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ARTICLE

Progress in Heat Conduction of Diamond/Cu Composites with High Thermal Conductivity

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Abstract: Diamond/Cu composite has advantages of low density, high thermal conductivity, and tailorable coefficient of thermal expansion (CTE) and possesses a good thermal matching performance with new generation chips. Therefore, it has a widespread application prospect in electronic packaging with high heat flux density and other fields. However, due to the poor wettability between diamond and Cu as well as high interfacial thermal resistance, the conductivity of the composite is even lower than that of the copper, which restricts its application. The interface of composite is modified to transform their mechanical and physical bonding state into a chemical and metallurgical bonding by pre-metallization, pre-alloying copper matrix and optimization of composite processing so as to improve wettability nowadays. In this research, surface modification, interface theory related to thermal conducting model and research development in finite element simulation were reviewed. The difficulties of the fabrication process, thermal conducting models, and key direction of future development were discussed. The effects of parameters such as diamond content and particle size on microstructure and thermal conducting performance were summarized.

Key words: diamond/Cu composites; interfacial wettability; interface theory; finite element simulation

With the rapid development of microelectronics technology, semiconductor chips and integrated circuits are used to pursue faster computing speeds and more complex functions, and core electronic devices of chip-level and module-level continue to develop towards the direction of being smaller in size and functionally integrated with high power density. Electronic equipment used in many high-tech fields such as national defense, electronics industry, new energy, aerospace, gets more and more powerful, and the level of integration becomes higher and higher, as shown in Fig.1. The problem of heat dissipation has become a key restricting the development of these industries[1]. As a consequence, the thermal conductivity (TC), density, thermal expansion coefficient of the heat dissipating material for core chip are requested with higher and higher requirements in the design of high-performance electronic package[2-4]. At present, thermal management materials as the first and second generation cannot meet the requirements of high thermal conductivity, while diamond/copper composites have the advantages of good thermal conductivity beyond 500 W/m·K, low coefficient of thermal expansion (CTE) in the range of 4×10^{-6}~6×10^{-6} K^{-1} and high strength, as shown in Fig.2. As the third generation of thermal management material, it is well matched with the new generation chip with thermal expansion coefficient of 5×10^{-6} K^{-1}, which is an excellent choice to replace the existing thermal sink materials, and has a broad application prospect in the field of high-performance electronic packaging[5-10]. The forming technology and the mechanism of diamond/copper composites will become a hot spot of electronic packaging research in the future.

In 1995, Sun Microsystems and Lawrence Livermore jointly studied a multi-chip module called Dymalloy. Diamond/Cu composite as the substrate of the module was prepared by vacuum pressure infiltration, and its thermal conductivity reached up to 420 W/m·K[11], which exceeded the thermal

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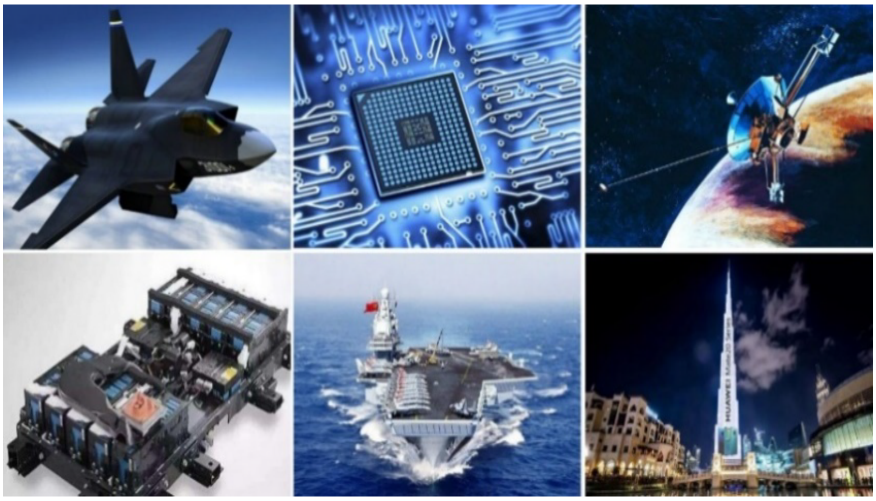

Fig.1 Application fields of diamond/copper composites

conductivity of electronic packaging materials and heat sink materials over the same period. Diamond/Cu composites have been widely concerned by researchers all over the world. However, the thermal conductivity of diamond/copper obtained by the fabrication process was far lower than the theoretical thermal conductivity, even lower than that of the copper matrix. For example, Ciupinski et al[12] prepared diamond/copper composite by pulse plasma sintering (PPS), and the thermal conductivity was only 200 W/m·K. And Chen et al[13] adopted spark plasma sintering (SPS) to synthesize diamond/Cu composites, and the thermal conductivity was only 167.4 W/m·K with 50% volume fraction of diamond and sintering temperature at 860 °C. Moreover, Chen et al[14] also prepared diamond/copper composites by high temperature and high pressure method, and its thermal conductivity was only 318.7 W/m·K. These results of diamond/copper do not meet the requirements for high power, high integration and high performance electronic devices. The reason is its poor wettability, weak interfacial bonding and high interfacial thermal resistance between diamond and copper leading to low thermal conductivity of the composites. Furthermore, there are a large number of free electrons in the copper matrix, while the heat transfer is mainly realized by the movement of electrons[15]. And diamond mainly relies on phonon to transfer heat, i.e., passing lattice vibration to finish heat transfer. Heat conduction of lattice can be regarded as the result of phonon diffusion motion. Hence, diamond/copper composites mainly rely on phonons and electrons for heat conduction[16], while the defect of impurities, pores and cracks in diamond particles can result in phonon scattering. In addition, the extremely poor wettability between diamond and copper matrix causes the defects of pores and voids in the interface, which restricts the phonon free path by means of the interaction with phonons, affecting the thermal conductivity and mechanical properties of the material.

Therefore, how to improve the bonding strength of interface and decrease the interfacial thermal resistance of diamond/Cu composites is of great significance. For the time being, researchers all over the world modified the interface of diamond/copper composites by pre-metallizing the surface of diamond[17-20] or pre-alloying the copper matrix[21-24] to enhance the thermal conductivity. Moreover, it is still the key to further obtain high density and high thermal conductivity diamond/copper matrix composites through improved preparation process. This research mainly reviews surface modification, interface theory related to thermal conducting and research development in finite element simulation of diamond/copper, introduces the influence law and mechanism of diamond with different addition elements and particle sizes on microstructure and thermal conductivity, and details the existing problems as well as prospects the future development trend.

1 Interfacial Modification Methods of Diamond/Copper Composites

Due to chemical inertness and extremely poor wettability between diamond and Cu, defects such as holes and voids are likely to appear at the interface, causing the weak interface combination, which affects the strength and TC. It is obviously unable to meet the requirements of application in the field of electronic packaging. At present, there are two important methods to improve the interfacial wettability. One is to pre-alloy the copper matrix. The other is pre-metallizing the diamond surface, changing the direct contact between Cu matrix and diamond into the contact between coating on the diamond particles and Cu matrix. These two methods mainly improve the wettability by selecting coating elements, coating thickness, distribution, and reaction products, etc.

