Structural Modeling and Thermal Conductivity of Graphite Film Reinforced Aluminum Matrix Laminated Composites

Yishi Su*, Jingyu Yang, Zichang Lin, Caihao Qiu, Xushun Chai, Xuyang Liu, He Cao, Xiaozhen Wang and Qiubao Ouyang*

State Key Laboratory of Metal Matrix Composites, School of Materials Science and Engineering, Shanghai Jiao Tong University, 800 Dongchuan Road, Shanghai 200240, China

Abstract: Excellent thermal conductivities of thermal management materials are expected to ensure the timely heat dissipation in lots of engineering applications and electronic devices. High in-plane thermal conductivity of laminated composites has become increasing significant for high energy and power density electronic devices. In this study, the continuous graphite film/aluminum (Gr film/Al) laminated composites were fabricated by vacuum hot pressing. In-plane and out-of-plane thermal conductivity of Gr film/Al laminated composites are tested. Two-dimensional structural models of Gr film/Al laminated composites are established, in which volume fraction, interfacial property, punching zone and orientation angle of Gr films can be controlled according to their actual composite microstructures. The effects of volume fraction and interfacial property on the thermal conductivity of Gr film/Al laminated composites are investigated. Two ways to reduce anisotropy of thermal conductivity are introduction of punching zones and control of Gr orientation, which are verified to be effective. On basis of the analysis above, a good understanding can be brought out for extensive thermal management applications of Gr/Al composites.

Keywords: Graphite film/aluminum composites, Structural modeling, Thermal conductivity, Volume fraction, Interfacial property.

1. INTRODUCTION

Nowadays, the ever-increasing power densities and decreasing transistor dimensions are two important trends of modern semiconductor devices. Effective thermal management materials can be applied to control surrounding temperature and improve device performance [1]. The material owning high integration, low cost and light weight is very promising to miniaturize the electronic equipment [2]. Useful lifetime of electronic devices can be shortened by the produced thermal stresses, while timely heat dissipation can clearly reduce surrounding temperature and release the thermal stresses [3]. In order to elevate the heat dissipation efficiency in mobile electronic devices, high in-plane thermal conductivity (TC) of laminated composites is an effective route [4]. Graphite (Gr) film, graphene film and carbon nanotube film are the promising materials with high in-plane TC, which have attracted lots of research attentions in thermal management applications [5, 6]. Gr films with in-plane TC of ~1500 $W{\cdot}m^{-1}k^{-1}$ has already been commercially used for heat dissipating in electronic chip cooling [7]. Graphene film containing debris-free giant graphene sheets with the in-plane TC of 1940±113 $W{\cdot}m^{-1}k^{-1}$ has been studied, which can be easily integrated into high power flexible devices [8, 9]. Carbon nanotube film with the in-plane TC of 770±10 $W{\cdot}m^{-1}k^{-1}$ has been produced by floating catalyst chemical vapor deposition, which can be suitable for effective heat dissipation applications [10, 11]. Gr film with high in-plane TC processes the thickness of tens of microns and weak mechanical properties, which have largely restricted its fabrication and application as large-scale electronic components [12]. Fortunately, Gr film with high in-plane TC can be extended to fabricate metal matrix composites in thermal management applications [13]. Among the Gr-based composites, the Gr flake reinforced aluminum (Al) matrix composites have attracted lots of research works [14, 15], but the Gr film reinforced Al matrix composites with high in-plane TC are rarely reported [16]. Due to their random orientations and lower densification of Gr flake/Al composites, Gr flakes are difficult to fully achieve the enhancing effect [17]. However, Gr films can be combined with Al foils to fabricate the Gr film/Al laminated composites, in which the volume fraction and orientation angle of Gr films are easily adjusted by the stacking of Gr films and Al foils [18]. For the Gr film/Al laminated composites, the damaged interfacial reaction would easily occur in high-temperature fabrication processing [19]. Thus, the low-temperature fabricating like vacuum hot pressing is a good solid-state way to produce Gr film/Al laminated composites, which can better control the interfacial reaction between Gr films

*Address correspondence to this author at the State Key Laboratory of Metal Matrix Composites, School of Materials Science and Engineering, Shanghai Jiao Tong University, 800 Dongchuan Road, Shanghai 200240, China; Tel: 86-21-34202634; Fax: 86-21-34202749;
E-mail: suyishi@sjtu.edu.cn; oyqb @sjtu.edu.cn

E-ISSN: 2410-4701/22

and Al foils [20]. Until now, besides the previous experimental work, few reports have been shown on the modeling and simulating works on the thermal conductivity of Gr/Al laminated composites [21]. Meanwhile, it is very meaningful to reveal ways to reduce high anisotropy of TC of Gr/Al laminated composites by modeling and simulating to increase the potential of this material for application.

