UDK 004.942:546.62
Original scientific article/Izvirni znanstveni članek

ISSN 1580-2949
Mater. Tehnol.

FINITE ELEMENT SIMULATION OF THERMAL PROPERTIES OF COATED GRAPHITE FLAKE/ALUMINUM MATRIX COMPOSITES

MKE SIMULACIJA TERMIČNIH LASTNOSTI KOMPOZITOV NA OSNOVI Al MATRICE Z VGRAJENIMI GRAFITNIMI KOSMIČI

Hongwei Shi, Xiaoming Du*, Siyu Li, Zhendong Sun

School of Materials Science and Engineering, Shenyang Ligong University, Shenyang 110159, China

Prejem rokopisa – received: 2025-01-19; sprejem za objavo – accepted for publication: 2025-02-24

doi:10.17222/mit.2025.1380

A three-dimensional, multi-scale finite element model was developed to predict the effective thermal conductivity of graphite flake/aluminum composites. The factors influencing the effective thermal conductivity and thermal expansion coefficient of graphite flake/aluminum composites were investigated, including the type and thickness of the coating, temperature, and the volume fraction of graphite flakes. The results indicate that a higher thermal conductivity and reduced coating thickness enhance the thermal conductivity of the composites. Taking into account the thermal expansion behavior and coating costs, it is advisable to select a copper coating with a thickness maintained between 1 μm and 1.5 μm. Temperature fluctuations influence the thermal resistance at the interface and the strength of the bonding. While an increase in the temperature can improve thermal conductivity, excessively high temperatures lead to a significant rise in the thermal expansion coefficient, adversely affecting the overall performance. The optimal temperature is 300 °C. Copper plating can enhance the thermal conductivity of the composites, and a higher volume fraction of graphite is associated with improved overall thermal performance, which aligns with the experimental results.

Keywords: graphite flake/aluminum matrix composites, thermal conductivity, coefficient of thermal expansion, numerical simulation

Avtorji so razvili več-nivojski tri-dimenzionalni model na osnovi metode končnih elementov (MKE) za napoved efektivne toplotne prevodnosti kompozitov z Al matrico v katero so vgrajeni grafitni »kosmiči« (tanke okrogle ploščice). Ugotavljali so faktorje, ki vplivajo na efektivno toplotno prevodnost in koeficiente toplotnega raztezanja izbranih sintetičnih kompozitov, vključno s tipom in debelino prevleke, temperaturo in volumskim deležem grafitnih kosmičev. Rezultati raziskave so pokazali, da višja toplotna prevodnost in zmanjšanje debeline prevleke izboljša toplotno prevodnost kompozitov. Z upoštevanjem toplotnega rastezanja in cene prevleke je priporočljivo izbrati bakreno prevleko debeline med 1,0 μm in 1,5 μm. Nihanje temperature vpliva na termično odpornost na meji s prevleko in njeno trdnost vezave s kompozitom. Medtem, ko naraščanje temperature lahko izboljša toplotno prevodnost, previsoka temperatura vodi do pomembnega povečanja koeficienta toplotnega raztezanja, kar obratno vpliva na kakovost sklopa prevleka-kompozit. Optimalna temperatura je 300 °C. Platiranje z bakrom izboljša toplotno prevodnost kompozitov in višji volumski delež grafita celovito izboljšata termične lastnosti, kar se ujema tudi z eksperimentalnimi rezultati.

Ključne besede: kompoziti, grafitni kosmiči v matrici iz aluminija, toplotna prevodnost, koeficient toplotnega raztezanja, numerična simulacija na osnovi metode končnih elementov (MKE)

1 INTRODUCTION

Emerging industries are driving advances in electronics, with components such as chips evolving towards miniaturization, increased power density, multifunctionality and lightweight designs. An increase in the temperature of electronic devices can significantly shorten the lifespan of chips. The enhancement of heat dissipation has emerged as a pivotal strategy to optimize the performance of devices and systems in the electronics industry. To ensure the thermal stability of heat dissipation materials, it is essential that these materials possess high thermal conductivity (TC), low density, and a coefficient of thermal expansion (CTE) that is compatible with the substrate.[1,2]

*Corresponding author's e-mail:
du511@163.com (Xiaoming Du)

To meet the increasing demand, thermal management materials have evolved into fourth-generation graphite flake/aluminum (GF/Al) composites. The bonding between GFs and the Al matrix at the interface significantly influences the thermal properties of these materials. Surface modification techniques can address related issues, with GF surface coating being a commonly employed method.[3] Recently, extensive research has been conducted on the interface design of GF/Al composites, revealing that the microstructure at the interface has a substantial impact on the TC. For instance, Xue et al. utilized a salt bath plating method to coat silicon carbide (SiC) and obtained a composite with an in-plane TC of 735 $W\cdot m^{-1}\cdot K^{-1}$.[4] Similarly, Huang et al. employed physical vapor deposition for copper plating and discovered that the coating thickness influenced the thermal expansion properties, necessitating careful regulation.[5] Zhou et al. conducted electroless plating with copper (Cu) or nickel (Ni) and found that Cu plating enhanced the TC, whereas Ni plating slightly reduced it, indicating that different coatings can have varying effects on the TC.[6] However, despite some studies on carbides and metal coatings, quantitative effects on the TC and thermal expansion have yet to be thoroughly evaluated and compared.

This study aims to evaluate the TC and CTE of both uncoated and coated graphite flake/aluminum (GF/Al) composites, while also exploring the influencing factors. The TC and CTE of the materials were calculated using the finite element method. The effects of the GF coating type, thickness, temperature, and volume fraction (ranging from 10 to 70 %) of the composites were quantitatively analyzed. Using copper-plated GF/Al composites with varying volume fractions as examples, the feasibility of the simulation was validated in conjunction with experimental results. This study represents a pioneering attempt to conduct a theoretically comprehensive evaluation of the effects of thermal conductivity and thermal expansion on GF/Al composites, a subject that has heretofore been under-explored in the academic literature.

2 MODELS AND METHODS

2.1 Microstructural model and material properties

In the production process, the morphology of GFs is characterized by irregular thin flakes.[7] A small number of GFs may be influenced by other GFs or matrix powders, or they may be oriented at a certain angle relative to the horizontal direction. GFs with a high volume fraction may exist in an incomplete or stacked state within the composite material.[8] Based on the actual morphology and distribution of GFs, and considering the feasibility of the simulation calculation time and mesh generation, the GF model is represented as a thin circular flake. **Figure 1** illustrates the finite element model and the local coating of coated GF/Al composites, in which GFs constitute 60 φ/%. **Figure 1** employs different colors to denote the types of materials: the green section represents the aluminum (Al) matrix, the red section indicates the GFs, and the white section corresponds to the coating.

The GF surface modified coating comprises a metal coating, a carbide modified layer, and other components. The selection of coating materials is primarily based on two considerations: first, the coating material must exhibit a high TC, such as copper (Cu), silver (Ag), and silicon (Si).[9–13] Second, the coating should be thin, as seen with silicon carbide (SiC).[14,15] Although titanium carbide (TiC) has a low TC, it exhibits a high strength, which significantly enhances the reinforcing effect on brittle GFs.[16] Consequently, Cu, Ag, Si, SiC, and TiC coatings have been selected for the analysis. The material properties relevant to this study are presented in **Table 1**.