1.1 Pre-alloying of Cu matrix

Pre-alloying of Cu matrix that improves the wettability

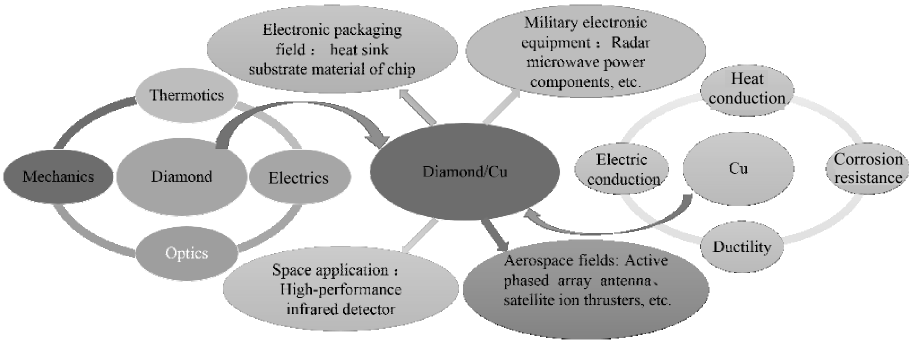

Fig.2 Introduction of diamond/Cu composites

between Cu and diamond by modifying the pores and other defects on the interface, and increasing the interfacial bonding strength of the composite is to add appropriate alloying elements into copper matrix to form carbide layer with diamond. It is worth noting that the addition of active elements should be neither too little nor too much. If the addition of alloying elements is too little, the carbide layer generated at the interface will be thin or uneven, resulting in low thermal conductivity of the composite. If the addition of alloying elements is too much, it is inevitable that some active elements will be dissolved in the Cu matrix, causing a significant decrease in TC of Cu matrix, which will further lead to lower TC and poor mechanical properties of the diamond/Cu composite. For the present, the high TC of diamond/Cu composites is mainly obtained by combining the alloying elements (Cr, B, Ti, Zr, etc) with the process to form carbide layer on the diamond surface.

Weber et al[8] synthesized Cu-*X* (*X*=B, Cr) alloys by vacuum induction melting, fabricated the Cu-*X*/diamond composites with the volume fraction of diamond at 60% by GPI and studied the effects of the addition of active elements B and Cr on TC and CTE of the composite. The results showed that the interfacial bonding gradually strengthens with the increase of the addition of alloy elements: TC of the composite increases first and decreases, and CTE decreases. The fabricated composites achieved a high thermal conductivity (>600 W/m·K) and a low coefficient of thermal expansion ($<10^{-5}$ K^{-1}). Bai et al[25] also added alloying element B into Cu matrix and studied the effect of B alloying element on the TC of diamond/Cu composite, and the coefficient of thermal expansion of the composite is shown in Fig.3. The research result is similar to the results of Weber's research: TC of Cu-B/diamond composite increases first and then decreases with increasing the addition of B element, and the maximum TC is obtained in the intermediate content, i.e. 868 W/m·K, and CTE reaches to 5.3×10^{-6} K^{-1}, which is significantly higher than that of the previous research results[26-28].

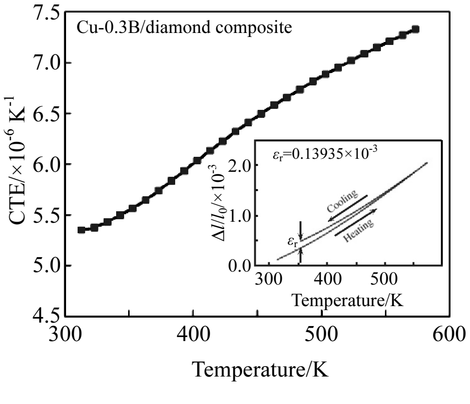

Fig.3 Coefficient of thermal expansion of the Cu-0.3wt% B/diamond composite[25]

Schubert et al[29] strengthened the interfacial bonding in Cu/diamond composites using atomized copper alloy with minor additions of chromium, and a thin nano-sized Cr_3C_2 layer was formed, achieving improvement in bonding strength and thermophysical properties of the composites. Mańkowski et al[30] also prepared Cu-Cr/diamond composites with 0.8vol% Cr by PPS. When the volume fraction of diamond is 50%, the thermal conductivity of the composite is up to 658 W/m·K. In addition, Ciupinski et al[31] also fabricated Cu-0.65Cr/diamond composite. The results attained that the thickness of the interfacial carbide layer reaches to 81 nm and TC is up to 687 W/m·K at 850 °C. Compared with the interfacial thickness of Cu-Cr/ diamond composite prepared by Grzonka et al[32], it was found that the Cr content of the added element has an optimal value, and SEM images of Cu-Cr/diamond composite interface are shown in Fig.4.

Chung et al[33] used mechanical method to uniformly mix diamond powder, copper powder with titanium powder, and sintered diamond/Cu-Ti composite by pressureless infiltration. And the effect of Ti content on the thermal properties of diamond/Cu-Ti composites was investigated. It was found that the diamond particles of 300 μm in size with the volume fraction of 50% and diamond particles of 150 μm in size with the volume fraction of 10%, as well as 2 at% Ti mixed composite attained the optimal TC of 608 W/m·K and CTE

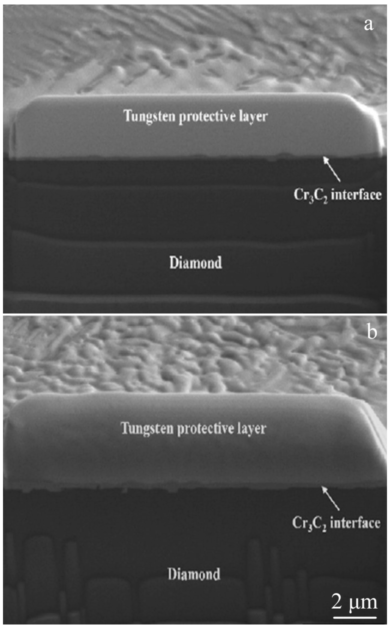

Fig.4 SEM images of Cu-Cr/diamond composite interface: (a) Cu-0.65Cr and (b) Cu-0.8Cr[29-32]

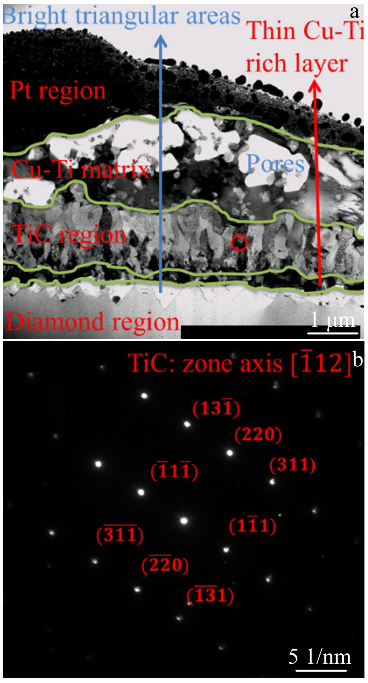

Fig.5 TEM images of diamond/Cu composite interface[33]

of 5.4×10^{-6} K^{-1}. The literature found that the alloying element Ti reacts with diamond at the interface to form the intermediate layer of TiC, which greatly enhances the interfacial bonding between diamond particles and Cu matrix, thereby improving the TC of the composite, as shown in Fig.5.