In this work, graphite film/Al laminated composites with high in-plane TC were fabricated by vacuum hot pressing method. Both in-plane and out-of-plane TC of graphite film/Al laminated composites with different volume fractions of Gr films are measured, while the anisotropic TC of laminated composites are simulated and discussed. Based on a structural modeling program where volume fraction, interfacial property, punching zone and orientation angle can be tuned accurately, two-dimensional (2D) structural models of Gr film/Al laminated composites with different volume fractions are established. The effect of interfacial thermal resistance is studied first to give a reasonable value of interfacial heat transfer coefficient. Then, two ways to reduce anisotropy of TC, *i.e.*, introduction of punching zone and control of orientation, are investigated. From the analysis above, a good understanding can be brought into light for the extensive thermal engineering applications of Gr/Al composites.

2. MATERIALS AND MODELING

2.1. Materials and Experimental Tests

In this study, the Gr film/Al laminated composites were fabricated by vacuum hot pressing. Continuous Gr film (99.9% in purity, 29.5 μm in thickness) and pure Al foil (99.6% in purity, 13 μm in thickness) were used as the raw materials. For the laminated composites, the thicknesses of Al foils are selected to 140.0, 52.0, 26.0 and 13.0 μm, while the volume fractions of Gr films are separately determined as 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. %. During the preparing process of Gr film/Al laminated composites, the Gr films were firstly cut into pieces and cleaned by acetone, and alumina on the surfaces of Al foils were washed and eliminated by alkali and acid solutions. Pre-treated Gr films and Al foils were first filled into a graphite mold, and then were hot-pressed in a vacuum furnace at 928 K for 100 min under a constant uniaxial pressure of 45 MPa. After the vacuum hot-pressing, the Gr film/Al laminated composite samples were cooled down with the furnace until room temperature.

TC of the produced Gr film/Al laminated composite was determined by the product of the bulk density, thermal diffusivity and specific heat capacity. The sample density was measured by the Archimedes method. The thermal diffusivity was measured by NETZSCH LFA447 thermal analyzer. The specific heat capacity was measured by differential scanning calorimeter. For the fabricated 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. % Gr film/Al laminated composites, the in-plane TC are separately provided as 380 ± 14.0, 545 ± 9.5, 772 ± 16.1 and 940 ± 19.2 $W\cdot m^{-1}k^{-1}$, while the Out-of-plane TCs are presented as 28.7 ± 1.4, 16.1 ± 0.9, 11.4 ± 0.5 and 7.2 ± 0.2 $W\cdot m^{-1}k^{-1}$ respectively. More details could be provided in our previous work [22].

2.2. Numerical Modeling and Simulation

In order to establish the relation between structure and thermal properties, 2D structural models of Gr film/Al laminated composites, where Gr films and Al foils are stacked in the y direction, are constructed by an independently developed structural modeling program. In these structural models, volume fraction, interfacial property, punching zone and orientation angle of Gr films can be controlled according to the real microstructures of Gr film/Al laminated composites. In this work, the bulk density of Gr films and Al foils are 2.2 and 2.7 $g\cdot cm^{-3}$, while the specific heat capacity of Gr films and Al foils are 710 and 880 $J\cdot kg^{-1}K^{-1}$, respectively. The used in-plane (along *x*-axis) and out-of-plane (along *y*-axis) TC of Gr films are separately 1222.6 and 7 $W\cdot m^{-1}k^{-1}$, and the in-plane and out-of-plane TC of Al foils are all equal to 234 $W\cdot m^{-1}k^{-1}$. During the simulating works, the mesh size is set to 2 μm and the element type is selected to DC2D3 in all cases. In order to simulate the in-plane (out-of-plane) TC of Gr films or Al foils or Gr film/Al laminated composites, the temperature on left (bottom) and right (top) sides of structural models are equal to 373 and 323 K (where ΔT = 50 K), while the other two sides are adiabatic, as is shown in Figure **1**. The length of structural model is denoted as L and the thickness H is dependent on the number of Gr film/Al layer, which is set to 3 here. In a single Gr film/Al layer, the thickness of Gr film is 29.5 μm, while that of Al foil is controlled to change volume fraction of Gr film. Heat flux in the x and y direction are denoted as q_x and q_y, respectively. Therefore, the in-plane and out-of-plane TC of Gr