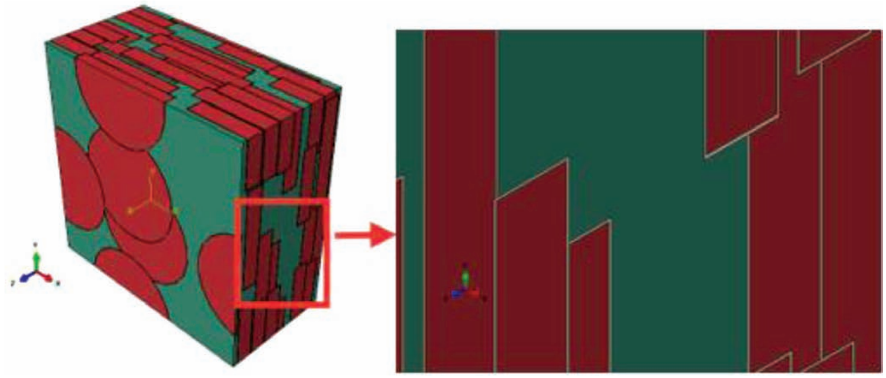

Figure 1: Microscopic model of a composite material with a 60-% graphite volume fraction

Table 1: Material parameters[17–20]

Material parameter		Aluminum matrix	Graphite	Cu	Ag	Si	SiC	TiC
Density (g·cm^{-3})		2.75	2.26	8.9	10.5	2.33	3.2	1.85
Specific heat capacity (J·kg^{-1}·K^{-1})		896	710	390	235	256.1	632	569
Thermal conductivity (W·m^{-1}·K^{-1})	in-plane	185.9	1000	400	428	150	170	36.4
	interlayer		38					
Young's modulus (GPa)	in-plane	69	1020	126	732	136	220	242
	interlayer		36.5					
Shear modulus (GPa)	in-plane	24	440	45	27	79.9	166	186
	interlayer		18					
Poisson's ratio	in-plane	0.33	0.16	0.35	0.38	0.27	2.9	0.19
	interlayer		0.012					
Coefficient of thermal expansion (10^{-6} K)	in-plane	22.8	1	16	19	2.59	2.9	6.4
	interlayer		3.4					

2.2 Heat transfer settings

In the context of a heat transfer analysis, radiation and convective heat transfer are not taken into consideration. The present research employs a three-dimensional steady-state thermal analysis.

Figure 2 illustrates the direction of heat transfer within the composites.

When investigating the TC of the composite material plane, various temperature loads are applied to the upper surface ($y = L$) and the lower surface ($y = 0$) of the composite material model along the Y-axis, while the remaining surfaces are maintained in an adiabatic state. In the study of the out-of-plane TC, the temperature loading surface is altered, while the other parameters remain constant. The governing equation for in-plane heat conduction is as follows:[21]

$$\frac{\partial}{\partial x}\left(\lambda\frac{\partial T}{\partial x}\right)+\frac{\partial}{\partial y}\left(\lambda\frac{\partial T}{\partial x}\right)=0 \quad (1)$$

$$q_x = \lambda\frac{\partial T}{\partial x} \quad (2)$$

$$T\big|_{y=L} = T_{\text{hot}} \quad (3)$$

$$T\big|_{y=0} = T_{\text{cold}} \quad (4)$$

$$\lambda = q^{\text{avg}} \times \frac{L}{\Delta T} \quad (5)$$

Here, q is the heat flux vector; q^{avg} is the average heat flux of the composite material (W/m^2); λ is the equivalent thermal conductivity of the composite material($W \cdot m^{-1} \cdot K^{-1}$); ΔT is the temperature difference (°C) in the direction of heat conduction; and L is the distance (m) between the model's high and low temperature surfaces. The TC of the composite can be determined using the equation provided above.

2.3 Thermo-force coupling setting

The phenomenon in which the volume or length of an object increases with rising temperature is known as thermal expansion. When analyzing thermal-force coupling, considering the transverse isotropy of mechanical properties and the coefficient of thermal expansion of GFs, we assume that the original length is l_0, the initial temperature is T_0, and the change in length after the temperature increases due to ΔT is Δl. The average linear expansion coefficient, α, of the material within this temperature range can be expressed as:

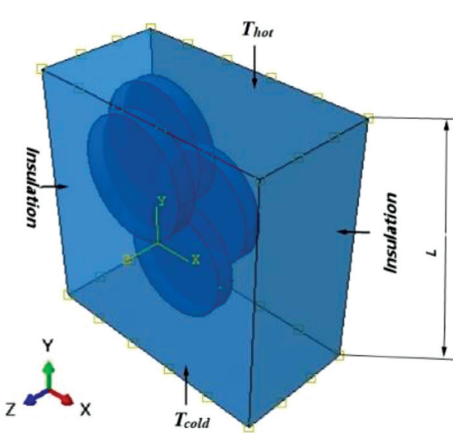

Figure 2: Direction and boundary conditions of heat transfer in composites

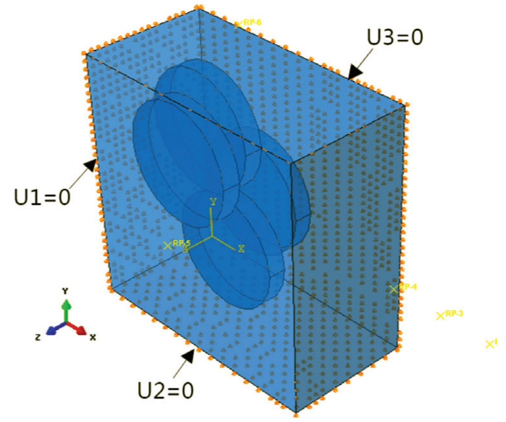

Figure 3: Thermal expansion boundary conditions of composites

$$\alpha = (\Delta l / \Delta T) / l_0 \quad (6)$$

To ensure the continuity of deformation and stress during the thermal expansion performance analysis, periodic displacement boundary conditions were applied to the geometric model.[22] **Figure 3** illustrates the displacements of the three orthogonal boundary surfaces, with restrictions applied in all directions. The initial temperature was room temperature, and the model was then heated from this temperature to 200 °C. The composite's thermal expansion performance was determined by defining the linear expansion coefficient.

3 EXPERIMENTAL PROCEDURES: MATERIALS AND METHODS

3.1 Materials and the coating on GFs

The GFs (purity >99.9 %, mesh size of –50, purchased from Greifa Carbon Material Co., Ltd., Nanjing, China) served as the filler material, while the 6061 aluminum alloy (particle size of 2–10 μm, purchased from Hongyu Material Co., Ltd., Beijing, China) served as the matrix material. The process of copper plating on the GF surface is as follows: Ultrasonic-cleaned, sensitized, and activated GFs were immersed in a metal salt solution containing copper sulfate as the reducing agent, ethylenediamine tetra acetic acid disodium as the complexing agent, and alcohol as the stabilizer. The pH value was adjusted using concentrated sodium hydroxide. The mixture underwent a water bath process, during which the temperature was 75 °C, and it was agitated at a speed of 30 min^{-1}. Subsequently, the electroless copper-plated GFs were removed to be dried, and then they were transferred into a tube furnace for thermal reduction. To prevent oxidation of the surface copper, the electroless copper-plated GFs had to be sealed and stored appropriately.[23,24]

3.2 Fabrication of the composites

The GF/Al composites, containing 10–70 φ/% coated GFs and non-coated GFs, underwent fabrication through the vacuum hot-pressing method. The coated and non-coated GFs were first mixed with 6061 aluminum alloy powder. Subsequently, the mixture was loaded into a F50 mm mold. The vacuum hot press furnace was subjected to a heating rate of 7 °C/min, reaching a temperature of 610 °C, and subsequently pressurized to 50 MPa. The temperature was sustained at 610 °C for a period of one hour. After the heat preservation period, the vacuum was maintained while the furnace cooled down.

3.3 Characterization

Microstructural characteristics were analyzed using scanning electron microscopy (SEM) with an S-3400N model from Japan. The TC of the composites was computed utilizing the equation: $K = \alpha\rho C_p$, where α represents thermal diffusivity, ρ indicates density, and C_p indicates specific heat of the composite. The thermal diffusivity and specific heat were measured under ambient conditions utilizing a Netzsch LFA467 instrument. The density was determined using the Archimedes drainage method.