Li et al[34] obtained Cu-Zr alloy with 0wt%~1.0wt% Zr by alloy melting method, then fabricated Cu-Zr/diamond composite by pressure infiltration. It was found that the TC increases first and then decreases with the increase of Zr addition, and the maximum TC is 930 W/m·K when the Zr content is 0.5 wt%. After analyzing, it was found that the high TC is caused by the formation of ZrC interfacial layer which is uniformly distributed between diamond and Cu, and the maximum TC is obtained at the interfacial thickness of 400 nm. It can be seen that the thickness of the interfacial layer is the key to improve TC.

In summary, the addition of different alloying elements reduces defects such as interfacial voids, enhances the interface bonding, and improves the interfacial wettability. However, alloying of Cu matrix reduces the TC of Cu matrix to some extent, and the TC of the interface is much lower than the TC of diamond and Cu, which inevitably affects the excellent thermal physical properties of the composite. Meanwhile, the TC of added elements and formed carbides should not be low, and the elements are supposed to be added in an appropriate amount. If the content of added alloying elements is little, the carbide formed is little, and the effect on the interface will not improve. If the content of alloying elements is too much, an excessive amount of carbides forms, and TC of the carbide is low, weakening the TC of the composite. Table 1 compares the composites fabricated by various techniques.

1.2 Pre-metallization of diamond surface

In addition to copper matrix pre-alloying, the improvement of interfacial wettability of diamond/copper composites is also an effective way to pre-metallize diamond surface. It uses different plating processes to deposit metal layer or metal carbide layer with the thickness from nanometer degree to micron degree on the surface of diamond. Metal layer or metal carbide layer can form better metallurgical bonding or chemical bonding with diamond surface, and the coating can also prevent graphitization of diamond surface during sintering process. Thus, the wettability difference between diamond and copper improves, and the thermophysical properties of the composites are optimized. The common methods of diamond surface metallization include vacuum microdeposition, magnetron sputtering, diffusion and salt bath.

Ueda et al[45] directly formed graphene layer on the surface of diamond by high temperature annealing under the action of vacuum Cu catalyst. The typical characteristics of monolayer graphene were observed in diamond heterostructures by means of diamond Raman spectroscopy. It was found that the coverage of graphene layer on diamond is about 85%.

Ren et al[9] used vacuum microdeposition to deposit Ti and Cr coating on diamond surface. The effect of coating on the microstructure and thermal conductivity of diamond/copper composite was studied. The results showed that the existence

Table 1 Comparison of composites fabricated by various techniques[31]

Diamond particle		Interfacial layer		Matrix material	λ/W·m^{-1}·K^{-1}	Fabrication technique	Ref.
Size/μm	Fraction/vol%	Material	Thickness/nm				
50~500	0.93~0.95	-	-	Cu-50wt% Ti	260~490	HTHPI	[35]
500~600	0.90	-	-	Cu	540~750	HTHPI	[36]
20~110	0.50~0.80	-	-	Cu	226~742	HTHPI	[7]
115±42	0.55~0.70	Cr_7C_3	0~2000	Cu	178~657	SPS	[9]
70	0.50~0.70	Mo_2C	1000	Cu	562~608	GPI	[37,38]
70	0.50~0.70	Cr_7C_3	1000	Cu	510~562	GPI	[39]
430	0.60~0.63	W-W_2C-WC	110~470	Cu	476~907	SI	[19]
550	0.80~0.90	Carbides	-	CuB/Cr/Ti	683~688	HTHPI	[40]
40~60	0.10~0.40	ZrC	-	Cu1Cr0.6Zr	240~430	SPS	[23]
230	0.90	B_4C	-	CuB	566~731	HTHPI	[41]
200	0.60	B_4C/Cr_3C_2	-	CuB	300~700	GPI	[8]
110	0.20~0.40	Cr_3C_2	188~294	Cu0.8Cr	478~591	HP	[42]
120	0.42	B_4C/Cr_3C_2/TiC/ZrC	-	Cu0.3B/0.8Cr/0.15Al/1Ti	495/490/195/295	HP	[43]
150~300	0.5~0.60	TiC	-	CuTi	409~608	S	[33]
110	0.55	ZrC	270/320/480	Cu0.8Zr/1.2Zr/2.4Zr	596/615/542	HP	[44]

Note: HTHP-high pressure high temperature, SPS-spark plasma sintering, GPI-gas pressure infiltration, SI-spontaneous infiltration, S-pressureless sintering, HP-hot pressure

of coating decreases the interfacial thermal resistance and improves the thermal conductivity of diamond/copper composites. Compared with titanium coating, chromium coating can reduce the thermal resistance of the interface between diamond and copper more effectively. The analysis concluded that the negative effect of Cr coating on the thermal conductivity of copper matrix is small, resulting in higher thermal conductivity of chromium-plated diamond/copper composite. The Cr on the surface of diamond particles exists in the form of Cr_7C_3. It is found that the thickness ratio of carbide layer to copper layer is 2:1, and the optimum thickness of interface layer is 0.6~0.9 μm. Finally, when the volume fraction of diamond is 70%, the thermal conductivity of Cr-coated diamond/copper composites reaches 657 W/m·K.

Abyzov et al[46] used diffusion method to deposit tungsten coatings and tungsten carbide coatings with a thickness of 260 nm on diamond particles with an average diameter of 200 μm at 1000 °C. Diamond/copper composites with volume ratio of 1:1 were prepared by pulse plasma sintering. It was found that the following chemical reactions occur on the surface of diamond during PPS: $W \rightarrow W_2C \rightarrow WC$. The coating has complete continuity and uniformity. The thermal conductivity of tungsten coating and tungsten carbide on diamond particle/copper composites is 690 and 550 W/m·K, respectively. Kang et al[38] formed molybdenum carbide coated diamond particles in mixed molten salt by salt bath method using MoO_3 as reaction medium. The formation mechanism of Mo_2C layer on diamond particles was discussed. Mo_2C-diamond/Cu composites were prepared by vacuum pressure infiltration. The effect of molybdenum carbide interlayer on the thermal properties of diamond/copper composites was studied. The results showed that the formation of Mo_2C layer can be divided into two steps, i.e., the reduction of MoO_3 to MoO_2 and the reduction of MoO_2 to Mo_2C. Moreover, Mo_2C effectively improved the wettability of diamond particles and copper. When the volume fraction of Mo_2C-diamond particles is 60%, the relative density of diamond/copper composites reaches 99.5%, the thermal conductivity reaches 596 W/m·K, and the coefficient of thermal expansion is 7.15×10^{-6} K^{-1}. In the temperature range of 900~1050 °C, Mo powder of 2 μm in size and diamond particles with diameter of 120~150 μm were used as raw materials. Mo_2C sub-micron coated diamond particles were also prepared by salt bath method. The effects of different plating time (10, 15, and 30 min) on the coating thickness were also discussed. Diamond/copper composites were prepared by vacuum pressure infiltration. The results showed that Mo_2C interlayer strengthens the interface bonding between diamond and copper, reduces the interface thermal resistance and improves the thermal conductivity. The thermal conductivity of diamond/copper composites is up to 657 W/m·K[47], and the morphology of diamond@Mo_2C particle coating is displayed in Fig.6.