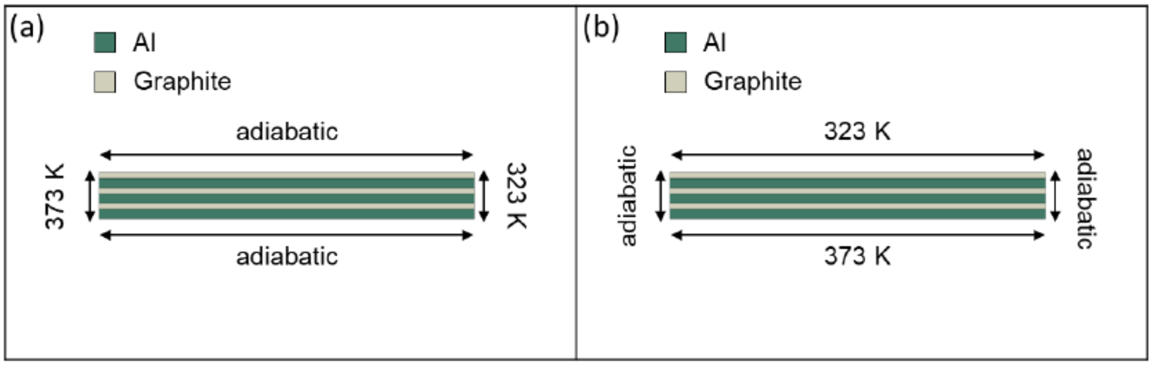

Figure 1: Boundary conditions of (**a**) in-plane TC and (**b**) out-of-plane TC.

film/Al laminated composites, *i.e.* $\lambda_x = q_x/(\Delta T/L)$ and $\lambda_y = q_y/(\Delta T/H)$, can be calculated by Fourier's law.

3. RESULTS AND DISCUSSION

3.1. Effect of Model Size on the Thermal Conductivity of Gr Films and Al Foils

Figure **2** provides the calculated heat flux along *x*-axis and *y*-axis (q_x, q_y) and in-plane and out-of-plane TC (λ_x, λ_y) of Gr films and Al foils with different model lengths. In the structural models, the model lengths L are selected as 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5 and 5 mm, while the model thicknesses H are 29.5 and 13 µm, respectively. In Figure **2**(**a**), along with the model lengths of Gr films L = 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5 and 5 mm, the heat flux along *x*-axis of Gr films (q_x) can be determined as 1.223×10^8, 6.113×10^7, 4.075×10^7, 3.056×10^7, 2.445×10^7, 2.038×10^7, 1.747×10^7, 1.528×10^7, 1.358×10^7 and 1.223×10^7 Wm^{-2}, respectively. Meantime, the in-plane TC (λ_x) of Gr films with different model lengths can be determined as a constant value of 1222.6 Wm^{-1}k^{-1}, which is consistent with the defined value. Figure **2**(**b**) provides the heat flux along *y*-axis (q_y) and out-of-plane TC (λ_y) of Gr films with varying model lengths. Although the model lengths of Gr films are different, the heat flux along *y*-