The linear CTE of the composite was determined using the ZRPY-1600 CTE tester manufactured by Xiangtan Instrument Co., Ltd. (Xiangtan, Beijing). The temperature range of the experiment was from ambient temperature to 200 °C.

4 RESULTS AND DISCUSSION

4.1 Effect of the coating type

Figure 4 shows the heat flow vector diagrams for a coated composite with a GF volume fraction of 10 % and a coating thickness of 1 μm, when heat transfer occurs parallel to the GFs. **Figure 4a** represents the scenario in which the coating material is copper, while **Figure 4b** represents the scenario with silicon carbide as the coating material.

The heat flow vector diagram is primarily utilized to observe the direction and magnitude of heat flow within the microscopic model unit. Observations indicate that the peak heat flux predominantly accumulates within or near the GFs, creating a continuous channel of high heat flow. When heat is transferred from the aluminum matrix to the GFs, minimal scattering is observed at the copper interface. In contrast, a slight scattering effect is evident when passing through the silicon carbide interface. This phenomenon occurs because, during heat conduction, some heat penetrates the interface layer and is transferred into the GFs, while another portion bypasses the interface layer. It is evident that the elevated interfacial thermal resistance of silicon carbide impedes the effective utilization of the GF's superior TC. Consequently, this leads to a diminished thermal conductivity of the composite material.[25]

Figure 5 illustrates the simulation results for the TC and CTE of composites with various coatings, specifically with a GF volume fraction of 10 % and a coating thickness of 1 μm. The findings indicate that the intrinsic properties of the coating significantly influence the overall performance. When the TC of the coating is higher than that of the matrix, the thermal performance of the composite is improved. Conversely, when the TC of the coating is lower than that of the matrix (as observed with materials such as Si, SiC, and TiC), the TC decreases. As Yang et al. noted, the TC of the filler material exceeds that of the matrix, emphasizing the critical role of the interface TC.[26] The CTE order for the coating materials is as follows: Ag > Cu > TiC > SiC > Si, which aligns with the simulation results.

Additionally, the CTE of the GFs in the z-plane is larger. In principle, the CTE of a composite material with an equivalent reinforcement phase is greater in the z-direction. However, contrary to the initial hypothesis, the experimental results reveal an alternative outcome. Zhou et al. suggested that this discrepancy arises from the compressive deformation of the GFs due to the high

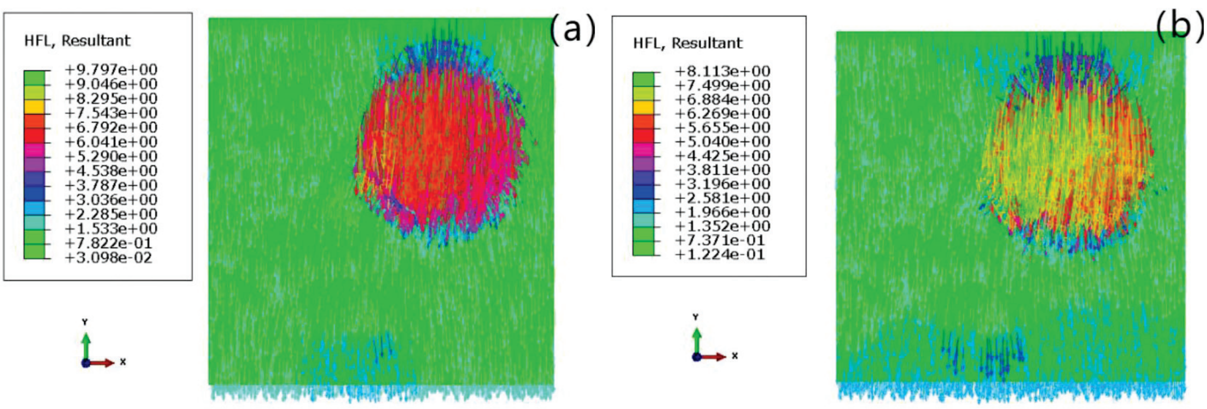

Figure 4: Heat flow vector diagrams of a composite material with different coating elements: a) copper (Cu), b) silicon carbide (SiC)

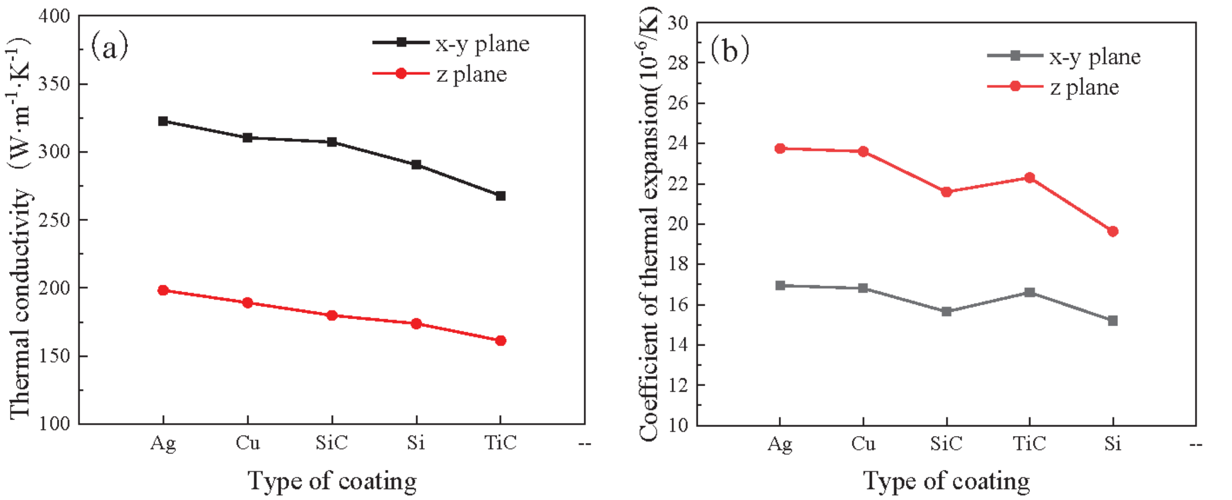

Figure 5: Effect of different coating types on the thermal properties of composite materials: a) thermal conductivity, b) coefficient of thermal expansion

pressure in the vertical direction during molding.[27] The residual stress resulting from cooling and shrinkage further inhibits thermal deformation in this direction. Theoretical calculations confirmed that compressive deformation absorbs most of the thermal expansion, leading to abnormal thermal expansion behavior.

Coatings of copper and silver significantly enhance the TC of the composites. However, it is imperative to consider the material's thermal expansion behavior. When evaluating the properties and costs of coating materials, the composite with a copper coating demonstrates superior overall performance.

4.2 Effect of the coating thickness

The preceding analysis demonstrates that interfacial thermal resistance is influenced by the TC of the composite, and is affected by both the composition and thickness of the coating. The thickness of a GF coating is typically in a range of 0.2–2 µm. For this analysis, a copper-plated composite material model with a 10 % GF volume fraction was selected, with coating thicknesses of (0.2, 0.5, 1, 1.5, and 2) µm.

