness values as a function of distance as seen in Fig. 6(b). After confirming serial number of indentation event conformed to the local mechanical property from each diffusion layer as well as parent materials, local hardness values of the layers A, B, C, D and F average at (1.58±0.32), (4.97±0.05), (6.33±0.00), (6.06±0.18) and (1.50±0.05) GPa, respectively. Noticeable higher hardness of the diffusion bonded interface than that of parent materials such as aluminum and copper confirms that layers B, C and D were high-hardness Al−Cu IMCs. Moreover, the hardness of the intermediate zone (layer C) is the highest among all of IMC layers, which means that AlCu+Al_3Cu_4 IMC layer could be the origin of brittle nature of the diffusion bonded Al−Cu macrocomposites.

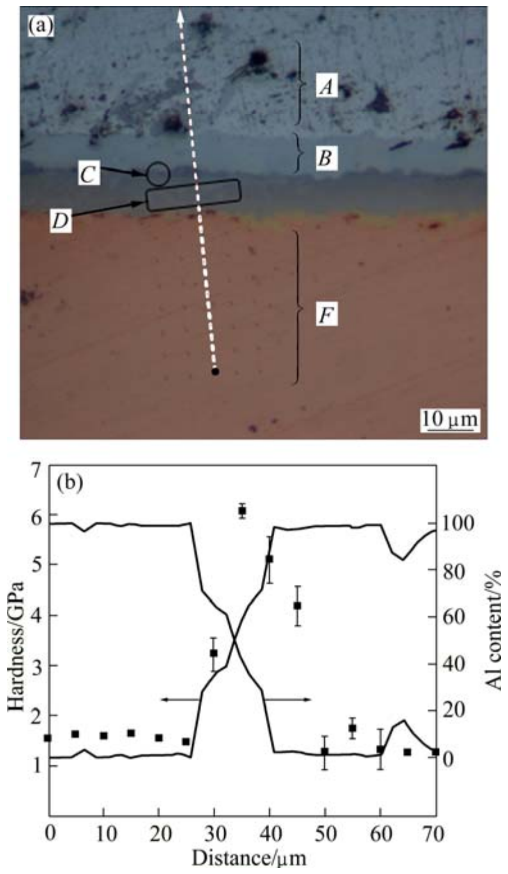

Fig. 6 SEM image of nanoindentation arrays obtained at interface between aluminum and copper (a), hardness variation (b) of Al/Cu composites overlapped onto evolution of Al−Cu contents through given arrow in (a)

4 Conclusions

1) Vacuum hot pressing between Al and Cu was carried out in the temperature ranging from 623 to 923 K for two distinguished compression steps. Feasible diffusion bonding temperature can be proposed as 823 K because low-temperature diffusion bonding below 773 K under the constant deformation rate exhibits lateral debonding between aluminum and copper.

2) The diffusion bonded interface between aluminum and copper at 823 K exhibits planar morphology composed of three distinguished layers. The layers adjacent to the aluminum and copper base metals are indexed as Al_2Cu and Al_4Cu_9, respectively. And the relatively thin middle layer is mainly composed of AlCu and Al_3Cu_4 intermetallic compounds.

3) Local hardness values of the aluminum, Al_2Cu, AlCu+Al_3Cu_4, Al_4Cu_9, and copper layers averages at (1.58±0.32), (4.97±0.05), (6.33±0.00), (6.06±0.18) and (1.50±0.05) GPa, respectively. The hardness values of intermetallic layers exhibit considerably higher than those of aluminum or copper base metals. The hardness of thin middle layer is the highest among diffusion layers, implying that AlCu+Al_3Cu_4 IMCs might induce brittle nature of the entire joint metals.

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真空热压铝和铜的固态连接

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摘　要：在温度 623~923 K 下采用真空热压扩散连接铝和铜，具体工艺为在预置温度下，变形率为 0.2 mm/min 时热压缩 10 min，再在炉冷过程中，以 0.2 mm/min 成型 10 min。通过界面分析可以看出，合适的扩散连接温度为 823 K，在扩散过程中产生了 3 种主要的金属间化合物层，分别为 Al_2Cu、$AlCu+Al_3Cu_4$ 和 Al_4Cu_9。3 种化合物层的局部硬度分别为(4.97±0.05)、(6.33±0.00)、(6.06±0.18) GPa。

关键词：真空热压；扩散结合；Al−Cu 金属间化合物；复合材料界面；界面微观结构；纳米压痕

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