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Effects of silicon addition on microstructure and properties of diamond/aluminum composites

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Abstract: Diamond/aluminum composites with excellent performance have broad application prospects in electronic packaging and heat dissipation. In order to improve the interfacial wettability between diamond and aluminum and to prevent excessive formation of Al_4C_3, silicon addition to the aluminum was used as the matrix. The composites with varying silicon content in the matrix were fabricated using the vacuum-assisted pressure infiltration method, and the effects of silicon content on microstructure and properties of the composites were systematically studied. Results show that the diamond/Al-Si composites exhibit a uniform and dense microstructure with good interfacial bonding. As the silicon content increases, both the thermal conductivity and bending strength of the composites first increase and then decrease, while the coefficient of thermal expansion decreases monotonically. When the silicon content in the aluminum matrix is 12wt.%, the composites reinforced with 180 μm diamond particles exhibit the highest thermal conductivity of 654 $W \cdot m^{-1} \cdot K^{-1}$, the highest bending strength of 296 MPa, and a remarkably low coefficient of thermal expansion of 5.96×10^{-6} K^{-1}. This study indicates that the silicon addition to the aluminum matrix can effectively suppress the excessive formation of Al_4C_3, and the coexistence of an appropriate amount of Al_4C_3 or SiC at the interface enhances interfacial bonding, thereby improving the performance of the composites.

Keywords: diamond/aluminum composites; thermal conductivity; electronic packaging; silicon-modified aluminum

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1 Introduction

The power consumption of electronic devices is increasing rapidly as they become more miniaturised, lightweight, and highly integrated[1, 2]. There is an urgent need to develop heat dissipation materials with excellent overall performance to meet the needs of high-power devices[3, 4]. Diamond is a more ideal material for heat dissipation due to its coefficient of thermal expansion (CTE) of as little as 1×10^{-6} K^{-1} and thermal conductivity (TC) of up to 2,200 $W \cdot m^{-1} \cdot K^{-1}$. However, diamond's difficulty in processing and its limited single-crystal size, as well as the poor match between coefficient of thermal expansion and semiconductor chips [$(3–7) \times 10^{-6}$ K^{-1}], limit its application in power devices[5, 6]. Diamond/aluminum composites have the advantages of high thermal conductivity, low coefficient of thermal expansion, and low density, etc., which can be used as electronic packaging substrates, heat sinks, and other materials in high heat flow density integrated circuits to meet the heat dissipation needs of high-power devices, and have a broad application prospect[7].

Due to the poor wettability of diamond and aluminum[4], it is difficult to achieve perfect bonding at the interface of diamond/aluminum composites. Therefore, the thermal conductivity of the composites is difficult to enhance significantly. In addition, diamond is also prone to interfacial reactions with the aluminum solution to produce Al_4C_3[7, 8], which is a brittle and easily deliquescent phase. Although a moderate amount of Al_4C_3 is beneficial to promote interfacial wetting, it is easy to generate excessive Al_4C_3 under sustained high-temperature environments, which greatly reduces the stability of the composite's properties when it undergoes deliquescence[9]. Aluminum matrix

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modification is an important method commonly used to modify the interface of diamond/aluminum composites[10, 11]. It is also known as aluminum matrix alloying, by adding a certain amount of alloying elements to aluminum to improve the properties of the aluminum matrix[1]. Beffort et al.[12] found that silicon (Si) enrichment at the diamond-aluminum interface effectively suppressed excess formation of Al_4C_3 when composites were prepared using an aluminum matrix modified with 7wt.% (mass percentage, hereafter wt.%) Si. However, the resulting thermal conductivity of the composites was only 130 $W \cdot m^{-1} \cdot K^{-1}$. Subsequently, Ruch[13] discovered the phenomenon of "selective bonding" based on Beffort's results, which suggested that the Si precipitated on the diamond surface hindered the generation of $A1_4C_3$, resulting in weaker interfacial bonding and lower thermal conductivity. Mizuuchi et al.[14] produced diamond/aluminum composites with a thermal conductivity of 403 $W \cdot m^{-1} \cdot K^{-1}$ by modifying the aluminum matrix with 5wt.% Mg. Subsequently, they switched to 7wt.% Si-modified aluminum matrix to further increase the thermal conductivity of the composites to 581 $W \cdot m^{-1} \cdot K^{-1}$, indicating that Si was beneficial in improving interfacial bonding. Wu et al.[15] found that the thermal conductivity of the composites increased from 210 to 330 $W \cdot m^{-1} \cdot K^{-1}$ as the Cu content was increased from 0 to 3wt.% when diamond/Al composites were prepared by using Cu-modified aluminum as the matrix. They concluded that Cu can lower the melting point of aluminum and improve the wettability by reacting with aluminum to produce Al_2Cu. In conclusion, the wettability of aluminum with diamond can be enhanced by aluminum matrix modification, which can inhibit the excess Al_4C_3 generation and strengthen the interfacial bonding[16]. However, the TC of composites prepared by aluminium matrix modification is generally low (up to 581 $W \cdot m^{-1} \cdot K^{-1}$ only)[14].