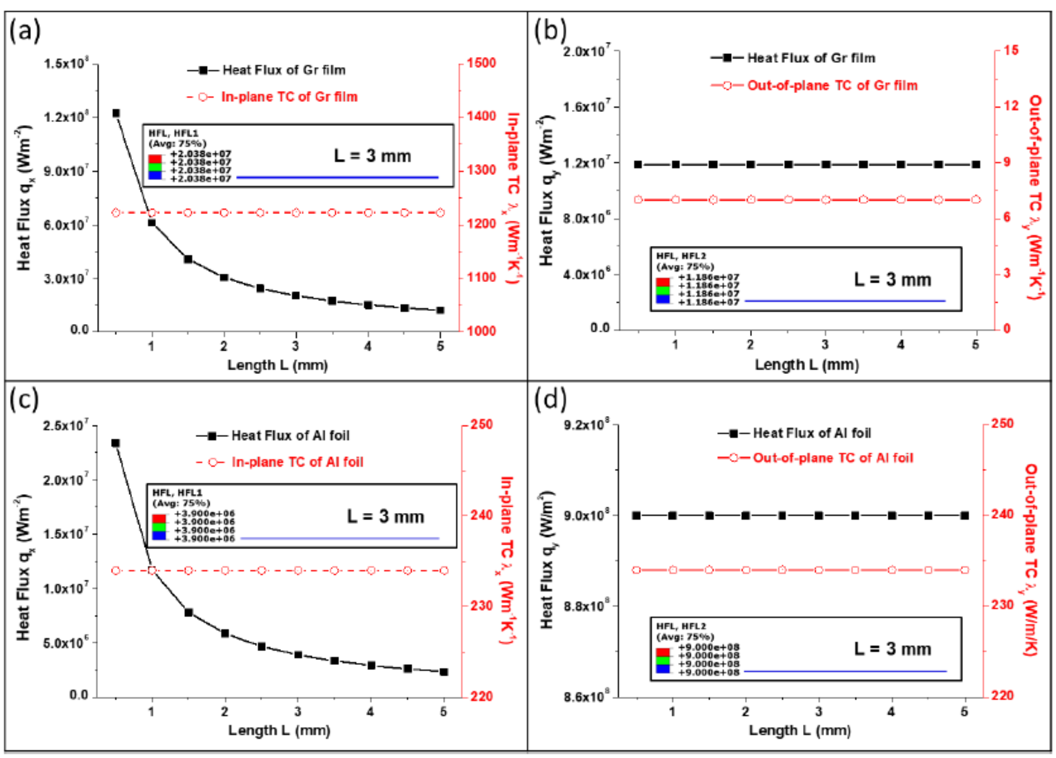

Figure 2: Heat flux and thermal conductivity of Gr films with varying model lengths: (**a**) along x-axis and (**b**) along y-axis; those of Al foils with changing model lengths: (**c**) along x-axis and (**d**) along y-axis.

axis (q_y) and out-of-plane TC (λ_y) are almost kept constant with the values of 1.186×10^7 Wm^{-2} and 7 $Wm^{-1}k^{-1}$ respectively. The calculated out-of-plane TC is also in agreement with the defined value.

In Figure **2**(**c**), along with the model lengths of Al foils L = 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5 and 5 mm, the heat flux along *x*-axis of Al foils (q_x) can be determined as 2.340×10^7, 1.170×10^7, 7.800×10^6, 5.850×10^6, 4.680×10^6, 3.900×10^6, 3.343×10^6, 2.925×10^6, 2.600×10^6 and 2.340×10^6 Wm^{-2}, respectively. Meantime, the in-plane TC (λ_x) of Al foils with different model lengths can be determined as a constant value of 234 $Wm^{-1}k^{-1}$. Figure **2**(**d**) provides the heat flux along *y*-axis (q_y) and out-of-plane TC (λ_y) of Gr films with varying model lengths. Although the model lengths of Gr films are different, the heat flux along *y*-axis (q_y) and out-of-plane TC (λ_y) are almost kept constant with the values of 9.0×10^8 Wm^{-2} and 234 $Wm^{-1}k^{-1}$, respectively. Both in-plane and out-of-plane TCs of Al are in consistence with the defined material parameters. Based on the simulated results, it can be indicated that the model lengths of Gr films and Al foils hardly produce effects on the in-plane and out-of-plane TC of Gr films and Al foils, which is set to 3 mm in the following simulations. At the same time, it is confirmed that the seriously anisotropic TC exists in the Gr films, while the Al foils own the isotropic TC.