Figure 6a illustrates that that an increase in the coating thickness is associated with a decrease in the in-plane TC and an increase in the out-of-plane TC. It is found that the TC of the silicon carbide-coated composite is below that of the copper-coated composite, and both exhibit a similar trend. Additionally, the interface layer, characterized by high thermal resistance and low TC, influences these results. Liu et al. validated the model's accuracy by predicting its behavior in accordance with the simulation.[19] **Figure 6b** illustrates the simulation results of the CTE for copper-coated GF/Al composites with varying coating thicknesses (0.2–2 µm) at a GF volume fraction of 10 %. An assessment of the coating thickness was conducted across a temperature range encompassing ambient temperature and a maxi-

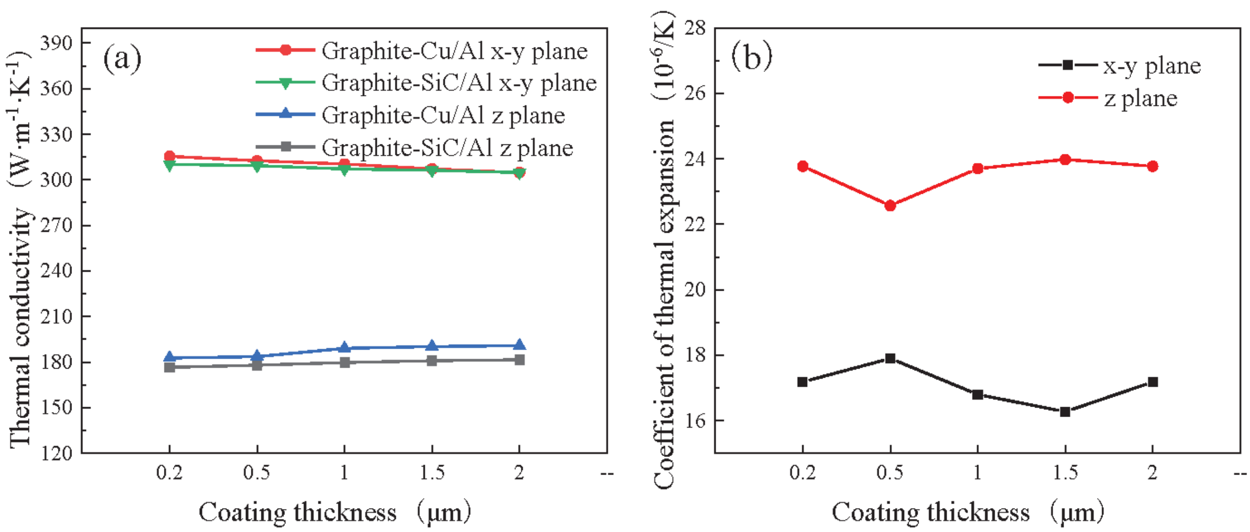

Figure 6: Simulation results: a) thermal conductivity, b) thermal expansion coefficient of composites with various coating thicknesses

mum of 200 °C. The effect of coating thickness on the CTE is significant, as it affects the overall structural properties of the composite. Moreover, it is imperative to emphasize that the thickness of the coating should not exceed 1.5–2 µm. Excessive thickness can introduce internal defects and other factors that compromise the stability of the CTE. Therefore, an optimal coating thickness of 1–1.5 µm is recommended, as it results in excellent thermal expansion performance. In general, copper plating on the graphite surface has a positive effect on the composite's thermal properties.

4.3 Effect of the temperature

The composite model with a GF content of 10 % was selected and tested at temperatures of (25, 100, 300, and 500) °C. To more directly observe temperature-related changes in the heat flux, heat-flux cloud image values were standardized within the same display range.

Figure 7 illustrates the heat-flux cloud map of the copper-plated GF/Al composite at various temperatures, with the volume fraction of GFs set at 10 %. The heat flow is directed parallel to the GFs. The cloud image indicates that the maximum and minimum heat flux values of the composite material fluctuate with the temperature. According to the principle of minimum thermal resistance, heat flow preferentially moves toward regions with lower thermal resistance. The hierarchy of thermal resistance is as follows: Al matrix, copper, and GFs, indicating that the heat flow is directed toward the GFs. When the temperature is below 300 °C, the TC of GFs continues to increase, resulting in a decrease in the thermal resistance of GFs as the temperature rises.[28–29] Consequently, the heat flow becomes more concentrated, resulting in an increased heat flux value with the TC increase of GFs, as illustrated in **Figure 7c**. Conversely, at temperatures exceeding 300 °C, a decrease in the TC of the GFs is observed, which consequently results in a reduction in the heat-flux value on the GFs.

Figure 8 shows the variation in the TC of the composite material with the temperature, both before and after copper plating, while maintaining the same GF volume fraction. It was observed that the in-plane TC of both composites increases with rising temperature but decreases after reaching 300 °C. In contrast, the out-of-plane TC consistently rises with an increase in the temperature. At room temperature, the copper content on the GFs is relatively low, indicating that the GFs serve as the primary heat conductor. The introduction of a copper coating effectively incorporates a material with a superior TC into the system, resulting in a higher internal TC of the copper-coated composite compared to the non-copper-coated composite. However, the TC of copper decreases with increasing temperature, resulting in greater thermal resistance that impedes the heat flow into the GFs. This results in a lower in-surface TC of the copper-coated composite compared to its non-copper-coated counterpart. In the z-plane, heat transfer mainly depends on the matrix and copper, with the TC in the direction of GF thickness being relatively low. As the temperature increases, this TC further diminishes, with the GFs playing a secondary role. This phenomenon can be attributed to the enhanced TC observed after copper plating in comparison to that of the composite without copper plating.

Figure 9 illustrates the stress distribution within the copper-coated GF/Al composite at different temperatures. As the temperature rises, the stress changes due to thermal expansion increase significantly, with the maximum stress potentially escalating several times. The region exhibiting the most significant variation in stress is primarily concentrated at the interface of the composite

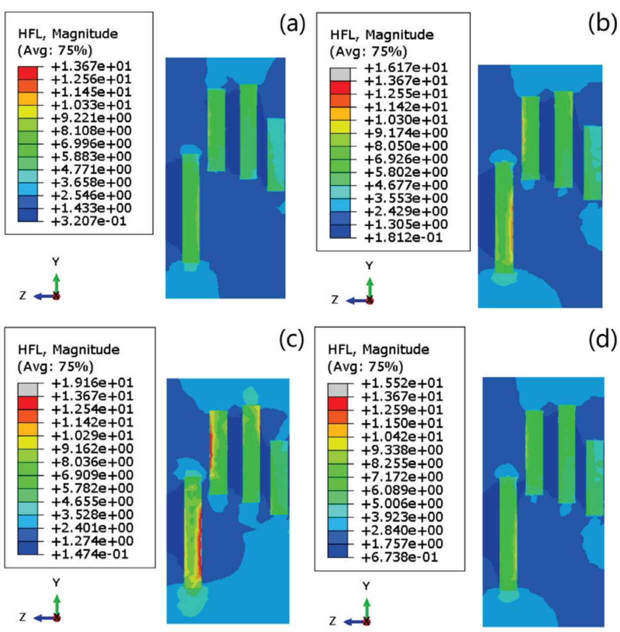

Figure 7: Heat-flux cloud images of the copper-coated GF/Al composite at different temperatures: a) 25 °C, b) 100 °C, c) 300 °C, d) 500 °C