There is a lack of systematic research on the composites modified by aluminum matrix, and the influence of alloying elements on their microstructure and properties is unclear. Furthermore, the effect of factors such as the size of the diamond particles on thermal conductivity has not been considered. It is reported in existing works that the SiC interfacial layer exhibits relatively prominent interfacial thermal conductivity among numerous materials used for diamond/aluminum composites interfaces[17]. Furthermore, aluminum-silicon (Al-Si) alloys possess advantages such as low cost, excellent fluidity, and favorable casting properties within the aluminum alloy family. Therefore, in this study, Al-Si alloys with different Si contents were formulated by modifying the aluminum matrix in the form of adding pure silicon to pure aluminum. Diamond/Al-Si composites with different Si contents were prepared by a vacuum-assisted pressure infiltration process. The effect of Si content on the microstructure and properties of the composites was systematically investigated.

2 Materials and methods

Synthetic single-crystal diamond particles (Henan Huanghe Whirlwind Co., China) with four different average sizes (45, 106, 180, and 425 μm) and self-formulated Al-Si alloys were used as raw materials. The Al-Si alloys with various Si contents were prepared by co-melting pure Al blocks and pure silicon blocks in a melting furnace. The diamond/Al composites (mass fraction of diamond about 60wt.%). The thermal conductivity of diamond particles can be estimated to be approximately 1,600 $W \cdot m^{-1} \cdot K^{-1}$ based on their nitrogen content [N] (about 180 ppm) by the equation $\lambda = 2200 - 3.27[N]$[2]. The Al-Si alloy was formulated proportionally with 99.8% pure aluminum and pure silicon (Tianjin Zuoyuan New Material Technology Co., China) as raw materials. The diamond/Al composites (mass fraction of diamond about 60wt.%) were prepared by a self-developed vacuum-assisted pressure infiltration method. Specifically, diamond particles with the above four sizes were used as reinforcements to fabricate diamond/Al-12Si composites with varied particle sizes via the same process, the diamond preform was first pre-heated in a muffle furnace at 600 °C for 1 h. Subsequently, the Al-Si alloy was melted, with the molten aluminum reaching a temperature of 800 °C. The molten metal was then poured into the mold while simultaneously activating the vacuum equipment. Finally, under a mechanical pressure of 8 MPa, the molten aluminum infiltrated into the diamond particles, with the infiltration process lasting approximately 1 min. More details about this preparation process have been reported in our previous work[4].

The microstructure of the composites was characterized by field emission scanning electron microscopy (SEM, ZEISS Merlin Compact, Germany) equipped with an X-ray energy spectrometer. The surface of the composites was etched using the electrochemical etching method with a DC stabilized power supply. The electrolyte used was a 15vol.% HNO_3 solution, the etching current was set to 2 A, and the etching time was approximately 2 min. Following etching, the samples were cleaned in anhydrous ethanol. The physical phase analysis of the composites was carried out by X-ray diffractometer (XRD, Rigaku S2, Japan). The thermal diffusion coefficient (α) of the composites was measured with a laser flash analyser (LFA 467, Netzsch, Selb, Germany). The density (ρ) of the composites was measured using the Archimedean drainage method. The specific heat capacity (C_p) of the composites can be calculated by the law of mixtures. The thermal conductivity (λ) of the composites was calculated according to the equation $\lambda = \alpha \times \rho \times C_p$. The thermal conductivity was measured with a sample size of Φ12.7 mm×3 mm at 25±5 °C. The coefficient of thermal expansion of the composites was measured by a thermal expansion meter (DIL 402, Netzsch, Germany) with a sample size of Φ6 mm×25 mm at a temperature ranging from 25 °C to 200 °C. An electronic universal testing machine (MTS E45, MTS Systems Corporation, USA) was used to test the bending strength of the composites. The specimen size was 3 mm×4 mm×38 mm, the span was 32 mm, the upper indenter was 5 mm, the loading rate was 0.3 $mm \cdot min^{-1}$, and 10 data points were collected per second. Four parallel specimens were used for the above tests and the results were averaged to minimize the testing error.

3 Results and discussion

3.1 Microstructures of diamond/Al-Si composites

SEM images in Fig. 1 show the microstructure of diamond/Al-Si composites with different silicon contents after sandpaper polishing. It can be seen that different diamond/Al-Si composites exhibit similar microstructure morphology. The diamond particles are uniformly distributed throughout the aluminium alloy matrix, with no obvious clustering or particle bias.

Further observation reveals that the aluminum alloy completely and uniformly fills the interstitial spaces between the diamond particles, and the diamond particles are firmly embedded in the aluminum alloy matrix, with good interfacial bonding between the two. The microstructure of the composites is dense, and there are no microstructural defects such as shrinkage porosity and shrinkage holes. The above characteristics show that the content of modified element Si hardly affects the distribution of diamond particles in the microstructure of the composites after the modification of the aluminum matrix.

