

Research Article

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Theoretical analysis of interfacial design and thermal conductivity in graphite flakes/Al composites with various interfacial coatings

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Abstract: Graphite flakes/Al composites are promising thermal management materials due to high thermal conductivity (TC) in basal plane orientation, matched coefficient of thermal expansion, and good machinability. In this article, the acoustic mismatch model and the effective medium approach are applied to predict the influence of different interfacial coatings on the interfacial thermal conductance (ITC) and the TC of graphite flakes/Al composites, respectively. With the increase in the thickness of interfacial coatings, the ITC and the TC of graphite flakes/Al composites decrease. For the composites with Ni, Cr/Cr₇C₃/Cr₃C₂, Si/SiC, Ti/TiC, WC, and Mo/Mo₂C coatings, the ITC is sensitive to coating thickness. In order to obtain ideal TC of graphite flakes/Al composites, the thickness of the coatings should be controlled below 1 μm. It is reasonable that the TC of the graphite flakes/Al composites increases as the volume fraction of graphite flakes increases. The TC of the graphite flakes/Al composites increases with the ITC and changes slowly when the ITC increases to a certain extent. Si/SiC and WC coatings are proposed to be the most promising candidates to improve the thermal performance of graphite flakes/Al composites.

Keywords: graphite flakes/Al composites, interfacial coatings, thermal conductivity

1 Introduction

The rapid development of smaller, more powerful, and more efficient electronic systems has created a very challenging set of heat dissipation, a critical factor in guaranteeing performance and reliability in electronic devices [1,2]. Hence, there is a great desire to develop very efficient thermal management materials with ideally lightweight, extremely high thermal conductivity (TC), and low coefficient of thermal expansion (CTE). In recent years, aluminum matrix composites with excellent comprehensive properties are therefore becoming promising materials for heat sinks and spreaders in multi-functional electronic packaging systems. Among these, SiC/Al composites, due to their low price, matched CTE, and near-net-shape fabrication versatility, are frequently used in the thermal management market [3,4]. However, their relatively low TC (below 250 W/m K) [5] cannot meet well with the increasing requirement in packaging application. Diamond/Al composites with higher TC (~750 W/m K) [6] have high potential in this regard; however, their high price and poor processability limit their application [7]. Due to their low price, high TC in basal plane orientation, and good machinability, graphite flakes/Al composites have received ever-growing attention [8–16].

Similar to other carbon/Al composites, the optimization of the interface plays a crucial role in determining the thermophysical properties of the graphite flakes/Al composites. It is well known that during the fabrication of graphite flakes/Al composites at elevated temperatures, it is extremely easy to form interfacial aluminum carbide (Al₄C₃), which is easy to hydrolyze under water vapor condition and can cause the interfacial de-bonding and deteriorate the properties of the composites [17]. Meanwhile, excessive Al₄C₃ may also greatly deteriorate the interfacial thermal conductance (ITC) to some extent due to its rather low intrinsic TC [18]. It is widely known that the ITC is important in various applications and metrologies of various forms of composites [19–23].

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Recent studies have shown that graphite flake surface coating can be applied to prevent the formation of Al_4C_3 and enable good bonding with the Al matrix, thus significantly improve the thermal and mechanical properties of graphite flakes/Al composites [24–29]. Although good interfacial bonding is helpful for desirable TC, the additional interface layers will probably also act as thermal boundary barriers, which will influence the thermal properties of composites [18]. Moreover, systematic experimental research studies on quantitative evaluation and comparison of interface layers on the thermal properties are still insufficient yet. Based on the performance parameters of the surface coatings [22,30–33], the prediction of the ITC and the TC of the composites will help to improve the process of the surface coating of reinforcements and the design of the composites.

The present work aims to theoretically evaluate the effects of various coating types and their thicknesses on the ITC and the TC of graphite flakes/Al composites. Typical interfacial coatings are divided into metals and their carbides, and a calculating method based on the acoustic mismatch model (AMM) and effective medium approach (EMA) are applied. The results may guide for choosing the appropriate interface types and their thicknesses for graphite flakes/Al composites.

