Effects of Silica-Modified Aluminum on the Microstructure and Properties of Diamond/Al-Si Composites

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Abstract： To improve the wettability of the Diamond/Aluminum (Dia/Al) composites interface and prevent the excessive formation of Al4C3, this study used silicon-modified aluminum as the matrix. The composites with different silicon content in the matrix were fabricated by the vacuum-assisted pressure infiltration method, and the effects of silicon content on the microstructure and properties of the composites was systematically studied. The results showed that the Dia/Al-Si composites had a uniform and dense microstructure with good interfacial bonding. With the increase in silicon content, both the thermal conductivity and bending strength of the composites first increased and then decreased, while the coefficient of thermal expansion decreased monotonically. When the silicon content was 12 wt.%, the composites prepared with 180 μm diamond particles exhibited the highest thermal conductivity and bending strength, reaching 654 $W \cdot m^{-1} \cdot K^{-1}$ and 296 MPa, respectively, and the coefficient of thermal expansion was as low as $6.13 \times 10^{-6} \cdot K^{-1}$. This study indicates that the silicon-modified aluminum matrix can effectively suppress the excessive formation of Al4C3, and the coexisting of appropriate amount of Al4C3 or SiC at the interface enhances interfacial bonding, thereby improving the performance of the composites.

Keywords: thermal conductivity; electronic packaging; Silicon-modified aluminum; diamond/aluminum.

1 Introduction

Dia/Al composites with excellent performance have broad application prospects in the field of electronic packaging and heat dissipation. However, due to the poor wettability of diamond with aluminum, it is difficult to significantly increase the thermal conductivity of Dia/Al composites[1-3]. At sustained high temperatures, diamond and aluminum liquids are also prone to generate excessive amounts of the brittle and easily deliquescent phase Al_4C_3, which greatly deteriorates the interface and reduces the stability of the properties[2]. Aluminum matrix modification is an important method commonly used for interfacial modification of Dia/Al composites. Mizuuchi et al.[4] concluded that wettability can be promoted and interfacial bonding can be strengthened by aluminum matrix modification Beffort et al.[5] obtained composites with 7 wt.% Si-modified aluminum as a matrix with a thermal conductivity of only 130 $W \cdot m^{-1} \cdot K^{-1}$. Subsequently, Ruch[6] concluded that Si precipitated on the diamond surface hindered the generation of Al_4C_3 such that the interfacial bonding became weaker, and the thermal conductivity was reduced. In order to further enhance the thermal conductivity of Dia/Al composites, this paper systematically investigated the effect of Si content on the microstructure and properties of the composites by using silicon-modified aluminum as the matrix.

2 Experimental procedure

The Pure aluminum and silicon (Tianjin Zuoyuan New Materials Technology Co., China) and synthetic diamond single-crystal particles (Henan Huanghe Whirlwind Co., China) were used as the raw materials. The average particle sizes of the diamond are 180 μm. The Dia/Al composites were prepared by the vacuum-assisted pressure infiltration (VAPI) process. More information concerning preparation details of VAPIed Dia/Al composites can be found in paper[1].The microstructure was examined by field emission scanning electron microscopy (SEM, ZEISS Merlin Compact, Germany). Electrochemical etching was performed by using a direct current (DC) power supply in a 15vol.% HNO3 solution at an etching current of 2A. The thermal conductivities of composites were given by the equation: $\lambda = \alpha \times \rho \times Cp$. The thermal diffusivity (α) was measured using a laser flash apparatus (LFA467, Netzsch, Selb, Germany). Density (ρ) is given by the Archimedes method. The specific heat capacity Cp was derived by the mixture rule based on the mass fraction of each component. The CTE was test by using a dilatometer (DIL 402, Netzsch, Germany). The test was conducted over a temperature range from 25 °C to 200 °C. Bending strength was evaluated using a universal testing machine (MTS E45, MTS Systems Corporation, USA).