Owing to the high hardness of diamond, it is difficult to prepare metallographic samples for observing the

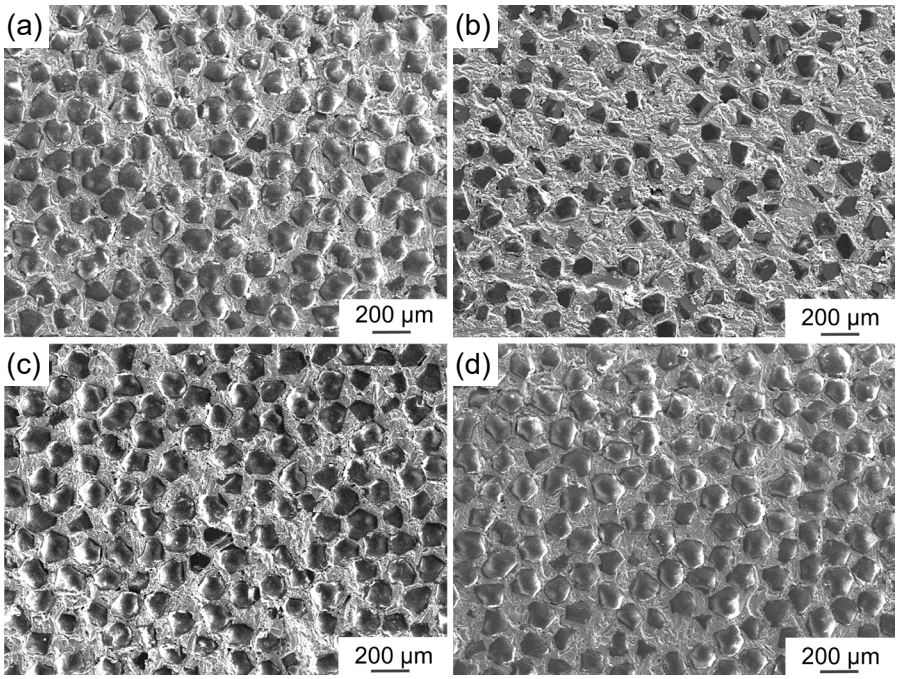


Fig. 1: Microstructure of diamond/Al-Si composites with different Si contents: (a) 4wt.%; (b) 8wt.%; (c) 12wt.%; (d) 16wt.%

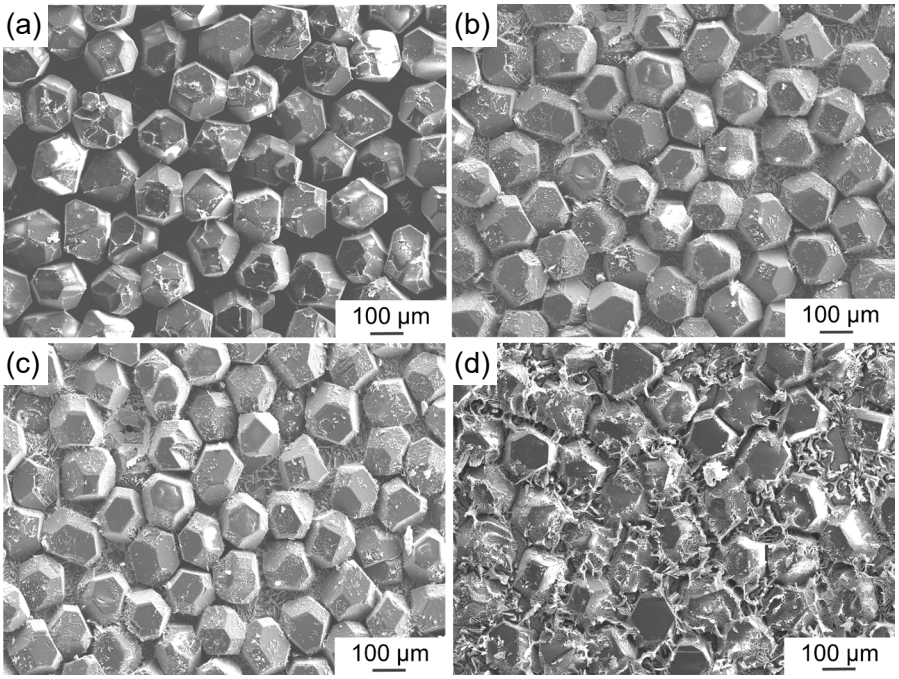


Fig. 2: Microstructure morphology of diamond/Al-Si composites with different Si contents after electrolytic etching: (a) 4wt.%; (b) 8wt.%; (c) 12wt.%; (d) 16wt.%

microstructure of composites using conventional grinding and polishing methods. Therefore, electrolytic etching was employed to selectively remove the aluminum alloy matrix, thereby clearly revealing the interfacial microstructure and distribution of diamond particles in the composites. The SEM images of diamond/Al-Si composites with different silicon contents after electrochemical etching are shown in Fig. 2. As can be seen in Fig. 2, the microstructure of different diamond/Al-Si composites is similar, and the diamond is uniformly distributed in different aluminum matrices. In particular, a certain number of interfacial products are also distributed on the surface of the diamond particles in the composites, which gradually increase with the increase of Si content in the aluminum matrix. The above characteristics indicate that the vacuum-assisted pressure infiltration method proposed in this study can prepare high-quality diamond/Al-Si composites with uniform and dense microstructure and good interfacial bonding.