2 Model employed for calculating the ITC and the TC

Considering the differences in the electronic and vibrational properties, there is an interface thermal barrier for heat transfer between Al matrix and graphite flakes, known as Kapitza resistance, which will limit the improvement of the TC of composites. The ITC h is the reciprocal of Kapitza resistance. For graphite flakes/Al composites with various coatings, the schematic of the multi-layered interfacial structure and physical model of the interfacial thermal resistance are shown in Figure 1. The total interfacial thermal resistance (R_{total}) includes the Kapitza resistance between Al matrix and the coating layer (R_{m-l}), the thermal resistance of the coating layer (R_l), and the Kapitza resistance between the coating and graphite flakes (R_{l-r}).

The AMM can be used to theoretically estimate the R_{m-l} and R_{l-r} :

$$R = \frac{2(\rho_m v_m + \rho_r v_r)^2}{C_m \rho_m^2 v_m^2 \rho_r v_r} \left(\frac{v_r}{v_m} \right)^2, \quad (1)$$

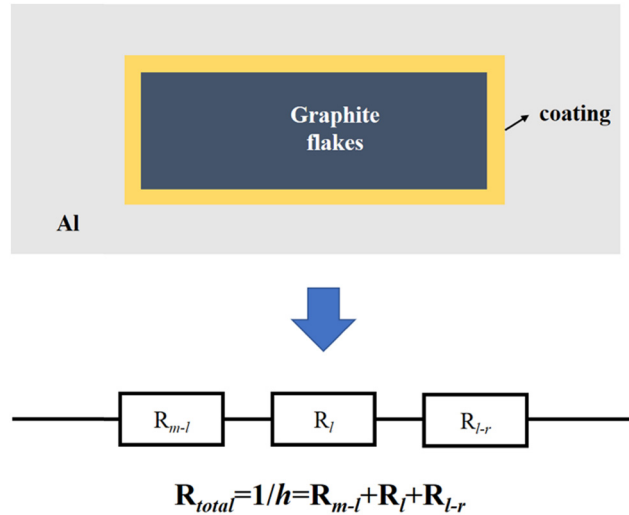


Figure 1: Schematic diagram of the interfacial thermal resistance of graphite flakes/Al composites with an interfacial coating.

where ρ , v , and C are the density, sound velocity, and specific heat capacity, respectively, and m and r denote the matrix and reinforcement. Generally, there are three modes of vibrations in isotropic materials: one longitudinal (v_l) and two transverse (v_t) [19]. The average sound velocity can be expressed as follows:

$$\frac{3}{v^2} = \frac{1}{v_l^2} + \frac{2}{v_t^2}. \quad (2)$$

As a typical anisotropic material, however, graphite flake still has three modes: one longitudinal, one transverse, and one out-of-plane mode (v_o), and the v_o is far less than v_l and v_t [19]. Hence, the average sound velocity of graphite flake can be expressed as follows:

$$\frac{2}{v^2} = \frac{1}{v_l^2} + \frac{1}{v_t^2}. \quad (3)$$

The R_l can be expressed by the equation:

$$R_l = \frac{l}{K_l}, \quad (4)$$

where l and K_l are the thickness and the TC of the coating layer, respectively.

The TC of graphite flakes/Al composites in the basal plane of graphite flakes (defined as the X–Y plane) is much higher (~ 880 W/m K) than that perpendicular to the basal plane (defined as the Z-direction). The schematic diagram of the X–Y plane and the Z-direction of graphite flakes/Al composites are shown in Figure 2.