In recent years, the integrated double-layer design of coating and copper matrix has attracted many researchers' attention. The double-layer design is a layer of carbide formed on the surface of diamond by one-time plating and then another layer of copper is deposited on the surface of carbide layer by chemical or magnetron sputtering. This design can improve the uniform distribution of diamond particles in copper matrix, strengthen the bonding strength of the interface between diamond particles and copper, increase the density of the composite, and then improve the mechanical and thermophysical properties of the composite.

Lin et al[48] firstly used salt bath method to plate Ti on the surface of synthesized single crystal diamond particles, and

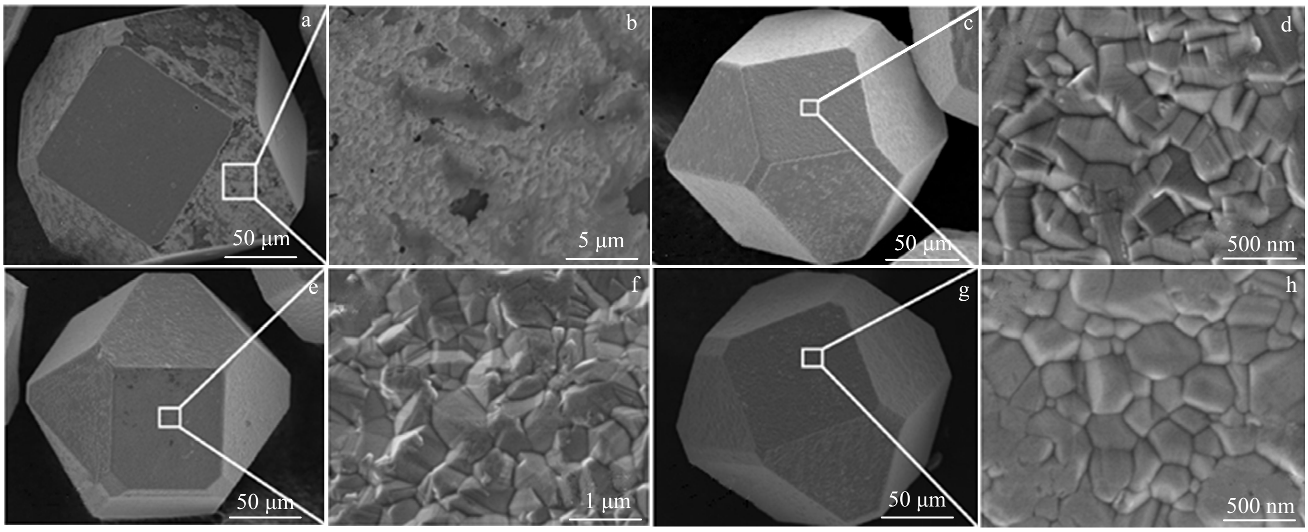

Fig.6 Morphologies of diamond@Mo_2C particle coating at 1050 °C for different plating time: (a, b) 10 min, (c, d) 15 min, (e, f) 30 min and (g, h) 60 min[47]

then a layer of copper was deposited on Ti-coated diamond particles by magnetron sputtering. In order to simulate the actual processing of diamond/copper composites, Cu-Ti coated diamond particles were heated in vacuum at 1073 K for 30 min. It was found that TiC forms between diamond particles and Ti layer. Between Ti layer and Cu layer, Cu diffuses into Ti layer and forms the composite phase at Ti/Cu interface. The results provided experimental basis for interface design and production of copper/diamond composites.

Zhang et al[49] deposited tungsten coatings on diamond surface by diffusion method, and then copper coatings were prepared by electroless plating. Finally, double-coated diamond/copper composites were prepared by vacuum hot pressing. The effect of double coating on the microstructure and thermal conductivity of the composite was studied. The results showed that tungsten coating improves the interfacial bonding and reduces the boundary thermal resistance between diamond and copper matrix to the greatest extent. And the copper coating is beneficial to the sintering and low temperature densification of diamond/copper composites. The thermal conductivity of the diamond/copper composites is 721 W/m·K, which is close to the theoretical value. Pan et al[50] also designed a double coating on the diamond surface. First, W was plated on the diamond surface by salt bath method, and then Cu was deposited on the W layer by chemical method. Diamond/copper preform was prepared by cold pressing with 1.2 GPa ultra-high pressure, and then sintered in vacuum at 1100 °C for 1 h. It was found that the thermal conductivity of the composites reaches 661 W/m·K when the volume fraction of diamond is 60%. It was found that the uniform distribution of diamond particles and strong interfacial bonding of double-layer particles in the composite are achieved by electroless plating instead of adding copper powder. Then, Pan et al[51] deposited Zr on the surface of diamond by magnetron sputtering, and the outer layer was still copper with electroless plating, as shown in Fig.7. Using the same equipment and method, the interface of the composite

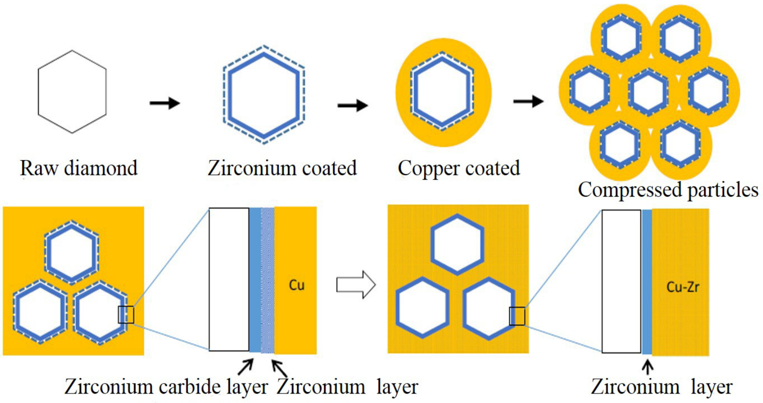

Fig.7 Isothermal oxidation curves of coated C/C samples in air at 1773 K[51]

prepared by double-coated diamond particles is well bonded and the particles are evenly distributed in the copper matrix. The thermal conductivity is as high as 720 W/m·K when the volume fraction of diamond is 65%.

According to the interfacial modification of diamond/ copper composites by metallization of diamond surface, the thermophysical properties of diamond/copper composites by changing the plating process were improved[45-51]. At present, the main reactive plating methods, such as salt bath plating and powder covered sintering, have the advantages of strong bonding, less interface defects and stable performance. The coating by physical methods such as magnetron sputtering and micro-deposition is thinner, but it requires more complicated equipment. Therefore, the coating conditions and processes should be selected according to the actual needs. In addition, few researches have been done on the effect of coating structure and interface structure on the thermophysical properties of diamond/copper composites, which is extremely difficult to further improve the thermophysical properties of diamond/Cu composites.