3 Result and discussion

Figure 1 shows the microstructure of Dia/Al-Si composite. Fig 2 shows the interfacial morphology of the composites. The microstructures of different composites are uniform and dense, with good interfacial bonding and a certain number of interfacial products distributed on the interface. With the increase of Si content, the interfacial products gradually increased and tended to be uniformly distributed, and their shapes gradually changed from point-like or rod-like to dendritic or coral-like. Fig 3 shows the EDS-SEM analysis of the interfacial products, indicating that the enrichment of silicon occurred on the surface of diamond particles during the preparation of the composites[5]. When the Si content in the aluminum matrix is low, the concentration of silicon atoms in the aluminum liquid is low and the driving force for diffusion is insufficient, so only a small amount of silicon is enriched at the interface. Whereas, when the Si content increases, the concentration of Si atoms in the aluminum liquid is higher, the driving force for diffusion is larger, and a large number of Si atoms are enriched at the interface and form dendrites.

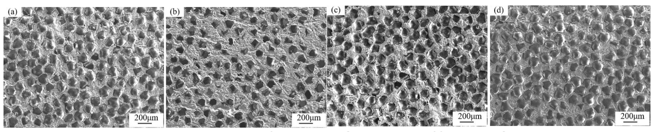

Fig. 1: Microstructure of Dia/Al-Si with different Si contents
(a) 4 wt.% ;(b) 8 wt.% ;(c) 12 wt.% ;(d) 16 wt.%
(b)

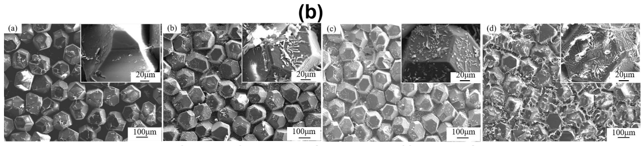

Fig. 2: Interface of Dia/Al-Si with different Si contents
(a) 4 wt.% ;(b) 8 wt.% ;(c) 12 wt.% ;(d) 16 wt.%
(b)

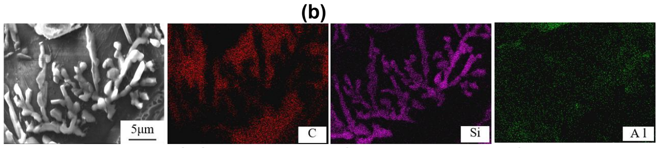

Fig. 3: EDS-SEM analysis of composites interfaces

Table 1 shows the properties of Dia/Al-Si composites. As the Si content in the aluminum matrix increases, both the bending strength (BS) and thermal conductivity (TC) of the composites tend to increase and then decrease, while the coefficient of thermal expansion (CTE) monotonically decreases. The former can be attributed to the changes in the interfacial structure in the composites[7]. When the Si content is low, the concentration of silicon atoms in the aluminum liquid is low and the driving force for diffusion is insufficient, the interfacial reaction is more difficult to carry out. The interfacial bonding is weaker, and at the same time, cracks are easier to germinate and extend, so the TC and BS of the composites are lower. With the increase of Si content, the concentration of silicon atoms increases, and it is easier to diffuse, the interfacial reaction is driven to form a metallurgical bond between the interfaces, and the interfacial strength increases, so the TC and BS are improved. However, when the Si content is too high, it leads to the formation of a thick brittle interfacial layer at the interface, which greatly reduces the interfacial bonding strength and increases the interfacial thermal resistance, so the TC and BS are reduced. The CTE of Al-Si alloys is lower with increasing Si content due to the decrease in the CTE of Al-Si alloys after alloying Al with Si and the transformation of tensile stresses into compressive stresses within the aluminum matrix[3].

Table 1. Properties of Diamond/Al-Si composites

Silicon content (*wt.*%)	TC ($W \cdot m^{-1} \cdot K^{-1}$)	BS (MPa)	CTE ($\times 10^{-6} \cdot K^{-1}$)
4.0	476	268	7.5
8.0	532	273	6.87
12.0	654	296	6.13
16.0	589	276	5.96

4 Conclusion

In summary, the silicon modified aluminum matrix can effectively promote interfacial wetting, reduce the generation of the side reaction Al_4C_3, and enhance the performance of Dia/Al-Si composites. The change of Si content leads to changes in the interfacial structure of the composites, but hardly affects their microstructure. With the increase of Si content in the aluminum matrix, the TC and BS of Dia/Al-Si composites show a tendency of increasing and then decreasing, while the CTE only monotonically decreases. When the Si content is 12 wt.%, the composites with 180 μm diamond exhibit excellent overall performance and meet the electronic packaging needs of typical semiconductor devices. In practical applications, composites with different properties can be prepared by regulating the alloy content to meet different application requirements.

5 Acknowledgments

The authors have no other competing interests to disclose.

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