The EMA can theoretically estimate the TC of graphite flakes/Al composites (K_c), which can be expressed as follows [16,20]:

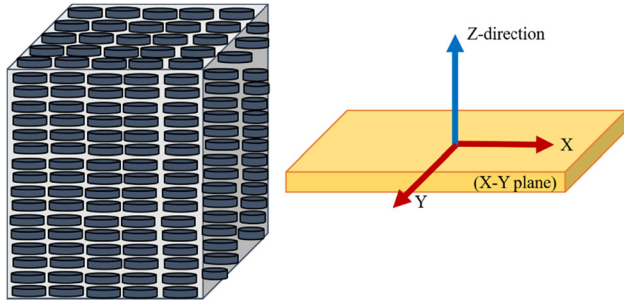


Figure 2: Schematic diagram of the X-Y plane and the Z-direction of graphite flakes/Al composites.

$$K_C^{XY} = K_m \frac{2 + f[\beta^{XY}(1 - S^{XY})(1 + \langle \cos^2 \theta \rangle) + \beta^Z(1 - S^Z)(1 - \langle \cos^2 \theta \rangle)]}{2 - f[\beta^{XY}S^{XY}(1 + \langle \cos^2 \theta \rangle) + \beta^ZS^Z(1 - \langle \cos^2 \theta \rangle)]}, \quad (5)$$

$$K_C^Z = K_m \frac{1 + f[\beta^{XY}(1 - S^{XY})(1 - \langle \cos^2 \theta \rangle) + \beta^Z(1 - S^Z)\langle \cos^2 \theta \rangle]}{1 - f[\beta^{XY}S^{XY}(1 - \langle \cos^2 \theta \rangle) + \beta^ZS^Z\langle \cos^2 \theta \rangle]}, \quad (6)$$

with

$$\beta^{XY} = \frac{K_r^{XY} - K_m}{K_m + S^{XY}(K_r^{XY} - K_m)}, \quad (7)$$

$$\beta^Z = \frac{K_r^Z - K_m}{K_m + S^Z(K_r^Z - K_m)}, \quad (8)$$

where K_C^{XY} and K_C^Z are the TC of composites in the X-Y plane and Z-direction, respectively. K_m is the TC of matrix alloy. K_r^{XY} and K_r^Z are the effective TC of reinforcements in the X-Y plane and Z-direction, respectively. f is the volume fraction of reinforcements. $\langle \cos^2 \theta \rangle$ is the orientation function, which can describe the distribution of the reinforcements in the Al matrix. When the graphite flakes are completely disordered in the matrix, $\langle \cos^2 \theta \rangle = 1/3$; when the graphite flakes are completely perpendicular to the X-Y plane, $\langle \cos^2 \theta \rangle = 0$; and when the graphite flakes are completely parallel to the X-Y plane, $\langle \cos^2 \theta \rangle = 1$. S is the geometrical factors related to the aspect ratio of the reinforcements, approximately regarding the shape of graphite flakes as a disk, and can be given by:

$$S^{XY} = \frac{\pi t}{4D}, \quad (9)$$

$$S^Z = 1 - \frac{\pi t}{2D}, \quad (10)$$

where D and t are the diameter and thickness of graphite flakes, respectively. Hence, the EMA can be reduced to:

$$K_C^{XY} = K_m \left[1 + \frac{f}{\frac{\pi t}{4D}(1 - f) + \frac{K_m}{K_r^{XY} - K_m}} \right], \quad (11)$$

$$K_C^Z = K_m \left[1 + \frac{f}{\left(1 - \frac{\pi t}{2D}\right)(1 - f) + \frac{K_m}{K_r^Z - K_m}} \right]. \quad (12)$$

The effect of multilayer interface (Figure 1) on the TC of graphite flakes may be equivalent to that of graphite flakes covered with a thermal barrier. Hence, the relationship between the effective TC of graphite flakes (K_r) and their intrinsic TC (K_{Gf}) and ITC ($h = 1/R_{\text{total}}$) is as follows [7,21]:

$$K_r^{XY} = \frac{K_{Gf}^{XY}}{1 + \frac{2K_{Gf}^{XY}}{h^{XY}D}}, \quad (13)$$

$$K_r^Z = \frac{K_{Gf}^Z}{1 + \frac{2K_{Gf}^Z}{h^Zt}}. \quad (14)$$

In our calculations and numerical analyses, the constants of materials involved are listed in Table 1. The representative values of 50 vol%, 500, and 20 μm are assigned to f , D , and t , respectively.