2 Thermal Conduction Models

In order to instruct and predict better the experiments on the thermal conductivity of diamond/copper composites, main corresponding theoretical thermal conductivity models are proposed, such as Maxwell-Eucken model, H-J model, and differential effective medium. Among all the three models, Maxwell-Eucken model[52,53] is the earliest and most widely used one, which can be described by the following formula:

$$\lambda_{\mathrm{c}} = \lambda_{\mathrm{m}} \frac{2(1-V_{\mathrm{d}})\lambda_{\mathrm{m}} + (1+2V_{\mathrm{d}})\lambda_{\mathrm{d}}}{(2+V_{\mathrm{d}})\lambda_{\mathrm{m}} + (1-V_{\mathrm{d}})\lambda_{\mathrm{d}}} \tag{1}$$

where λ_{c} is the thermal conductivity of diamond/copper composite; λ_{m} and λ_{d} are the thermal conductivity of Cu matrix and the reinforced diamond particles, respectively; V_{d} is the volume fraction of diamond spherical particles in the diamond/copper composite.

This model assumes that the reinforced particles are spherical and uniformly dispersed in the continuous phase, so there are some restrictions in the case when the volume fraction of the reinforced phase in the matrix is lower. However, some researchers applied the Maxwell-Eucken model in the case of large volume fraction of the reinforced phase, and the results obtained are not accurate. When the volume fraction of diamond reinforced phase is less than 20%, the theoretical calculation results are close to the experimental results. When the volume fraction of diamond reinforcement phase is more than 20%, the theoretical results are quite different from the experimental results mainly because of the interaction between particles and defects such as voids. In addition, Maxwell's theoretical model neglects the thermal resistance at the interface among the matrix, the reinforced phase, and the non-spherical reinforced phase particles, which makes the theoretical thermal conductivity predicted by the model larger than the experimental thermal conductivity. The thermal resistance at the interface between matrix and reinforcement phase in composites is mainly due to the poor bonding force between the two phases and the difference of linear thermal expansion coefficient between the two materials. This thermal resistance is called Kapitza resistance[54]. According to the heat theory, the thermal conductivity of the composite is a function of the thermal conductivity of the particles and the volume fraction of the matrix phase and each component, but it has nothing to do with the particle size. Yoshida et al[7] found that the thermal conductivity of composites also depends on the size of diamond particles, as shown in Fig.8.

Therefore, in order to predict the thermal conductivity more comprehensively and accurately, Hasselman et al[55] improved Maxwell's theoretical model and proposed H-J model, which takes fully consideration on the dimensions (radius of reinforced phase particles) of diamond reinforced phase and the interfacial thermal resistance between the matrix (reciprocal of the thermal conductivity h_{c} of the interface phase):

$$\lambda_{\mathrm{eff}} = \lambda_{\mathrm{m}} \frac{[2(\frac{\lambda_{\mathrm{d}}}{\lambda_{\mathrm{m}}} - \frac{\lambda_{\mathrm{d}}}{ah_{\mathrm{c}}} - 1)V_{\mathrm{d}} + \frac{\lambda_{\mathrm{d}}}{\lambda_{\mathrm{m}}} + 2\frac{\lambda_{\mathrm{d}}}{ah_{\mathrm{c}}} + 2]}{[(1 - \frac{\lambda_{\mathrm{d}}}{\lambda_{\mathrm{m}}} + \frac{\lambda_{\mathrm{d}}}{ah_{\mathrm{c}}})V_{\mathrm{d}} + \frac{\lambda_{\mathrm{d}}}{\lambda_{\mathrm{m}}} + 2\frac{\lambda_{\mathrm{d}}}{ah_{\mathrm{c}}} + 2]} \tag{2}$$

where λ_{eff} is the thermal conductivity of diamond/copper composites; λ_{m} and λ_{d} are the thermal conductivity of Cu matrix and reinforced phase diamond particles, respectively; V_{d} is the volume fraction of diamond spherical particles in diamond/copper composites; a is the radius of diamond reinforced phase spherical particles; h_{c} is the thermal conductivity of interface, which can be determined by the materials of reinforced phase and matrix.

From this model, it can be seen clearly that when $h_{\mathrm{c}} \rightarrow \infty$, H-J model is consistent with Maxwell-Eucken model expression. However, when calculating the theoretical thermal conductivity, the model assumes that the matrix and diamond

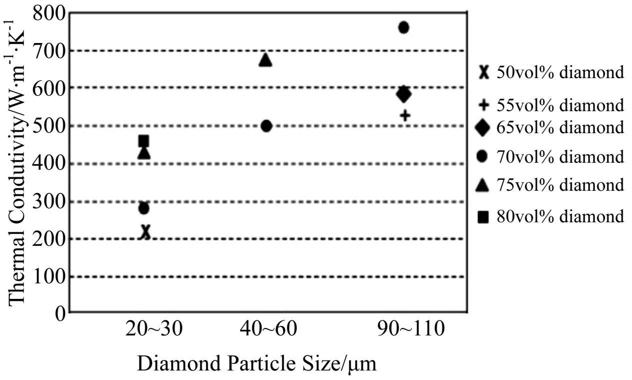

Fig.8 Thermal conductivity of diamond/copper composites with different particle sizes[7]

are perfectly integrated.

The interfacial bonding of diamond/copper composites changes from non-bonding or physical bonding to chemical bonding or diffusion bonding due to the alloying of copper matrix or the introduction of surface metallization of diamond particles, which is conducive to heat transfer through the interface. Yuan et al[56] considered an interfacial modification of diamond/copper consisting of carbide layer, carbide forming element layer, alloy layer or intermetallic compound layer from diamond to copper matrix in turn. In this case, the diagrams and physical models of diamond/copper composites with multi-layer interfaces are shown in Fig.9:

$$R_{\mathrm{c}} = \sum (R_i + R_{(i-1)\to i}) \ (i\geq 1) \tag{3}$$

where R_i represents the interface thermal resistance of interface i, $R_{(i-1)\to i}$ represents the thermal resistance of the layer from interface i–1 to interface i.

Thermal conductivity is the most important physical parameter to characterize the heat dissipation of high thermal conductivity materials, which reflects the heat transfer capacity of materials. In insulator, the carrier of heat conduction for diamond is phonon, and the carrier of heat conduction for copper is electron. For heat transfer through the interface between two dielectrics or dielectrics and conductive materials, phonons are the main heat carriers. Because of the low concentration of free electrons in dielectrics, the contribution of electrons can be neglected.

Phonon is the energy quantum of lattice vibration. The motion of a large number of phonons is the lattice vibration medium. It can be seen as the air mass of phonon diffusion movement. The interaction and collision between phonons are the main mechanism of heat conduction. Therefore, the ideal gas equation can be used to describe the thermal conductivity of phonon diffusion movement. For ideal gases, the expression of thermal conductivity is

$$\lambda_{\mathrm{c}} = \frac{1}{3} cvl \tag{4}$$

where c is the heat capacity per unit volume (J/kg·K), l is the average free path of phonon (m), and v is the average velocity of phonon motion (m/s).