3.2. Effects of Interfacial Property and Volume Fraction on the Thermal Conductivity of Gr Film/Al Composites

Interfacial thermal resistance and volume fraction of Gr films could produce effects on the thermal conductivity of Gr film/Al laminated composites. Heat transfer coefficient (α) is commonly used to describe the interfacial thermal resistance at Gr film-Al interfaces, while volume fraction of Gr films is tuned by changing the thickness of Al foil in the Gr film/Al layer. 55 values of heat transfer coefficient at Gr film-Al interfaces ranging from 1×10^4 to 1×10^{10} $W\cdot m^{-2}k^{-1}$ are selected to study the effects of interfacial thermal resistance, while the considered volume fractions of Gr films are 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. %. Figure **3**(**a**) and (b) presents the in-plane and out-of-plane TC (λ_x,λ_y) of Gr film/Al laminated composites with different heat transfer coefficients and volume fractions of Gr films. In Figure **3**(**a**), with the heat transfer coefficient ranging from 1×10^4 to 1×10^{10} $W\cdot m^{-2}k^{-1}$, the in-plane TC of Gr film/Al laminated composites with a certain volume fraction of Gr films keeps almost constant, indicating that heat transfer coefficient at Gr film-Al interfaces has no effect on the in-plane TC of Gr film/Al laminated composites. This is because Gr film-Al interface is parallel to the direction of heat flux and thus, heat has no need to transfer across the interface. The in-plane TC of Gr film/Al laminated composites with volume fractions of 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. % are 406.4, 595.6, 763.6 and 908.1 $W\cdot m^{-1}k^{-1}$, respectively, showing an obvious increasing trend caused by more contributions of Gr films. In Figure **3**(**b**), the out-of-plane TC of Gr film/Al laminated composites with a certain volume fraction of Gr films gradually increases with increasing heat transfer coefficient. When the heat transfer coefficient is below 1×10^7 $W\cdot m^{-2}k^{-1}$, the out-of-plane TC of Gr film/Al laminated composites sharply increase from near zero. However, when the heat transfer coefficient is above 1×10^7 $W\cdot m^{-2}k^{-1}$, the out-of-plane TC of Gr film/Al laminated composites nearly keep the constant values. Besides, higher volume fraction of Gr films decreases the out-of-plane TC of Gr

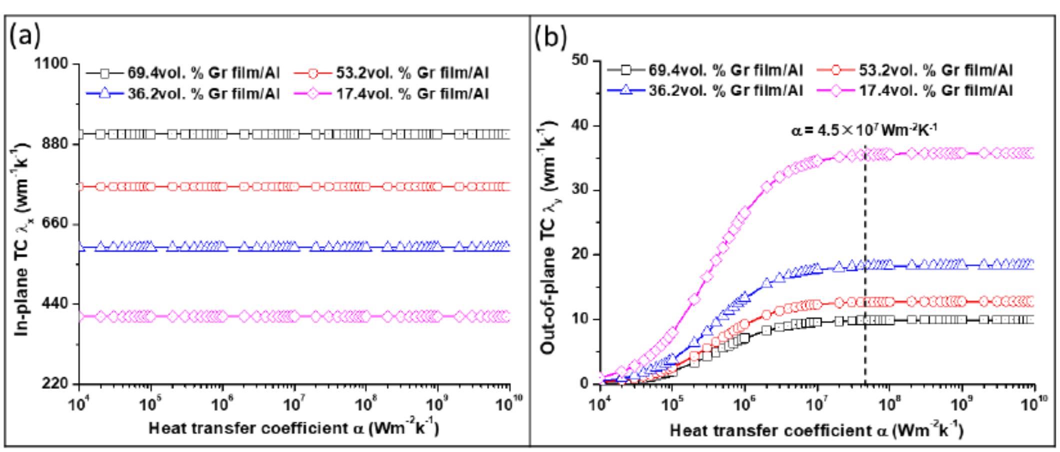

Figure 3: Thermal conductivities of Gr film/Al laminated composites with different heat transfer coefficient at Gr film-Al interfaces and different volume fractions of Gr films: (**a**) in-plane TC and (**b**) out-of-plane TC.