3 Predictions and numerical analyses

Based on the aforementioned models, the effects of various coating types and their thicknesses on the ITC and the TC of graphite flakes/Al composites can be theoretically evaluated. Furthermore, the variation of the ITC and the TC of graphite flakes/Al composites with a different volume fraction of graphite flakes has been systematically analyzed.

3.1 ITC of graphite flakes/Al composites

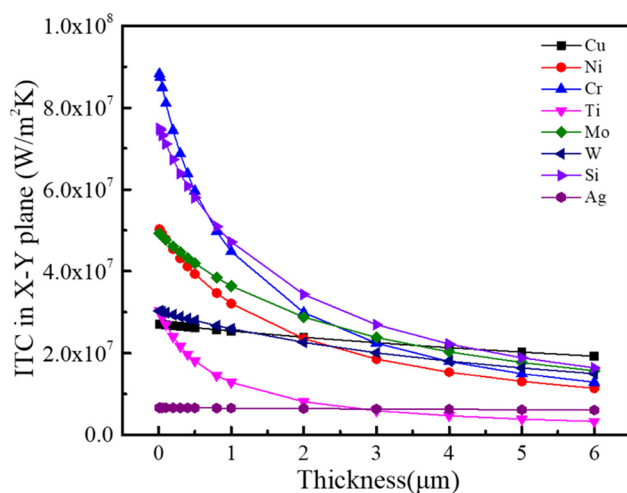
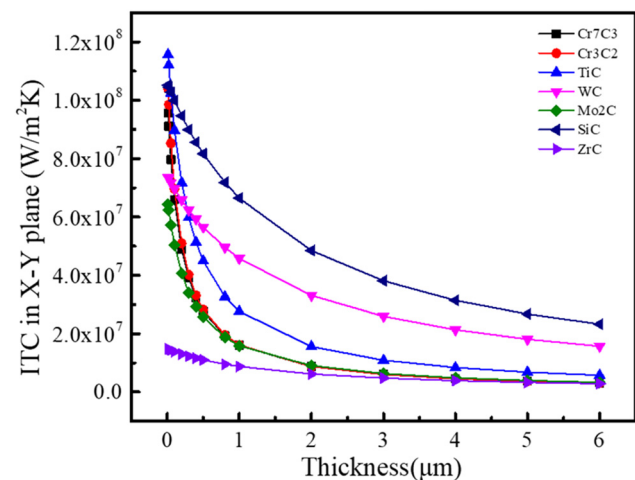
As a bridge between the matrix and the reinforcements, the interface can transfer the external force borne by the matrix to the reinforcements; meanwhile, the appropriate interface bonding can prevent crack propagation and slow down stress concentration. Light wave, acoustic wave, thermoelastic wave, and shock wave will scatter and absorb at the interface, resulting in discontinuity of physical properties. From this point of view, the carrier transport at the interface is blocked, which is not conducive to the heat conduction of the material, especially the condition of a weak bonding at the interface. Due to the poor wettability of graphite and Al, it is more feasible and necessary to improve interfacial bonding by introducing additional interface layers by surface metallization of graphite flakes. Generally, the commonly used coating materials include metals and their carbides.