Figure 3 indicates the SEM images of the electrolytically etched interfacial morphology in diamond/Al-Si composites with varying silicon contents. It can be seen that the electrolytic etching interfaces of different composites exhibit distinct morphological characteristics, while interface products are consistently present on the surface of diamond particles. Further observation reveals that the interfacial products on the surface of diamond particles gradually increase with the increase of Si content in the aluminum matrix. When the Si content in the aluminum matrix is as low as 4wt.%, the composites' electrolytic etching interface is only sporadically distributed with point-like or rod-like interface products. As the Si content in the aluminum matrix increases from 4wt.% to 12wt.%, the interfacial products on the surface of the diamond particles gradually increase and tend to be uniformly distributed, and the shape gradually changes from point- or rod-like to dendritic. When the Si content in the aluminum matrix is further increased to 16wt.%, the interfacial reaction products gradually increase in amount and begin to connect into larger regions, with the morphology evolving toward dendritic or coral-like structures.

The excessive interfacial product on the electrolytic etching interface of the composites is found to be mainly composed of three elements, C, Si, and Al, by EDS analysis, and it is initially determined that the products may be Si or SiC. As shown in Fig. 4, the higher content of Si indicates that Si atoms in the aluminum matrix are enriched on the surface of the diamond particles in the preparation of the composites[12]. When the Si content in the aluminum matrix is low, the Si atoms in the aluminum alloy liquid exhibit a lower driving force for diffusion due to the lower concentration of Si atoms. Therefore, only a small amount of Si is enriched on the surface of the diamond particles in the composites. While when the Si content in the aluminum matrix increases, the Si atom concentration in the aluminum alloy liquid is higher, and the driving force for the diffusion of Si atoms is larger. As Si atoms continuously enrich at the interface of the composites, they begin to nucleate and grow into dendritic structures during the cooling stage of composite fabrication.This indicates that the addition of an appropriate amount of Si (12wt.% in this study) to the aluminum matrix can promote interfacial wetting and improve the interfacial bond strength.

The XRD patterns of diamond/Al-12Si composites after electrochemical etching are shown in Fig. 5. Small amounts of Al_4C_3 and SiC are detected in addition to diamond, Si, and Al, further confirming that the interfacial reaction between diamond and aluminum matrix can produce interfacial products, as shown in Fig. 6. Actually, studies have shown that adding silicon to aluminum can effectively suppress the formation of Al_4C_3 by

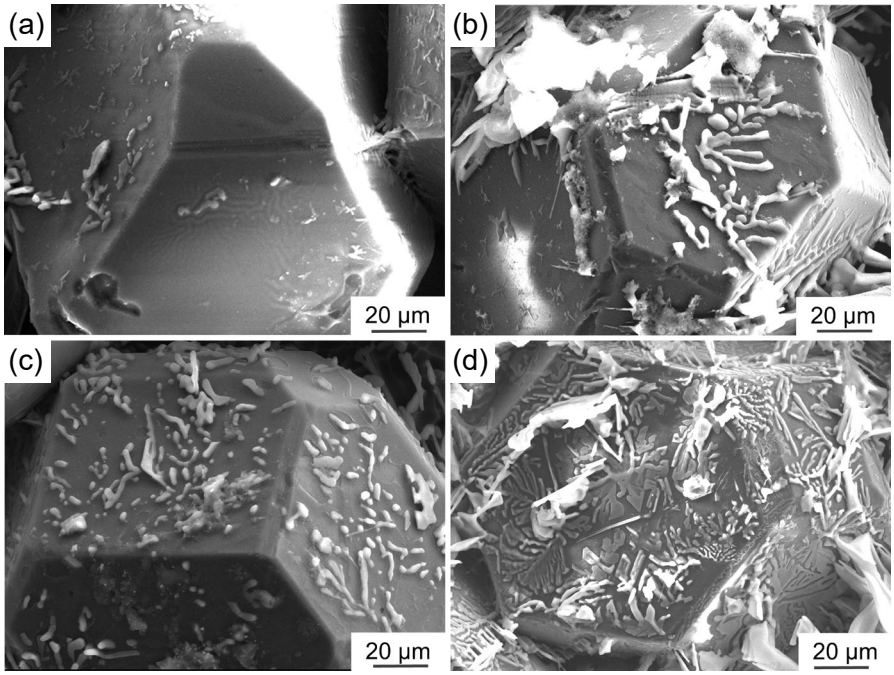


Fig. 3: Morphologies of electrolytically etched interface of diamond/Al-Si composites with different Si contents: (a) 4wt.%; (b) 8wt.%; (c) 12wt.%; (d) 16wt.%