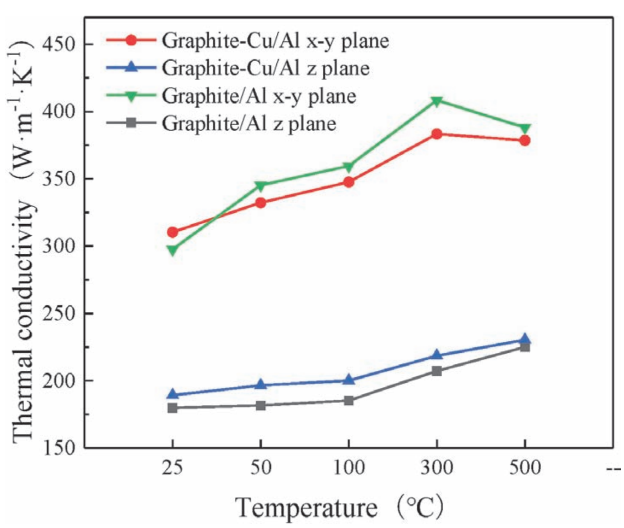

Figure 8: Thermal conductivity of composites at different temperatures

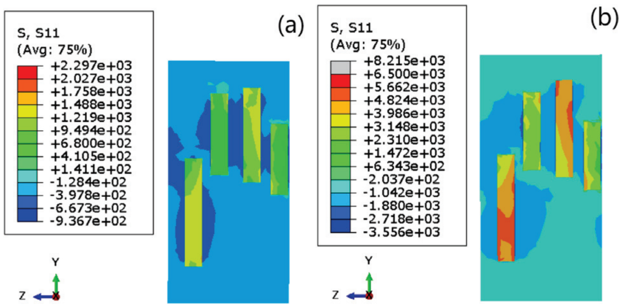

Figure 9: Stress distribution of composites at different temperatures: a) 200 °C, b) 500 °C

material. This observation suggests that the copper-coated GFs impose a limiting effect on the expansion of the matrix.

Figure 10 illustrates the results of the CTE simulation for both copper-coated and uncoated composites with a 10-% GF volume fraction at various temperatures. As the temperature increases, there is a noticeable rise in the coefficient of thermal expansion (CTE) of the coated composite. This behavior is consistent with that observed for the uncoated copper composite. Furthermore, the CTE of the z-plane in the copper-coated composite does not differ from that of the uncoated composite, suggesting that thermal expansion in the z-plane is not significantly influenced by temperature changes. Additionally, the CTE of the copper-coated GF/Al composite is lower compared to that of the uncoated composite. This finding indicates that the CTE of the composites can be further reduced through the surface modification of the GFs.

Although the TC of the copper-coated GF/Al composite improves with increasing temperature, the CTE of the composite also continues to rise. If the temperature becomes excessively high, it may not meet the material's performance requirements. Therefore, it is advisable to maintain the temperature as close to 300 °C as possible.

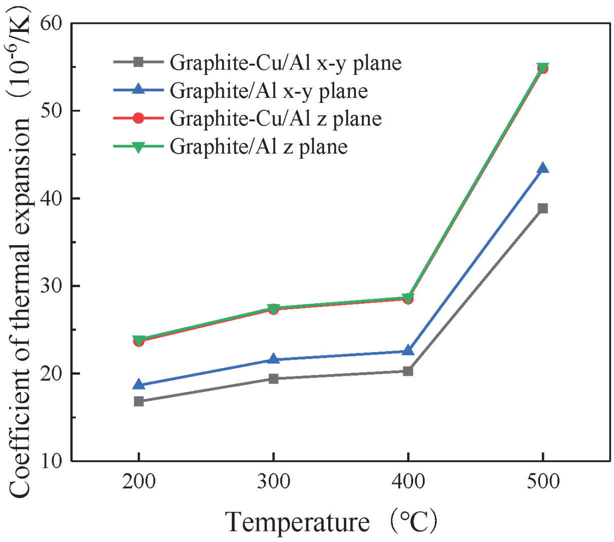

Figure 10: Simulation results for the effect of temperature on the thermal expansion coefficient of copper-coated and uncoated composites

4.4 Effect of the GF volume fraction

The volume proportion of GFs in the composite significantly influences its thermal conductivity. To investigate these effects, a model was selected, including both copper-coated and uncoated composites. The volume fraction of the enhancement phase varied from 10 % to 70 %.

Figure 11 shows the heat-flux diagrams of copper-coated and uncoated composites with GF volume fractions of 30 % and 50 % in the x–y plane. The increase in the volume fraction of GFs resulted in the adjacent GFs interacting with one another, thereby expanding the maximum heat-flux area. The overall heat-flux value for copper-coated GFs is higher than that for uncoated GFs, indicating that the presence of the coating reduces the heat loss at the GF-aluminum matrix interface. This enhancement allows for a greater heat transfer through the GFs, ultimately improving the overall TC.

Figure 12 illustrates a comparison of the TC simulation results for both uncoated and coated composites. It is evident that the TC of the composites exhibits a positive correlation with the GF content in the *x–y* plane, while demonstrating a negative correlation with the GF content in the z-plane. Following surface modification, the TC of both surfaces is improved.

Under the same integral number of the GF/Al composite, the TC at the surface of the copper plating is increased by a certain proportion compared to that without copper plating. It was demonstrated that the gain effect of the out-of-plane TC following copper plating is greater than that of the in-plane TC. This phenomenon can be attributed to the fact that when heat flow is transferred perpendicularly to the GFs, the copper coating exerts a predominant influence. Conversely, when heat flow is transferred parallel to the GFs, the graphite content becomes the primary factor governing the process. This accumulation enhances the z-plane coating and improves the TC in the thickness direction. Consequently,

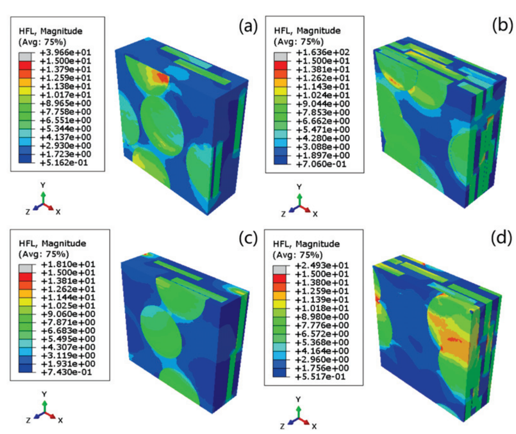

Figure 11: Heat-flux cloud images of copper-coated and uncoated composites with different GF volume fractions: a) coated – 30 %, b) coated – 50 %, c) uncoated – 30 %, d) uncoated – 50 %

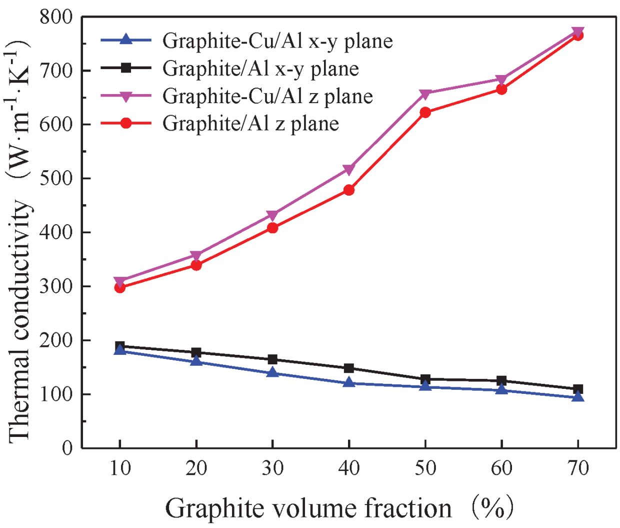

Figure 12: Simulation values of the thermal conductivity for composites with different graphite volume fractions

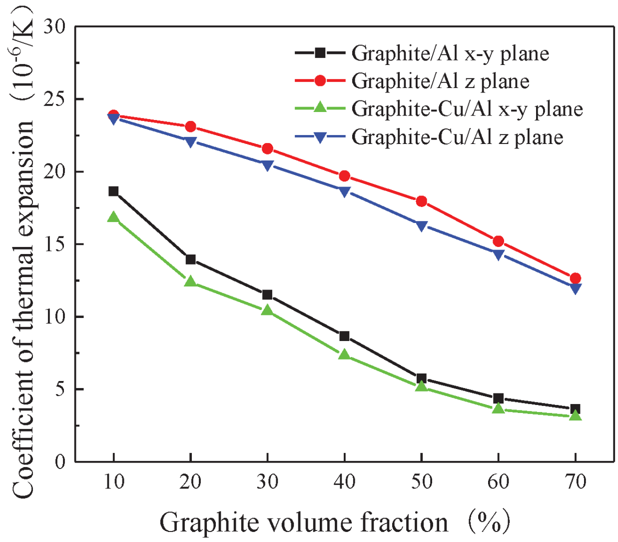

Figure 14: Simulation values of the CTE for composites with different graphite volume fractions

the influence of the copper coating in the thickness direction is more pronounced.