Table 1: Constants of coating materials [22,30–33]

Materials	Density/(kg m ⁻³)	Sound velocity/(m s ⁻¹)			Specific heat/(J K ⁻¹ kg ⁻¹)	TC/(W m ⁻¹ K ⁻¹)
		Longitudinal	Transverse	Average		
Al	2,700	6,240	3,040		880	237
Graphite flakes	2,250	23,600	15,900		710	880 (<i>a</i> -axis)
		1,960	700			27 (<i>c</i> -axis)
		8,970	5,332		703	126
SiC	3,100	12,810	7,530		678	179
W	19,320			4,029	133	178
WC	14,900			4,697	203	120
Mo	10,200			3,916	248	138
Mo ₂ C	9,000	6,257	3,605	4,003	347	21
Cr	7,190	6,980	4,100	4,545	446	90
Ti	4,540	6,070	3,120		522	22
TiC	4,930	9,330	6,092		569	36
Cu	8,900	4,910	2,500	2,801	386	398
Ni	8,800	5,630	2,960	3,310	447	88
Ag	10,500	3,780	1,740	1,960	237	427

As shown in Figure 3, the ITC of graphite flakes/Al composites with metal coatings decreases with the increase in coating thickness, but the trend of variation is obviously different. For the composites with W, Cu, and Ag coatings, the ITC decreases slowly with the increase in coating thickness. Particularly, the ITC of composites with Ag coating is relatively lower than others, which is not suitable for coating material of graphite flakes/Al composites. That is because of the lower sound velocities of Ag. For the composites with Cr, Si, Mo, Ni, and Ti coatings, the ITC is sensitive to coating thickness. When the coating thickness is 1 μm , the ITC of composites with Cr, Si, Mo, Ni, and Ti coating decreases 49.3, 37.1, 26.2, 36.2, and 57.3%, respectively.

Hence, in order to obtain satisfied interface heat transfer behavior, the thickness of the coating of the composites should be strictly controlled. Among these coatings, the ITC of composites with Ti coating is relatively low, and the coating thickness should be controlled at tens of nanometers. Cr and Si are better coating materials due to their higher ITC. However, the solubility of Cr in the Al matrix is so large that it is not advisable to be used as a coating material of diamond/Al composites [18]. Mo, as a diamond coating material, has been found to form intermetallic compounds with Al at the interface, so the TC of the diamond/Al composites has not been improved [23]. Therefore, Si is a suitable coating material of the graphite flakes/Al

**Figure 3:** ITC of graphite flakes/Al composites with typical metal coatings as a function of coating thickness.**Figure 4:** ITC of graphite flakes/Al composites with typical carbide coatings as a function of coating thickness.

composites from the point of view of improving the ITC of the composites; however, the thickness of the coating still needs to be controlled.

For graphite flakes/Al composites with carbide coatings, as shown in Figure 4, the ITC similarly decreases with the increase in coating thickness and changes gradually after falling to a certain extent. The ITC of graphite flakes/Al composites with ZrC coating is at a lower level; hence, ZrC is not a suitable coating material. The graphite flakes/Al composites with other coating are all sensitive to the thickness of the coating, and the ITC has been significantly reduced when the coating thickness is more than 500 nm. Particularly, for Cr_7C_3 , Cr_3C_2 , TiC, and Mo_2C coatings, the ITC of composites with 100 nm thickness coatings decreases 30.1, 32.9, 22.4, and 21.6%, respectively. Considering the coating process of flake graphite, it is relatively difficult to realize the coating thickness under 100 nm. Compared with other carbide coatings, SiC is more suitable as a coating material for graphite flakes/Al composites. The ITC of graphite flakes/Al composites with SiC coating is higher as a whole; meanwhile, when the coating thickness is 500 nm, the ITC only decreases 22.35%.