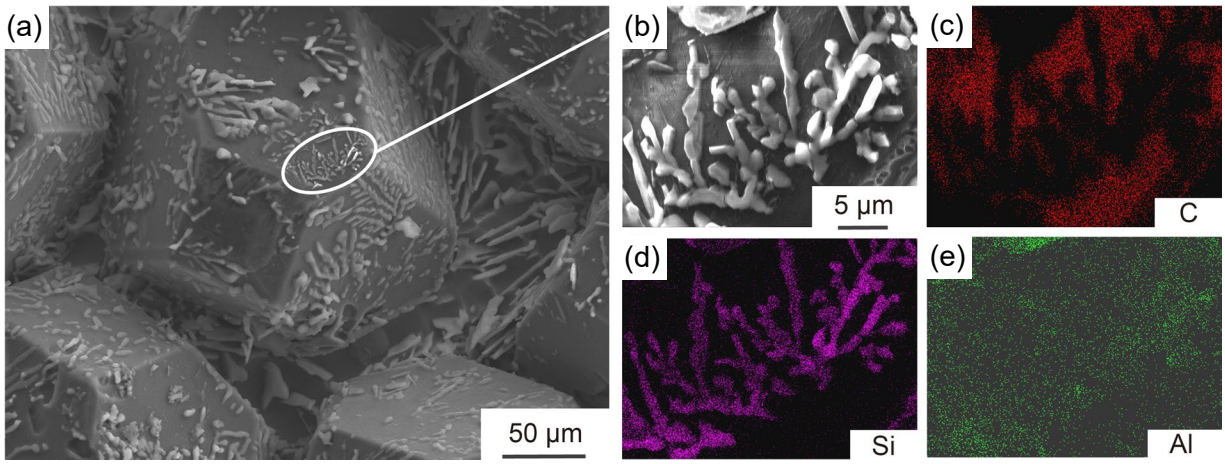


Fig. 4: Energy spectrum analysis of interfacial products of diamond/Al-12Si composite (a–e)

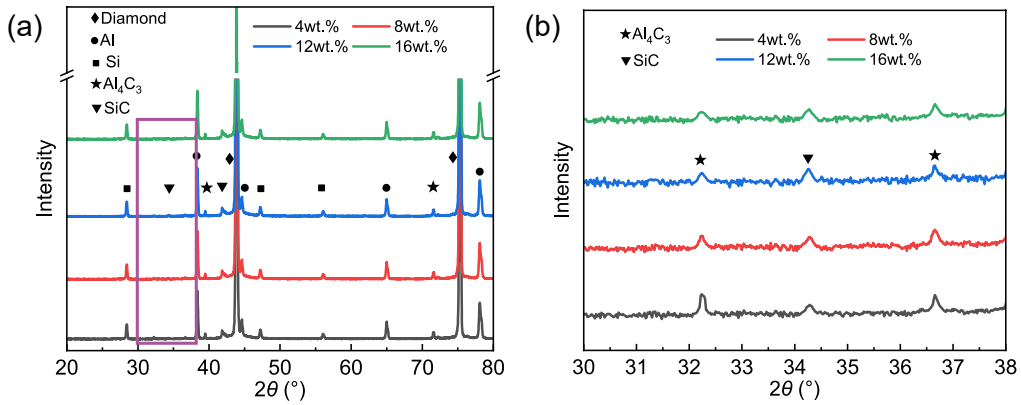


Fig. 5: XRD patterns of diamond/Al-12Si composite after electrolytic etching (a, b), (b) is magnified image of purple box region in (a)

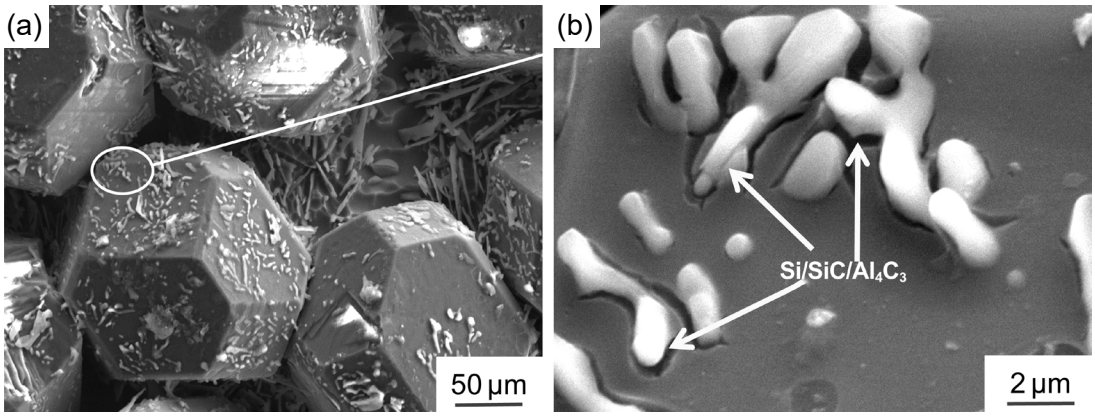


Fig. 6: Interfacial morphology of diamond/Al-12Si composite after electrolytic etching (a, b)

favoring the formation of SiC according to Eq. (1)[8]. However, it is also possible that some pure silicon phases may form through subsequent reactions according to Eq. (2)[18]. According to the elemental distribution map of the diamond/Al-12Si composites after electrochemical etching, as shown in Fig. 4, the gray-white phase primarily consists of Si, C, and Al (with Si accounting for a higher proportion), which is consistent with the EDS elemental mapping (Fig. 4) and XRD results (Fig. 5) confirming the coexistence of SiC, Al_4C_3, and residual Si phases at the interface.