The frequent collisions between phonons and the scattering of phonons at defects are caused by various defects (such as holes, voids, impurities and dislocations) in diamond/copper composite, which reduces the average free path of phonons. Therefore, the higher the density, the higher the interfacial bonding force; the fewer internal defects, the better the thermal conductivity. According to the phonon heat transfer theory, the interface thermal resistance is expressed by the following formula according to the acoustic mismatch model (AMM)[9,57]:

$$R_{\mathrm{c}} = \frac{1}{h_{\mathrm{c}}} = \frac{2}{\rho_{\mathrm{m}} c_{\mathrm{m}}} \frac{v_{\mathrm{d}}^2}{v_{\mathrm{m}}^3} \frac{(\rho_{\mathrm{m}} v_{\mathrm{m}} + \rho_{\mathrm{d}} v_{\mathrm{d}})^2}{\rho_{\mathrm{m}} v_{\mathrm{m}} \cdot \rho_{\mathrm{d}} v_{\mathrm{d}}} \tag{5}$$

where R_{c} represents the interfacial thermal resistance; ρ_{m} and ρ_{d} denote the density of matrix and diamond reinforced phase, respectively; v_{m} and v_{d} denote the phonon velocity of matrix and diamond reinforced phase, respectively; c_{m} denotes the specific heat capacity of matrix. As for the definition of reinforcement phase and matrix next to each interface, the incident side is considered as reinforcement phase and the other side is considered as matrix. v can be established from longitudinal (v_{l}) and transverse (v_{t}) velocities[58,59]:

$$\frac{3}{v^3} = \frac{1}{v_{\mathrm{l}}^3} + \frac{2}{v_{\mathrm{t}}^3} \tag{6}$$

Yang et al[60] considered the interfacial thermal resistance when studying the thermal conductivity of Cu-Ti/diamond composites. So the overall interfacial thermal resistance R of Cu-Ti/diamond composites when carbides formed at the interface can be expressed by the thermal resistance (R_{c}) of the interfacial carbides and the Kapitza resistance (R_{K}). R_{K} has two sub-interfaces: diamond ($R_{\mathrm{D/C}}$) and copper carbide ($R_{\mathrm{Cu/C}}$).

$$R = R_{\mathrm{c}} + R_{\mathrm{K}} = R_{\mathrm{c}} + R_{\mathrm{Cu/C}} + R_{\mathrm{D/C}} \tag{7}$$

where $R_{\mathrm{c}}=l/\lambda_{\mathrm{c}}$, l and λ_{c} are the thickness and intrinsic thermal conductivity of carbide layer, respectively.

Therefore, the interfacial thermal resistance is also related to the thickness of the interlayer and the intrinsic thermal conductivity. The total thermal resistance of the interfacial layer is the sum of the thermal resistance of the carbide layer, the matrix copper-carbide layer and the diamond carbide layer. If the interfacial layer is multilayer, the total thermal resistance of the interfacial layer is the sum of the carbide layer and the diamond carbide layer. Fig.10 is a comparison between H-J theoretical calculation of thermal conductivity of chromium-plated diamond and experimental values at different volume fractions of diamond. It can be seen that the theoretical value is higher than the experimental value. Because of the limitations of the model itself, the H-J theoretical model has more excellent prediction effect and applicability under the condition of low volume fraction of reinforced phase particles. With the increase of volume fraction of reinforced phase particles, the theoretical value is higher than the experimental value. Additionally, the theoretical model deviates significantly from the experimental results and the reliability reduces.

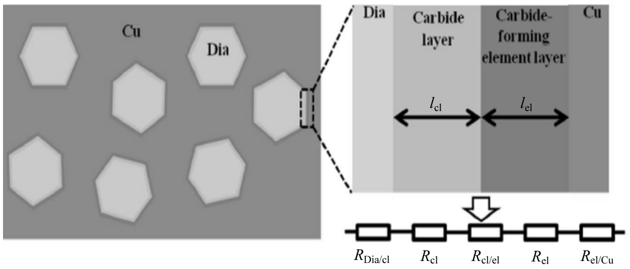

Fig.9 Schematic of diamond/copper composite multilayer interface[56]

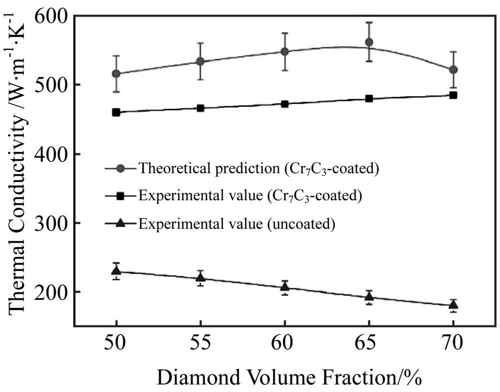

Fig.10 Comparison to the theoretical and experimental values of H-J model[39]

In conclusion, the phonon mismatch model does not consider the diffusion scattering of phonons at the interface, but the diffuse mismatch model (DMM) assumes that all phonons scatter at the interface, which can be better estimated by Kapitza resistance[61] as there are a few cracks and a little air at the interface of the coated diamond-copper composites, leading to the enhancement of phonon scattering at low density or high porosity. Specific formula is as follows:

$$R_{\mathrm{K}} = \left[1.02 \times 10^{10} \times \frac{(\sum_{j} v_{i,j}^{-2})(\sum_{j} v_{3-i,j}^{-2})}{\sum_{i,j} v_{i,j}^{-2}} \right]^{-1} \times T^{-3} \tag{8}$$

where the subscript 3–*i* is used to indicate the opposite side of side *i*, *v* is used to indicate the phonon velocity (which can be obtained by Eq.(6)), and *T* is used to indicate the absolute temperature.

Table 2 Interfacial resistance and theoretical thermal conductivity of diamond/Cu-2%Ti composites[33]

$R_{\mathrm{TiC/diamond}}$/m^2·K·W^{-1}	R_{TiC}/m^2·K·W^{-1}	$R_{\mathrm{Cu/TiC}}$/m^2·K·W^{-1}	R_{K}/m^2·K·W^{-1}	$K_{\mathrm{P}}^{\mathrm{eff}}$/W·m^{-1}·K^{-1}	K_{c}/W·m^{-1} K^{-1}	$K_{\mathrm{c\text{-}p}}$/W·m^{-1} K^{-1}
5.2×10^{-10}	5.8×10^{-8}	1.4×10^{-10}	5.9×10^{-8}	1019	707	662

The R_{K} based on DMM only depends on the phonon velocity *v* and absolute temperature *T* in phase *j*. The phonon velocity of carbides can be described as follows:

$$v_{\mathrm{c}} = \sqrt{G/\rho} \tag{9}$$

where *G* represents the shear modulus and *ρ* denotes the density of carbides.