3.2 TC of graphite flakes/Al composites

Figures 5 and 6 show the variation of the TC of graphite flakes/Al composites with different coatings along the X–Y plane, which is consistent with the ITC. With the increase in the thickness of interfacial coatings, the TC of graphite flakes/Al composites decreases. The overall

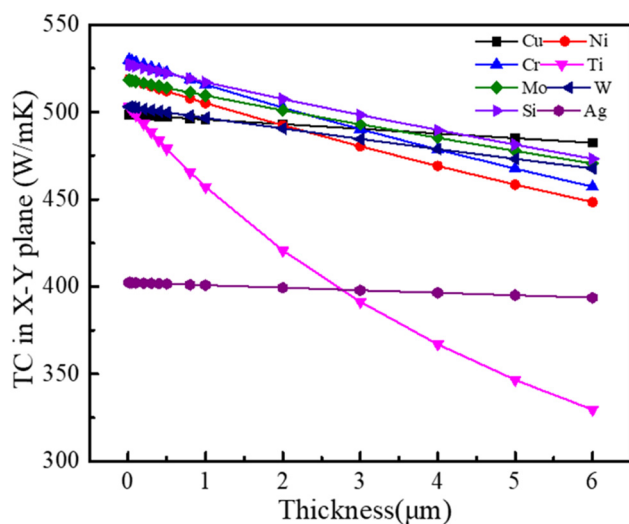


Figure 5: TC of graphite flakes/Al composites with typical metal coatings as a function of coating thickness.

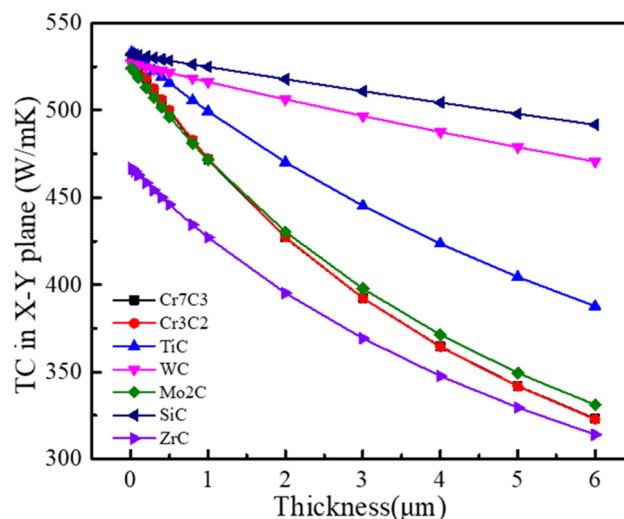


Figure 6: TC of graphite flakes/Al composites with typical carbide coatings as a function of coating thickness.

level of the TC of Ag coating composite is low, and the TC of Ti coating composites decreases rapidly with the increase in coating thickness. When the coating thickness is 1 μm , the TC of composites with Ti coating decreases 9.1%. The graphite flakes/Al composites with Cr and Si coatings exhibit higher TCs when coating thickness is less than 1 μm .

In comparison, the thickness of the carbide coating has a more significant effect on the TC of the composites. The TC of composites with ZrC coating is relatively lower, which is consistent with the variation trend of the ITC. The TC of Cr_7C_3 , Cr_3C_2 , TiC, and Mo_2C coating composites changes significantly with the coating thickness. The overall TC of SiC and WC coating composites is relatively high, and SiC or WC coating can be used for graphite flakes/Al composites.

Figure 7 shows the calculated ITC and the TC of 50–70 vol% graphite flakes/Al composites with various coatings with a thickness of 0.2–1.0 μm . It is well reasonable that the TC of the graphite flakes/Al composites increases as the volume fraction of graphite flakes increases. Since the graphite flakes as high thermal reinforcements construct the heat conduction path of the composites, the increase in their contents is bound to improve the TC of the composites. Furthermore, as for the case of the introduced interface coatings, it can be seen that the TC of the graphite flakes/Al composites increases with the ITC and changes gradually when the ITC increases to a certain extent. The ITC plays an important role in improving the TC of graphite flakes/Al composites. Because of the differences between the physical properties of graphite and Al, thermal flux through the interfaces changes and this so-called thermal