$$4Al + 4C + Si \rightleftharpoons Al_4C_3 + SiC \quad (1)$$

$$4Al + 3SiC \rightleftharpoons 3Si + Al_4C_3 \quad (2)$$

Since the composites' surface contains hardly any aluminum matrix after electrochemical etching, it can be inferred that Al exists as the compound Al_4C_3. while Si appears in two forms: elemental Si and the compound SiC. In fact, the results in Fig. 5 corroborate this interpretation. Furthermore, by observing Fig. 6 (which shows a different location within the same field of view as Fig. 4), it can be seen that the gray-white phase exhibits dendritic (dendritic Si) and thin hexagonal plate-like morphologies (Al_4C_3)[19]. Therefore, it can be inferred that the gray-white phase in Fig. 6 is a mixture of elemental Si, SiC, and Al_4C_3. The interfacial reaction products are firmly embedded in the diamond

particles by atomic interdiffusion, which can significantly improve the interfacial bond strength between diamond and aluminum matrix. In the composites, the interfacial reaction products as a transition layer bridge the gap between diamond and aluminum matrix, and this structure significantly improves their interfacial bonding strength, prevents the formation of interfacial gaps, and enhances the densification of the composites. During the vacuum-assisted pressure infiltration process, suitable high-temperature and high-pressure conditions provide the required kinetic and thermodynamic conditions for the interfacial reaction, overcoming the challenge of poor wettability between the diamond and the aluminum melt[20–22]. It is thanks to this suitable preparation environment that the diamond/Al composites thus exhibit excellent microstructure and unique interfacial structure.

3.2 Thermal performance of diamond/Al-Si composites

Figure 7(a) shows the thermal conductivity of diamond/Al-Si composites with different Si contents. The thermal conductivity of the composites tends to increase firstly and then decrease with the increase of Si content in the aluminum matrix. The thermal conductivity of the composites increases from 476 $W \cdot m^{-1} \cdot K^{-1}$ to 654 $W \cdot m^{-1} \cdot K^{-1}$ when the Si content is increased from 4wt.% to 12wt.%. However, when the Si content is further increased to 16wt.%, the thermal conductivity of the composites decreases to 589 $W \cdot m^{-1} \cdot K^{-1}$, which indicates that the effect of the Si-modified aluminum matrix on the thermal conductivity of diamond/Al-Si composites is great. Notably, the thermal conductivity of the composites can be as high as 654 $W \cdot m^{-1} \cdot K^{-1}$ when the Si content is 12wt.%, indicating that there exists an optimum Si content for diamond/Al-Si composites, and that the composites prepared with this content of Si-modified aluminum matrix can obtain the highest thermal conductivity. The thermal conductivity of diamond/Al-12Si composites with different particle sizes is shown in Table 1. In particular, the composites prepared using diamond particles of larger size (425 μm) and also containing 12wt.% Si, the thermal conductivity is up to 713 $W \cdot m^{-1} \cdot K^{-1}$, which is much higher than the highest thermal conductivity value of the composites prepared by the aluminum matrix modification method reported in similar literature[14]. It indicates that the thermal conductivity of the composites can be effectively improved by simultaneously considering Si modification of the aluminum matrix and increasing the diamond particle size[23].

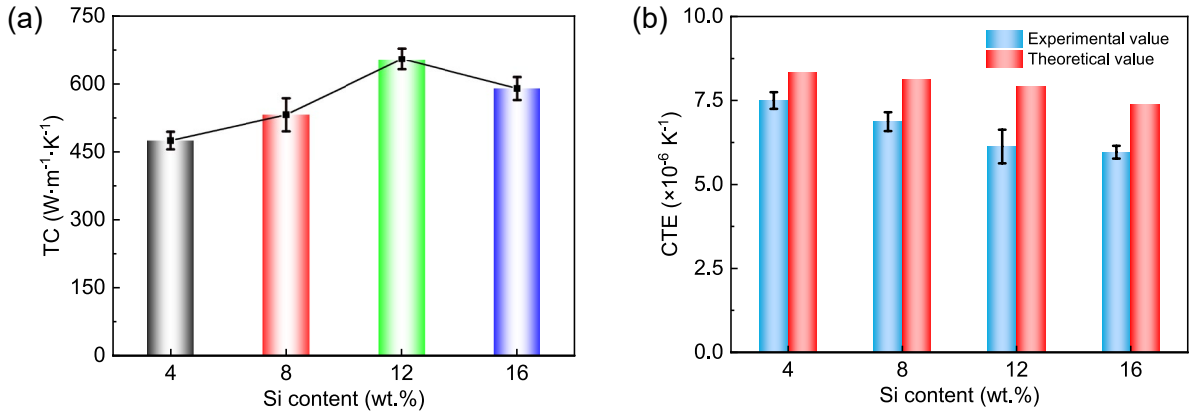


Fig. 7: Thermal performance of diamond/Al-Si composites: (a) thermal conductivity (TC); (b) coefficient of thermal expansion (CTE)

Table 1: Thermal conductivity of diamond/Al-12Si composites with different particle sizes

Diamond particle size (μm)	Thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$)
45	455
106	507
180	654
425	713