It is evident that the deformation behavior is inconsistent at different temperatures due to the differences in the CTE and mechanical properties among the graphite flakes, the aluminum matrix, and the copper coating. **Figure 13** illustrates the stress distribution in composite materials with 30-% and 50-% GF volume fractions. The CTE of interfacially bonded GFs is lower than that of the aluminum matrix, leading to negligible volume changes in response to increasing temperatures. The limitation on the expansion of the aluminum matrix induces tensile stress, which arises from interfacial tension. As the temperature increases, the aluminum matrix experiences compressive forces exerted by the GFs, while the GFs are subjected to tensile forces from the aluminum matrix. Consequently, the GFs restrict the free expansion of the aluminum matrix; therefore, a higher volume fraction of GFs results in greater constraint.

Figure 14 illustrates the simulation results of thermal expansion for both uncoated and coated composite materials. It is evident that the CTE of the composites de-

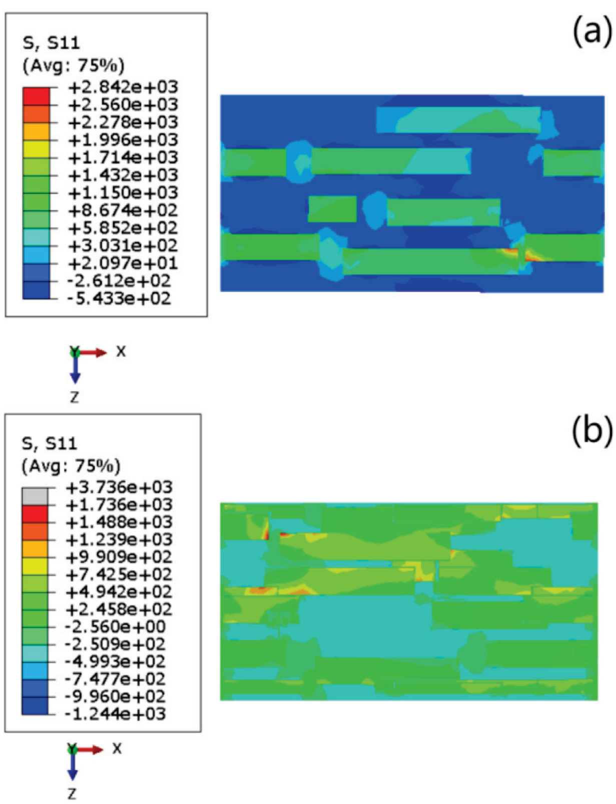

Figure 13: Stress images for composites with different copper-coated graphite volume fractions: a) 20 %, b) 50 %

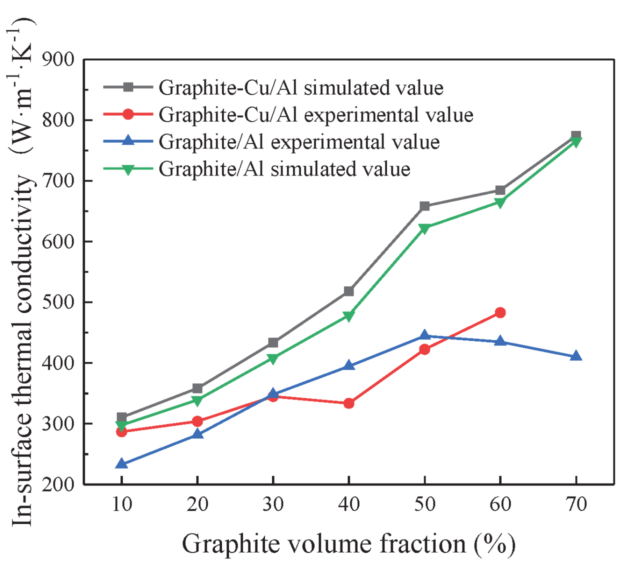

Figure 15: Simulation values of the thermal conductivity for composites with different graphite volume fractions

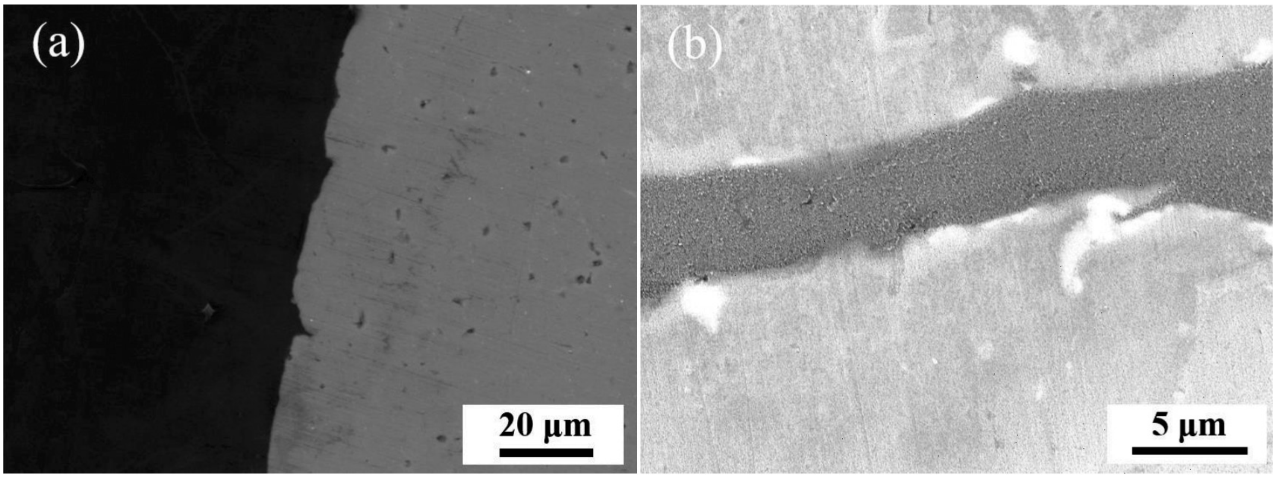

Figure 16: SEM images of composites with a graphite volume fraction of 10 %: a) without copper coating, b) with copper coating

creases proportionally as the GF volume fraction increases. Furthermore, copper plating on the GF surfaces was shown to enhance the bond strength and reduce the thermal deformation behavior of the composites.

4.5 Comparison of simulation and experiment

When preparing Al-matrix composites with a higher volumetric proportion of GFs, the GFs tend to obstruct the flow of the matrix powder, reducing the material's density, and negatively impacting the quality of the chemical coating.[8] Consequently, only composite material with a 60-% volume fraction of GFs was prepared. Since the thermal conductivity parallel to the GF direction is higher than that perpendicular to the GF direction, only the in-plane TC of the composite is considered.

Figure 15 presents the experimental and simulation results for the TC of both uncoated and copper-coated GF/Al composites. It is evident that as the GF content increases, the TC also increases, with copper plating further enhancing this property. To directly observe the interface bonding, scanning electron microscopy (SEM) was employed to analyze the interfaces between the different phases.