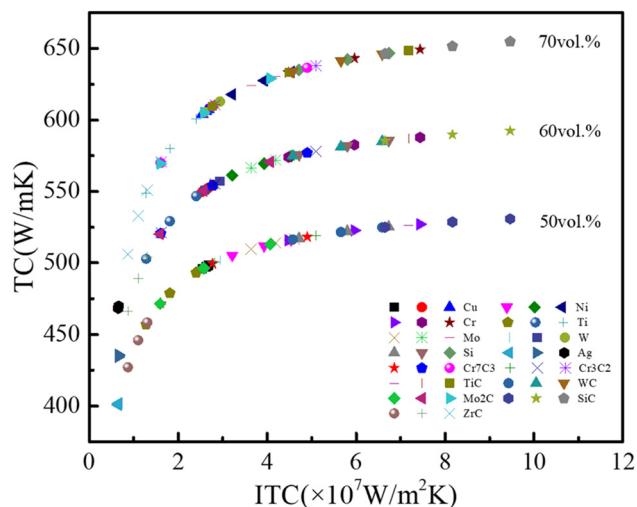


Figure 7: Predicted ITC and TC of 50–70 vol% graphite flakes/Al composites with a coating thickness of 0.2–1.0 μm .

resistance limits the heat transfer of composites. Once the interface coating layers introduced in the composites can improve the ITC, such as Si and SiC, and the TC of the composites can reach higher level. The TC of the graphite-SiC/Al composite in the X-Y plane fabricated by vacuum hot pressing technology can reach (528–735) W/(mK) [24]. In order to obtain better ITC, the coating layers should be with a nanoscale thickness. However, the interface is one of the factors affecting the TC of composites. Once the perfect interface bonding is obtained, the TC of composites often depends on other factors, such as the size and volume fraction of graphite, the distribution of graphite in the matrix, the composition of matrix alloy, and so on. Apparently, the size and volume fraction of graphite flake are also the factors affecting the ITC.

It can be seen that there are still some errors in the prediction model of the ITC and the TC of the composites. AMM assumes that phonons are reflected and refracted at the interface. The smaller the ratio of interface roughness to phonon mean free path, the closer the interface thermal resistance is to the calculated value of the AMM model. Hence, the actual situation of the ITC of composites is higher than that predicted by the AMM model. Meanwhile, other factors, such as relative density and interfacial reaction of the graphite/Al composite, are not considered in these models. When there are pores in the composites, it is easy to form microcrack under stress and further reduce the mechanical properties of the composites. Meanwhile, these pores can also affect the heat conduction of phonons and electrons and reduce the thermophysical properties of the composites. It is well known that graphite and Al will react during preparation in high temperature and the TC of the

reaction products Al_4C_3 is very low, which is not benefit for the improvement of the TC of the composites. It should be noted that these predictive models are still very effective to evaluate the effects of various coatings on the ITC and select a desired interface layer for graphite flakes/Al composites.

4 Conclusion

Graphite flakes/Al composites are promising thermal management materials due to high TC and low CTE. In this work, the prediction models based on the AMM and EMA are developed to evaluate the effect of various interfacial coatings on the ITC and the TC of graphite flakes/Al composites. The ITC decreases with the increase in coating thickness. For the composites with Ni, Cr/Cr₇C₃/Cr₃C₂, Si/SiC, Ti/TiC, WC, and Mo/Mo₂C coatings, the ITC is relatively sensitive to coating thickness. With the increase in the thickness of interfacial coatings, the TC of graphite flakes/Al composites decreases. The coatings with a nanoscale thickness are positive to improve the thermal properties of graphite flakes/Al composites. Si/SiC and WC coating can be used as suitable coating materials of the graphite flakes/Al composites from the point of view of improving the TC of the composites; however, the thickness of the coatings still needs to be controlled. The TC of the graphite flakes/Al composites increases as the volume fraction of graphite flakes increases. The TC of the graphite flakes/Al composites increases with the ITC and changes slowly when the ITC increases to a certain extent.

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Conflict of interest: The authors declare that they have no conflict of interest.

Data availability statement: All data included in this study are available upon request by contact with the corresponding author.

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