The change in the interfacial structure between the diamond particles and the aluminum matrix in the composites can be attributed to the change in the thermal conductivity of the diamond/Al-Si composites with varying Si content. Si atoms do not have enough driving force for diffusion due to their low concentration in the aluminum alloy liquid when the Si content in the aluminum matrix is low. At this time, only a small amount of Si atoms are enriched in the interface of the composites, the interfacial reaction is more difficult to carry out, and the interfacial bonding is also weak, therefore, the thermal conductivity of the composites is low[24, 25]. As the Si content in the aluminum matrix rises, the Si concentration in the aluminum alloy liquid rises, and the driving force of Si atom diffusion increases. A large number of Si atoms are continuously enriched toward the interface of the composites and undergo interfacial reaction with diamond to generate SiC, which enhances the interfacial bonding and improves the interfacial thermal conductivity[25]. At the same time, the scattering effect of hot carriers (phonons) is weakened, so the thermal conductivity of diamond/Al-Si composites is enhanced. However, a large number of Si atoms diffuse to the interface of the composites when there is too much Si in the aluminum matrix. It is easy to form a thicker interfacial layer

at the interface, so that the interfacial thermal resistance of the composites increases and the thermal conductivity decreases. When increasing the diamond particle size, the composites exhibit a high thermal conductivity due to the strong interfacial bonding, large diamond particle size, and high interfacial thermal conductivity of the joint role. The relatively small specific surface area of the larger diamond size compared to the smaller diamond size reduces the interfacial area of the composites and lowers the total interfacial thermal resistance, thus improving the efficiency of heat flow transfer[26-28]. Furthermore, the strong interfacial bonding, can be attributed to the strong anchoring and bridging effect of the appropriate amount of interfacial products, Al_4C_3 or SiC, present between the interfaces.

The coefficient of thermal expansion was predicted by a theoretical model to enable a thorough analysis of the thermal expansion properties of diamond/Al-Si composites after aluminium matrix modification. The Kerner's model is more accurate in predicting the results because it takes into account both the microstresses of the composites and the effect of crystal boundary shear stresses in the commonly used theoretical model[1, 4, 5]. Therefore, in this study, the Kerner's model was used to predict the coefficient of thermal expansion of diamond/Al-Si composites according to the following equation:

$$\alpha_c = \alpha_m V_m + \alpha_d V_d + \frac{V_m V_d (\alpha_d - \alpha_m) \times (K_d - K_m)}{V_m K_m + V_d K_d + (3 K_m K_d / 4 G_m)} \quad (3)$$

where the α_c, α_m, and α_d refer to the coefficient of thermal expansion of the composites, aluminum, and diamond, respectively, $\times 10^{-6}$ K^{-1}. The V_m and V_d are the volume fractions of aluminum and diamond, respectively, vol.%. The K_m and K_d are the bulk modulus of aluminum and diamond, respectively, in GPa. The G_m is the shear modulus of aluminum. The material parameters required for the calculations are listed in Table 2, where the relevant parameters of Al-Si alloys are measured experimentally.

The coefficient of thermal expansion of diamond/Al-Si composites with different silicon contents is shown in Fig. 7(b). It can be seen that the coefficient of thermal expansion of diamond/Al-Si composites shows a decreasing trend with the increase of Si content in the aluminum matrix, which is the same trend as the predicted value of Kerner's model. This may be related to the fact that the microstress and shear stress of the composites are considered in the Kerner's model, which indicates that the Kerner's model can predict the coefficient of thermal expansion of the composites more accurately. When the Si content in the aluminum matrix was increased from 4wt.% to 16wt.%, the coefficient of thermal expansion of diamond/Al-Si composites first decreases rapidly from 7.5×10^{-6} K^{-1} to 6.13×10^{-6} K^{-1}. Subsequently, it decreases slowly to 5.96×10^{-6} K^{-1}, which is in agreement with the typical semiconductor coefficient of thermal expansion of $(3-7)\times10^{-6}$ K^{-1}[7]. This indicates that diamond/Al-Si composites can match typical semiconductor devices well. According to Kerner's model, it is known that the coefficient of thermal expansion of diamond/Al-Si composites decreases as the volume fraction of diamond increases or the coefficient of thermal expansion of the aluminum matrix decreases.

Table 2: Properties of diamond particles and Al-Si alloys

Material	α ($\times10^{-6}$ K^{-1})	K (GPa)	G (GPa)
Diamond	1.2	417	468
Al-4Si	22.8	76.5	27.1
Al-8Si	22.3	76.9	28.2
Al-12Si	21.8	77	28.9
Al-16Si	20.2	78	30.2

Since the coefficient of thermal expansion (CTE) of Al-Si alloy decreases with the addition of Si (the CTE of pure Al is ~23×10^{-6} K^{-1}, while that of Al-Si alloy is ~20×10^{-6} K^{-1} at room temperature)[26, 27], the CTE of the diamond/Al-Si composites decreases as the Si content in the aluminum matrix increases, which is also consistent with the prediction of the Kerner model. This behavior can be explained in terms of thermal strain. Within a certain temperature range, the tendency of thermal expansion of the composites increases with increasing temperature, and the tensile stress within the aluminum matrix is gradually transformed into compressive stress[29, 30], so that the thermal expansion of the composites is suppressed.