Based on DMM model, Chung et al[33] also considered the porosity between diamond and Cu-Ti interface, and the result of interfacial resistance and theoretical thermal conductivity of diamond/Cu-2%Ti composites is shown in Table 2. In the model, the effect of porosity is considered by redistributing effective thermal conductivity to the composite. According to the mixing rules, the effective thermal conductivity of the composites was obtained by combining the thermal conductivity of the composites with air.

$$\lambda_{\mathrm{c\text{-}p}} = \lambda_{\mathrm{c}} V_{\mathrm{c}} + \lambda_{\mathrm{air}} V_{\mathrm{porosity}} \tag{10}$$

where $\lambda_{\mathrm{c\text{-}p}}$ represents the thermal conductivity of the composite with porosity; V_{c} and V_{porosity} are the volume fraction of the composite and porosity, respectively.

In addition to Maxwell-Eucken and H-J models, Bruggeman[62] predicted the thermal conductivity of heterogeneous composites in 1935 and proposed Bruggeman theoretical model:

$$1 - V_{\mathrm{d}} = \frac{\lambda_{\mathrm{d}} - \lambda_{\mathrm{c}}}{\lambda_{\mathrm{d}} - \lambda_{\mathrm{m}}} \left(\frac{\lambda_{\mathrm{m}}}{\lambda_{\mathrm{c}}} \right)^{\frac{1}{3}} \tag{11}$$

Based on Eq.(10), Every[63] presented a theoretical model of differential effective medium (DEM)[64] for particle reinforced composites, which is expressed as follows:

$$(1 - V_{\mathrm{d}}) \left(\frac{\lambda_{\mathrm{c}}}{\lambda_{\mathrm{m}}} \right)^{\frac{1}{3}} = \frac{\lambda_{\mathrm{d}}^{\mathrm{eff}} - \lambda_{\mathrm{c}}}{\lambda_{\mathrm{d}}^{\mathrm{eff}} - \lambda_{\mathrm{m}}} \tag{12}$$

where λ_{c} and λ_{m} are the interfacial thermal conductivity of composite materials and metal matrix, respectively; V_{d} is the volume fraction of diamond reinforced particles. Considering that *r* is the average size of the diamond reinforced phase and h_{c} is the interface thermal conductivity between the reinforced phase and the matrix, the effective thermal conductivity of the reinforced phase is as follows[65]:

$$\lambda_{\mathrm{eff}} = \frac{\lambda_{\mathrm{d}}}{1 + \frac{\lambda_{\mathrm{d}}}{h_{\mathrm{c}} r}} \tag{13}$$

In conclusion, because of the poor wettability between diamond and copper matrix, neither solid solution nor carbide can be formed. The interface between diamond and copper matrix is a simple mechanical combination, and there are holes and voids, which seriously affect the strength and thermal conductivity. Maxwell-Eucken, H-J model and DEM models consider that the interface of diamond/copper composites is perfect and there is no defect such as voids. At the same time, these theoretical models mainly consider phonons as carriers. The electrons in the copper matrix are not involved which also have some influences on the thermal conductivity. Therefore, the interface thermal conductivity of diamond/copper composites cannot be reflected in practice, and there is a certain gap between theoretical prediction and experimental value.

However, DEM model considers not only the shape and size of reinforcement phase, but also the effect of interface voids and cracks on the thermal conductivity of composites. The predicted results of DEM model are closer to the experimental values than those of other models.

3 Thermal Conduction Models

From the theoretical model, diamond/copper composites with high thermal conductivity, low coefficient of thermal expansion and good mechanical properties are obtained, relying only on the function of volume fraction, content, grain size, etc to predict thermal conductivity coefficient of composite, which is not ideal and needs to be further improved. With the development of computer technology, the function of finite element analysis software is constantly improved, and it is easy to analyze and establish related model of complex problems through finite elements. Therefore, a very effective and feasible approach is provided by finite element analysis theory and methods to predict the thermal conductivity of diamond-reinforced/copper matrix composites.

Current researches on the analysis of metal matrix composites using numerical simulation techniques are mostly focused on the effects of volume fractions of diamond reinforced phase, coating type, thickness and distribution on thermal conductivity. Rape et al[66] established 3D model of single-grain diamond/CuAgZr composites with different volume fractions of diamond, compared the results of finite element simulation prediction with theoretical calculations and experimental results, and found that the results are consistent with theoretical calculations and experimental results. The model predictions, experiments and theoretical results are shown in Fig.11. This discovery fully illustrates that the method of finite elements can be used to predict the thermal conductivity of Cu/Zr based composites containing diamond particles. Cho et al[67] simulated the distribution of current density in the electro-deposition process of diamond/copper composite using the COMAOL software when the size of the diamond particles is 66 and 420 μm. It is worth noting that the current density pattern and the distribution of microstructure are consistent. Zain-UI-Abdein et al[68] studied the effect of the coating under different diamond volume fractions on the effective thermal conductivity of the diamond/copper composite with the methods of experiment and numerical simulation. The finite element model was established by the microcosmic analysis diagram of the composite, and the thermal conductivity under different volume fractions was simulated. Finally, compared with the experimental results, the two results were in good agreement. Wu et al[69] conducted numerical simulation on heat flux in diamond/copper composites using COMSOL software and the results showed that its thermal conductivity and thermal expansion coefficient were 614.87 W/m·K and 9.45×10^{-6} K^{-1}, respectively. Yang et al[70] fabricated diamond/aluminum composites with tungsten carbide coatings by pressure infiltration method with the thermal conductivity of 588 W/m·K. In order to better understand its thermal conduct mechanism, the finite element method is used to establish 3D model of an imperfect interface. According to the different fracture characteristics of composites, the effects of non-homogeneous interface and homogeneous interface thermal conductivity (h) on the thermal conductivity of composites are studied. The results showed that when the interface thermal conductivity of the diamond {111} surface (h_{111}) is lower than 10^5 W/m^2·K or higher than 10^9 W/m^2·K, the total thermal conductivity of the composite is mainly related to the interface thermal conductivity (h_{100}) on the surface of the diamond {111}. While h_{111} ranges from 10^5 W/m^2·K to 10^9 W/m^2·K, the combined effect of h_{111} and h_{100} on thermal conduction behavior of composites cannot be ignored. In addition, the carbide coating leads to the additional interface thermal resistance on the diamond surface {100}. However, the average interfacial thermal conductivity of composites improves, which leads to the increase of the total thermal conductivity of composites.