Figures 16a and **16b** illustrate the interfaces of uncoated and copper-coated GF/Al composites with a 10-% GF volume fraction, respectively. These figures clearly demonstrate that after copper plating, the GFs are tightly bonded to the matrix, resulting in a smooth interface free of voids and defects. In comparison to the uncoated interface, the bonding in the copper-coated interface is significantly enhanced, improving the thermal properties of the composite.

The TC of the uncoated GF/6061 Al composite generally aligns with experimental results, except at high volume fractions (>50 %). In the simulations, the GF structure is idealized, and the treatment of pores and interfaces is inadequate, leading to significant deviations in the heat flow obstruction at higher contents. Additionally, the high GF volume fraction in the experiments tends to deform, affecting the heat flow and resulting in reduced data accuracy. Furthermore, it was observed that upon exceeding the GF volume fraction of 20 %, the in-plane TC of certain copper-coated composites does not increase. This phenomenon occurs because the Cu coating introduces multiple interfaces; as the GF content increases, so does the number of interfaces and defects. This leads to increased phonon scattering from these interfaces and defects, thereby prolonging the temperature transfer time and reducing the TC.[31] Surface modifications, which may be caused by the GF orientation, interfacial thermal resistance, interfacial impurities brought on by plating oxidation, and porosity defects that were missed in the simulation, are the reasons for the discrepancies between the simulated and experimental values of the TC of the two composites. However, the overall trend remains consistent, validating the conclusions drawn.

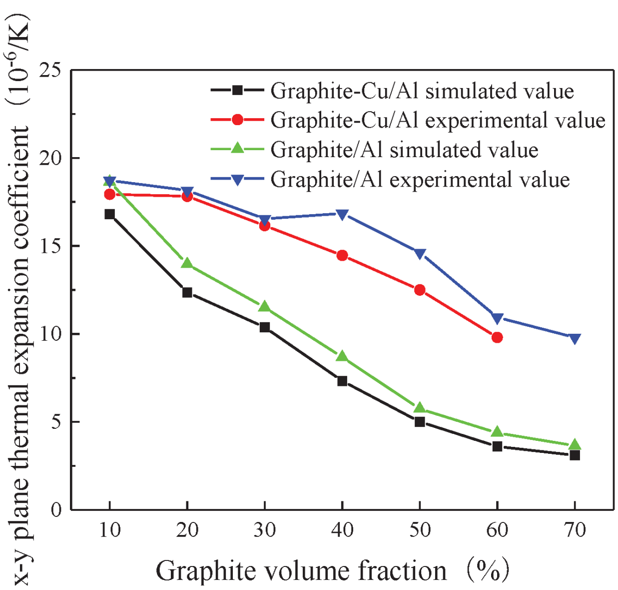

Figure 17: Comparison of thermal expansion coefficients for the uncoated and copper-coated GF/Al composites with different graphite volume fractions

Figure 17 presents the experimental and simulation results for the CTEs of the *x–y* planes of the composites with different volume fractions of copper-coated GFs.

It can also be observed from the diagram that the CTE of the composites is reduced following the GF modification. The improved interfacial bonding achieved through the coating enhances the stress-strain transfer, thereby increasing the constraint of the GFs on the Al matrix and reducing interfacial porosity. This reduction in porosity helps to mitigate the thermal expansion caused by temperature fluctuations.[32] Consequently, the overall thermal expansion of the material decreases as the expansion capacity of the pores increases, resulting in the CTE of the copper-coated GF/Al composites being greater than that of natural GFs. Additionally, the simulation results are lower than the experimental values, likely due to the non-ideal arrangement of the GFs and the high CTE in the z-direction. Nevertheless, a consistent trend is evident across both experimental and simulated results. Despite the differences in the TC and CTE between uncoated and copper-coated GF/Al composites, the overall patterns remain consistent. Thus, it can be concluded that the numerical finite element method is an effective approach for predicting thermal properties. Although quantifying the CTE of the z-plane through a finite element analysis presents challenges, achieving a low CTE remains a primary objective. This composite configuration, which incorporates various volume fractions of GFs into an Al matrix, satisfies the requirements for high TC and low expansion, making it well-suited for the desired material properties. By optimizing the design of the composite structure, it is possible to achieve an ultra-low CTE in the z-direction.

5 CONCUSIONS

This paper investigates the effects of the coating type and thickness, GF volume fraction and temperature on the TC and CTE of coated GF/Al matrix composites through numerical simulations. The conclusions derived from the simulations were subsequently validated through experimental procedures. For coated GF/Al composites, it is advisable to select a coating with a higher intrinsic TC than that of the substrate. A thicker coating may result in increased interfacial thermal resistance, which adversely affects the heat transfer and thermal expansion. Balancing both performance and cost, a copper coating with a thickness of 1–1.5 μm is deemed optimal. It is evident that temperature exerts a substantial influence on the thermophysical properties of the composites. As the temperature increases, the thermal conductivity also rises; however, it should not be elevated excessively, thus preventing a high coefficient of thermal expansion. The optimal temperature is approximately 300 °C. A higher GF volume fraction results in an increased in-plane TC but decreased out-of-plane TC and CTE. Applying a copper coating to the GFs is beneficial, and increasing the GF volume can enhance the overall performance, provided it does not lead to a reduction in effectiveness. Compared to the experimental results, the simulation outcomes were slightly higher due to the ideal distribution and shape of GFs, as well as the imperfect configuration of pores, interfaces, and residual stresses. Nevertheless, the overall trend remained consistent, demonstrating that numerical simulation technology is both feasible and reliable. This approach can serve as an invaluable reference for designing the thermal properties of composites.

Acknowledgment

This research was funded by the Liaoning Provincial Applied Basic Research Project (2023JH2/101300233) and Basic Research Projects of Higher Education Institutions in Liaoning Province (JYTZD20230004, JYTMS20230193) in 2023.

6 REFERENCES

[1] Q. B. Chi, X. M. Du, Z. D. Sun, S. Y. Li, F. G. Liu, The Effect of Graphite Flake Volume Fraction on the Thermophysical Proper Ties of Graphite Flake/Aluminum Composites, Advanced Engineering Materials, 26 (**2024**) 7, 2301836, doi:10.1002/adem.202301836

[2] X. H. Qu, L. Zhang, W. U. Mao, S. B. Ren, Review of metal matrix composites with high thermal conductivity for thermal management applications, Progress in Natural Science: Materials International, 21 (**2011**) 3, 189–197, doi:10.1016/s1002-0071(12)60029-x

[3] S. Ren, Q. Hong, J. Chen, X. He, X. Qu, The influence of matrix alloy on the microstructure and properties of (flake graphite + diamond)/Cu composites by hot pressing, Journal of Alloys and Compounds, 652 (**2015**), 351–357, doi:10.1016/j.jallcom.2015.08.191

[4] C. Xue, H. Bai, P. F. Tao, J. W. Wang, N. Jiang, S. L. Wang, Thermal conductivity and mechanical properties of flake graphite/Al composite with a SiC nano-layer on graphite surface, Materials & Design, 108 (**2016**), 250–258, doi:10.1016/j.matdes.2016.06.122

[5] Y. Huang, Y. Su, X. Guo, Q. Guo, Q. Ouyang, G. Zhang, D. Zhang, Fabrication and thermal conductivity of copper coated graphite film/aluminum composites for effective thermal management, Journal of Alloys and Compounds, 711 (**2017**), 22–30, doi:10.1016/j.jallcom.2017.03.233

[6] C. Zhou, G. Ji, Z. Chen, M. Wang, A. Addad, D. Schryvers, H. Wang, Fabrication, interface characterization and modeling of oriented graphite flakes/Si/Al composites for thermal management applications, Materials & Design, 63 (**2014**), 719–728, doi:10.1016/j.matdes.2014.07.009