When the temperature continues to increase, the thermal stress on the composites increases. Since the elastic modulus and strength of the diamond particles are much higher than those of the aluminum matrix, the aluminum matrix starts to undergo thermal expansion, while the diamond particles are almost unaffected under the same thermal stress conditions. Therefore, the change of the coefficient of thermal expansion of the composites is mainly caused by the thermal expansion behavior of the aluminum matrix, so the coefficient of thermal expansion of the composites has a similar trend to that of the aluminum matrix. In addition, the interfacial reaction products also play an important role in the change of the coefficient of thermal expansion of composites. When the Si content in the aluminum matrix increases to 12wt.%, the bonding of the composites changes from simple mechanical bonding to chemical metallurgical bonding, which is evidenced by the EDS elemental mapping (Fig. 4) and XRD results (Fig. 5) confirming the mutual diffusion of Al, Si, and C atoms and the formation of interfacial reaction products (SiC and Al_4C_3) at the diamond/aluminum interface. This makes the interfacial bond of the composites stronger, and the limiting effect of the interface on the aluminum matrix is enhanced. When the composites are subjected to thermal expansion, the anchoring effect of the interfacial reaction product on the aluminum matrix causes the aluminum matrix to tend to change the trend of diamond particles, which greatly restricts the expansion of the aluminum alloy matrix. Thus, the expansion effect of the composites is reduced, so the composites have a low coefficient of thermal expansion.

3.3 Bending properties of diamond/Al-Si composites

The bending strength of diamond/Al-Si composites with different Si contents is shown in Fig. 8. It can be seen that the bending strength of the composites tends to increase and then decrease with the increase of Si content. When the Si content in the aluminum matrix increases from 4wt.% to 16wt.%, the bending strength of the composites initially rises, reaching a peak value of 296 MPa at 12wt.% Si, and then slightly declines to 276 MPa at 16wt.% Si. This maximum value of 296 MPa is significantly higher than the 149 MPa reported for diamond/Al-Cu composites fabricated using a Cu-modified Al matrix with 22 μm diamond particles[15], and higher than the 76 MPa of diamond/(Ti)/Al composites prepared with titanium plating on the diamond surface using diamond particles measured 165 μm[31, 32]. This indicates that the alloying of the aluminum matrix with Si can significantly enhance the bending strength of the composites.

The reason why the bending strength of diamond/Al-Si composites shows a first increase and then decrease with the increase of Si content in the aluminum matrix can be attributed to the change of the interfacial structure between the diamond particles and the aluminum matrix[4, 33]. When the Si content in the aluminum matrix is low, the Si atoms in the aluminum alloy liquid do not have enough driving force for diffusion due to the low concentration. Only a small number of Si atoms are enriched at the interface of the composites, which leads to a more difficult interfacial reaction and a weaker interfacial bond. When the composites are fractured due to the load, the crack initiation and expansion at the early stage of fracture is easier, and the ability to resist the load is poorer, so the bending strength of the composites is lower. As the Si content in the aluminum matrix increases, the concentration of Si atoms in the aluminum alloy liquid rises, and the driving force of Si atom diffusion increases. A large number of Si atoms are continuously enriched at the interface and generate SiC by interfacial reaction with diamond, and thus the interfacial bonding of the composites is enhanced, which greatly reduces the probability of crack initiation in the composites, and therefore the bending strength of the composites is improved[9]. However, when the Si content in the aluminum matrix is too high, a large number of Si atoms in the aluminum alloy liquid diffuse to the interface between diamond and aluminum during the preparation of the composites. During the cooling-down stage of the composites, a large number of Si atoms crystallize and precipitate at the interface, forming a thick brittle interfacial layer. This significantly degrades the interfacial bonding strength, thereby reducing the bending strength of the composites.

4 Conclusions

In this study, the interface of diamond/Al composites was regulated by means of aluminum matrix modification, and diamond/Al-Si composites with different Si contents were successfully prepared by a vacuum-assisted pressure infiltration process. The effects of Si content on the microstructure and properties of the composites were systematically investigated.

(1) Changes in Si content lead to transitions in the interfacial structure of the composites, but have little effect on the distribution of their structures. The Si addition to the aluminum matrix can effectively promote the interfacial wetting of the composites, reduce the generation of the interfacial side reaction Al_4C_3, and improve the performance of the composites. As the Si content in the aluminium matrix increases from 4wt.% to 16wt.%, both the thermal conductivity and bending strength of the diamond/Al-Si composites first increase and then decrease, while the coefficient of thermal expansion decreases monotonically.

(2) When the Si content in the aluminum matrix is 12wt.%, the composites prepared from 180 μm diamond particles exhibit excellent overall performance, with the highest values of thermal conductivity and bending strength of 654 $W \cdot m^{-1} \cdot K^{-1}$ and 296 MPa, respectively, as well as a low coefficient of thermal expansion of 5.96×10^{-6} K^{-1}.

(3) When the diamond particle size is 425 μm, the thermal conductivity of diamond/Al-12Si composites can be as high as 713 $W \cdot m^{-1} \cdot K^{-1}$. The thermal conductivity of the composites can be effectively improved by considering both the Si addition to the aluminium matrix and the diamond particle size simultaneously. In practical applications, composites with different properties can be prepared by adjusting the Si content in the aluminum matrix and selecting appropriate diamond particle sizes to meet the requirements of different scenarios.

Conflict of interest

Prof. Yuan Liu is an EBM of *CHINA FOUNDRY*. He was not involved in the peer-review or handling of the manuscript. The authors have no other competing interests to disclose.

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