Because the distribution of diamond in the copper matrix is random, its heat transfer process is also more complex. Scholars start to adopt finite element simulation method to study the thermal conductivity of composite materials with the development of computer technology, for the advantages of saving cost, saving time and labor, avoiding recording errors, and certain variables can be fixed so that the influence of the parameters on thermal conductivity by changing one parameter individually can be studied. Generally speaking, the thermal conductivity of diamond/copper composite is mainly related with diamond content, particle size, coating type, thickness and interface binding condition. Currently, simulation

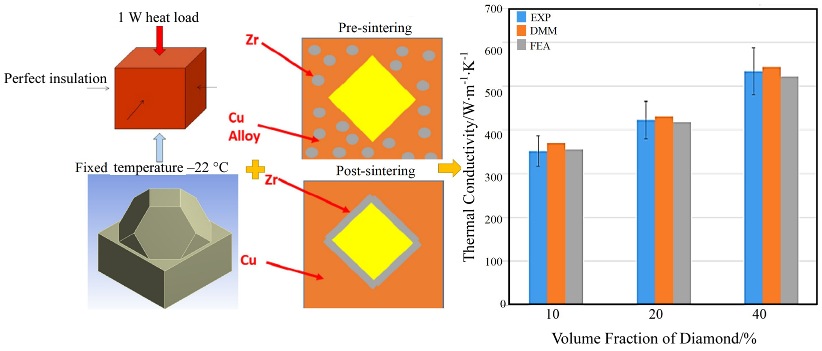

Fig.11 Experiment and theoretical results of single-grain diamond/CuAgZr composite in 3D model[66]

studies on composites are also focused on these areas, while numerical simulation of thermal performance of diamond particles with random distribution in diamond/copper composites is very few, especially the systematic study of defects such as voids. Due to the poor wettability of diamond particles and copper, no matter what process method is used, it is impossible to achieve complete density, and pores, cracks, holes and other defects exist at the interface, which have a large impact on the strength, corrosion resistance, especially the thermal conduction of the diamond/copper composite. At the same time, the distribution, size and shape of the pores are difficult to quantify by experiments. Therefore, numerical simulation is necessarily adopted to study the effect of defects such as pores on the thermal conductivity of diamond/copper composites.

4 Conclusions and Outlook

Diamond/copper composites are considered to be excellent thermal management materials in areas of aerospace, military, electronics, etc, due to their high thermal conductivity, low thermal expansion and isotropy, etc, which have received widespread attention around the world. The United States, Japan, the United Kingdom and other countries with more developed industries have applied diamond/copper composites as electronic packaging materials in the field of aerospace electronics, optoelectronic systems and high-performance servers; for example, Raytheon, Thales and other companies have carried out the research of engineering application on phase change substrate with high thermal conductivity, diamond/metal substrate and carbon matrix composite diffuser substrate with high thermal conductivity. While some researches mainly focus on the deposition of diamond thick film on the copper substrate for improving the diamond/copper composites. According to research, the demands of new weapons with high-power microwave devices and high-power laser devices with high thermal conductivity, low expansion coefficients of high heat dissipation performance parts are increasing. At present, the Non-ferrous Metal Research Institute, the Chinese Academy of Sciences, etc, have also carried out the research on packaging material of diamond/copper, high thermal carbon composite and sample development, but the theoretical and technical level needs to be further improved.

Aiming at the problem of poor wettability and difficult machining of diamond/copper composite, the main challenge of the current development is how to obtain a good interfacial bonding strength between diamond and copper. Researchers all over the world improve the thermal conductivity of composites from the aspects of surface modification, near-net forming technique and other points of view. For example, the near-net forming technique such as powder injection molding is used to prepare the porous preformed skeleton of diamond. Powder injection molding in combination with gas pressure infiltration can prepare diamond/copper composite or the final formed parts. The thermal conductivity exceeds 500 W/m·K or even reaches to 1000 W/m·K. However, the research of high performance electronic packaging materials such as diamond/copper composite in China is still relatively behind, and there is still a big gap compared with the researches in other countries. Therefore, in order to improve the interfacial bonding strength of diamond/copper composite, increase the density and thermal conductivity, optimize the process and then put it into engineering application to accelerate the rapid development of China's electronics industry, the future research should be focused on the following aspects:

1) Control of the content, distribution and thickness of pre-metallized elements on diamond surface or pre-alloyed elements on copper matrix. The introduction of coating improves the interface bonding, but also increases the interface thermal resistance. Therefore, how to develop a new formulation with new coating process to control the content, distribution form, and thickness of diamond surface pre-plated carbide layer or copper matrix alloying element to obtain a perfect interface is instructive.

2) Optimize the thermal conductivity model of diamond/copper composite. In the existing predictive model, the phonon is mainly considered as the carrier. However, the electrons in the copper can also have an effect on thermal conductivity. While the current thermal conductivity theoretical model does not consider the effects of electron-phonon coupling. Therefore, these models do not actually reflect the interfacial thermal conductivity of diamond/copper composite.

3) Diamond/copper composites with high density and high thermal conductivity obtained by improving the preparation method are still important. Several mentioned processes have been reported for the preparation of high thermal diamond/copper composites, but these methods are based on high temperature and high pressure. These strict conditions increase energy consumption and require extremely high requirements for equipment. Since the thermal expansion of copper matrix does not match with diamond particles, the interface will crack slightly during the cooling process. At the same time, graphitization of diamond occurs at high temperature or diamond even reacts with the coated metal, causing changes in the form, distribution, and thickness of diamond surface carbide. These factors increase the interface thermal resistance, resulting the significant reduction of the thermal conductivity of diamond/copper composites. There- fore, optimizing the high temperature method is very challenging, and there is an urgent need to carry out researches on sintering mechanism and interface structure.

4) Engineering applications. On the basis of diamond/copper composites prepared with excellent comprehensive properties by optimizing the interface between diamond and copper and the sintering process, the next urgent problem to be solved is to achieve engineering applications. Due to the high hardness of diamond-reinforced particles, it is difficult to

process diamond/copper composites. For composites with simple shapes and restricted size, one-step molding can be achieved. However, for parts with more complex structures, near-net forming techniques need to be developed to fabricate complex structure shapes. In addition, the surface roughness of composites is high, and diamond is exposed on the surface of composites, which will have a certain influence on the performance of subsequent welding. As a result, the research of near-net forming and processing techniques will be developed to solve the problem of forming and welding of diamond/copper composites to provide theoretical and technical support in the large-scale engineering applications of diamond/copper composites.

5) Aiming at the requirement of core electronic equipment for high heat flux chip, etc. It is necessary to conduct the research on the design and forming mechanism of heat sink components based on micro-channel core chips, as well as the low-cost and high-efficiency fabrication technology to apply in the fields of national defense, aerospace, new energy, electronics and other fields.

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高导热金刚石/铜复合材料的导热研究进展

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摘　要：金刚石/铜复合材料具有密度低、热导率高及热膨胀系数可调等优点，且与新一代芯片具有良好的热匹配性能，在高热流密度电子封装领域具有非常广泛的应用前景。然而由于金刚石与铜界面润湿性差，界面热阻高，导致材料热导率比铜还低，限制了其应用。为了改善其界面润湿性，通过在金刚石表面金属化或对铜基体预合金化等手段来修饰复合材料界面，以提高金刚石/铜复合材料的热导率。本文综述了表面改性、导热模型相关的界面理论以及有限元模拟的研究进展，讨论了制备工艺、导热模型和未来发展的关键方向，总结了金刚石添加量、颗粒尺寸等制备参数对其微观组织结构和导热性能的影响规律。

关键词：金刚石/铜复合材料；界面润湿性；界面理论；有限元仿真

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