[7] Q. Liu, X. B. He, S. B. Ren, C. Zhang, L. Ting-Ting, X. H. Qu, Thermophysical properties and microstructure of graphite flake/copper composites processed by electroless copper coating, Journal of Alloys and Compounds, 587 (**2014**), 255–259, doi:10.1016/j.jallcom.2013.09.207

[8] J. K. Chen, I. S. Huang, Thermal properties of aluminum–graphite composites by powder metallurgy, Composites Part B: Engineering, 44 (**2013**) 1, 698–703, doi:10.1016/j.compositesb.2012.01.083

[9] C. Guan, Y. Zhao, G. Chen, X. Kai, W. Qian, R. Tao, L. Huang, X. Gao, L. Jin, Synergistic strengthening and toughening of copper coated graphene nanoplates and in situ nanoparticles reinforced AA6111 composites, Materials Science and Engineering: A, 822 (**2021**), 141661, 10.1016/j.msea.2021.141661

[10] N. Trifale, E. Nauman, K. Yazawa, Thermal and mechanical modeling of metal foams for thermal interface application, Journal of Heat Transfer, 138 (**2016**) 7, 072801, doi:10.1115/1.4032957

[11] X. Han, Y. Huang, Q. Gao, M. Yu, X. Chen, High thermal conductivity and mechanical properties of nanotube@Cu/Ag@graphite/aluminum composites, Industrial & Engineering Chemistry Research, 57 (**2018**) 31, 10365–10371, doi:10.1021/acs.iecr.8b01567

[12] X. Han, Y. Huang, Q. Gao, R. Fan, X. Peng, Effects of nanotube content on thermal and mechanical properties of NT@Cu/Ag@GF/Al composites, Journal of Alloys and Compounds, 766 (**2018**), 594–600, doi:10.1016/j.jallcom.2018.06.333

[13] J. M. Molina, A. Rodríguez-Guerrero, E. Louis, F. Rodríguez-Reinoso, J. Narciso, Porosity effect on thermal properties of Al-12 wt% Si/graphite composites, Materials, 10 (**2017**) 2, 177, doi:10.3390/ma10020177

[14] X. Han, Y. Huang, J. Yan, Y. Zhu, X. Gao, Highly thermally conductive nanocomposites synthesized by PVD for thermal management applications, Journal of Alloys and Compounds, 825 (**2020**), 153917, doi:10.1016/j.jallcom.2020.153917

[15] H. Tong, F. Qiu, R. Zuo, P. Shen, X. Cong, J. Liu, Q. Jiang, RETRACTED: The effect and mechanism of alloying elements on Al/SiC interfacial reaction in Al melt, Applied Surface Science, 501 (**2020**) 31, 144265, doi:10.1016/j.apsusc.2019.144265

[16] A. R. Kennedy, S. M. Wyatt, Characterising particle–matrix interfacial bonding in particulate Al–TiC MMCs produced by different methods, Composites Part A: Applied Science and Manufacturing, 32 (**2001**) 3–4, 555–559, doi:10.1016/s1359-835x(00)00052-x

[17] Y. Wu, J. Luo, Y. Wang, G. Wang, H. Wang, Z. Yang, G. Ding, Critical effect and enhanced thermal conductivity of Cu-diamond composites reinforced with various diamond particles prepared by composite electroplating, Ceramics International, 45 (**2019**) 10, 13225–13234, doi:10.1016/j.ceramint.2019.04.008

[18] Y. Liu, W. Li, Y. Cui, Y. Yang, J. Yang, Theoretical analysis of interfacial design and thermal conductivity in graphite flakes/Al composites with various interfacial coatings, Science and Engineering of Composite Materials, 29 (**2022**) 1, 500–507, doi:10.1515/secm-2022-0152

[19] Q. Liu, C. Zhang, J. Cheng, F. Wang, Z. Lv, Y. Liu, X. Qu, Modeling of interfacial design and thermal conductivity in graphite flake/Cu composites for thermal management applications, Applied Thermal Engineering, 156 (**2019**), 351–358, doi:10.1016/j.applthermaleng.2019.04.063

[20] S. S. Sidhu, S. Kumar, A. Batish, Metal matrix composites for thermal management: A review, Critical Reviews in Solid State and Materials Sciences, 41 (**2016**) 2, 132–157, doi:10.1080/10408436.2015.1076717

[21] Y. Wang, X. Du, Q. Chi, Numerical Simulation of Thermal Conductivity of Graphene Reinforced Aluminum Matrix Composites, Journal of Physics: Conference Series, 2459 (**2023**) 1, 012003, IOP Publishing, doi:10.1088/1742-6596/2459/1/012003

[22] S. L. Omairey, P. D. Dunning, S. Sriramula, Development of an ABAQUS plugin tool for periodic RVE homogenisation, Engineering with Computers, 35 (**2019**), 567–577, doi:10.1007/s00366-018-0616-4

[23] X. M. Du, Q. B. Chi, Y. Wang, F. G. Liu, Research on Preparation Process and Properties of Graphite Film/Aluminum Composites, Journal of Shenyang Ligong University, 42 (**2024**) 2, 58–63, doi:10.3969/j.issn.1003-1251

[24] N. Li, X. M. Du, F. G. Liu, Effect of Copper-Plated Graphene on the Structure and Properties of Aluminum-Matrix Composites, Journal of Shenyang Ligong University, 40 (**2021**) 4, 73–87, doi:10.3969/j.issn.1003-1251

[25] Q. Liu, F. Wang, W. Shen, X. Qiu, Z. He, Q. Z. Zhang, Influence of interface thermal resistance on thermal conductivity of SiC/Al composites, Ceramics International, 45 (**2019**) 17, 23815–23819, doi:10.1016/j.ceramint.2019.07.358

[26] W. Yang, K. Peng, J. Zhu, D. Li, L. Zhou, Enhanced thermal conductivity and stability of diamond/aluminum composite by introduction of carbide interface layer, Diamond and Related Materials, 46 (**2014**), 35–41, doi:10.1016/j.diamond.2014.04.007

[27] C. Zhou, W. Huang, Z. Chen, G. Ji, M. L. Wang, D. Chen, H. W. Wang, In-plane thermal enhancement behaviors of Al matrix composites with oriented graphite flake alignment, Composites Part B: Engineering, 70 (**2015**), 256–262, doi:10.1016/j.compositesb.2014.11.018

[28] L. Zhao, J. Tang, M. Zhou, K. Shen, A review of the coefficient of thermal expansion and thermal conductivity of graphite, New Carbon Materials, 37 (**2022**) 3, 544–555, doi:10.1016/j.carbon.2022.06.052

[29] N. Burger, A. Laachachi, M. Ferriol, M. Lutz, V. Toniazzo, D. Ruch, Review of thermal conductivity in composites: Mechanisms, parameters and theory, Progress in Polymer Science, 61 (**2016**), 1–28, doi:10.1016/j.progpolymsci.2016.05.001

[30] K. Dash, S. Sukumaran, B. C. Ray, The behaviour of aluminium matrix composites under thermal stresses, Science and Engineering of Composite Materials, 23 (**2016**) 1, 1–20, doi:10.1515/secm-2013-0185

[31] Y. Zhao, K. Sugio, Y. Choi, S. Gen, Z. Xu, J. Yu, Effect of anisotropic thermal conductivity of graphite flakes and interfacial thermal resistance on the effective thermal conductivity of graphite flakes/aluminum composites, Materials Transactions, 62 (**2021**) 1, 98–104, doi:10.2320/matertrans.mt-m2020071

[32] X. Peng, Y. Huang, X. Han, R. Fan, High volume fraction of copper coated graphite flake\Nitrogen doped carbon fiber reinforced aluminum matrix composites, Journal of Alloys and Compounds, 822 (**2020**), 153584, doi:10.1016/j.jallcom.2019.153584