film/Al laminated composites, which is due to pretty low out-of-plane TC of Gr. At small value of heat transfer coefficient, large interfacial thermal resistance is the main factor in restricting the out-of-plane TC, leading to an obvious effect of heat transfer coefficient on the out-of-plane TC. As heat transfer coefficient becomes larger, interfacial thermal resistance is too small to influence the out-of-plane TC, resulting in a steady value of out-of-plane TC. In practice, researchers usually assume that the Gr film/Al interface is good for heat conduction and take 4.5~5$\times 10^7$ W·m^{-2}k^{-1} as the value of heat transfer coefficient at Gr film/Al interface, which is in consistence with the results in this work. Therefore, heat transfer coefficient is set to 4.5$\times 10^7$ W·m^{-2}k^{-1} for the subsequent research and the corresponding simulated TCs are compared with experimental values in Table **1**, which shows good agreement between simulation results and experiments and great anisotropy in Gr film/Al laminated composites.

In a word, the heat transfer coefficient at Gr film-Al interfaces and volume fraction of Gr films both play a key role in the TC of Gr film/Al laminated composites. For the Gr film/Al laminated composite with a certain volume fraction of Gr films, the in-plane TC shows very few changes with the varying heat transfer coefficient, while the out-of-plane TC shows first sharp increases and then keep constant with the raising heat transfer coefficient. Increasing volume fraction of Gr films increases the in-plane TC but deceases the out-of-plane TC of Gr film/Al laminated composites.

3.3. Effect of Punching Zones on the Thermal Conductivity of Gr Film/Al Composites

On basis of the severe thermal conductivity anisotropy of Gr film/Al laminated composites and growing requirement for high-perforce thermal management materials, the punching zone method is considered to effectively balance the in-plane and out-of-plane TC of Gr film/Al laminated composites. In this work, attempts to punch zones in Gr films and replace with Al matrix are possible to improve the in-plane and out-of-plane TC of Gr film/Al laminated composites. The volume fraction of punching zones in Gr films is defined as the punching zone rate (β), which is considered to study the effect of punching zones on the in-plane and out-of-plane TC of Gr film/Al laminated composites. Besides, punching zone number may also affect TCs of Gr film/Al laminated composites. Figure **4** shows TCs and heat flux distributions of Gr film/Al laminated composites with different punching zone rates and punching numbers. Figure **4**(**a**) shows the in-plane TC of Gr film/Al laminated composites with varying punching zone rates. Along with the punching zone rate increasing from 10% to 70%, the in-plane TC of Gr film/Al laminated composites with a certain volume fraction of Gr films gradually reduces. As to the Gr film/Al laminated composites with Gr volume fractions of 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. %, the in-plane TC of Gr film/Al laminated composites with the punching zone rate β of 10% and punching zone number of 1 are calculated as 362.1, 490.5, 596.1 and 692.8 W·m^{-1}k^{-1}, while those of Gr film/Al composites with the punching zone rate β of 70% and punching zone number of 1 are provided as 262.7, 280.6, 290.9 and 298.2 W·m^{-1}k^{-1}. It shows that the in-plane TC of Gr film/Al laminated composites generate nonlinear decreases with the increasing punching zone rates. Besides, although the punching zone numbers are different, the in-plane TC of Gr film/Al laminated composites with a certain volume fraction of Gr films and a certain punching zone rate are close to each other, which is caused by insignificant effect of interfacial thermal resistance. Figure **4**(**b**) presents the heat flux along *x*-axis of 69.4vol. % Gr film/Al laminated composites, where the punching zone numbers are 1, 2, 3 and the punching

Table 1: Thermal Conductivities and Volume Fractions of Gr Film/Al Laminated Composites

Materials	Thermal conductivity (Wm^{-1}k^{-1})				Layer thickness (μm)		Content (vol. %)
	In-plane TC		Out-of-plane TC		Gr film	Al foil	Gr film
	Num.	Exp.	Num.	Exp.			
Gr film/Al	908.1	940$\pm$19.2	9.9	7.2$\pm$0.2	29.5	13.0	69.4
Gr film/Al	763.6	772$\pm$16.1	12.7	11.4$\pm$0.5	29.5	26.0	53.2
Gr film/Al	595.6	545$\pm$9.5	18.2	16.1$\pm$0.9	29.5	52.0	36.2
Gr film/Al	406.4	380$\pm$14.0	35.5	28.7$\pm$1.4	29.5	140.0	17.4

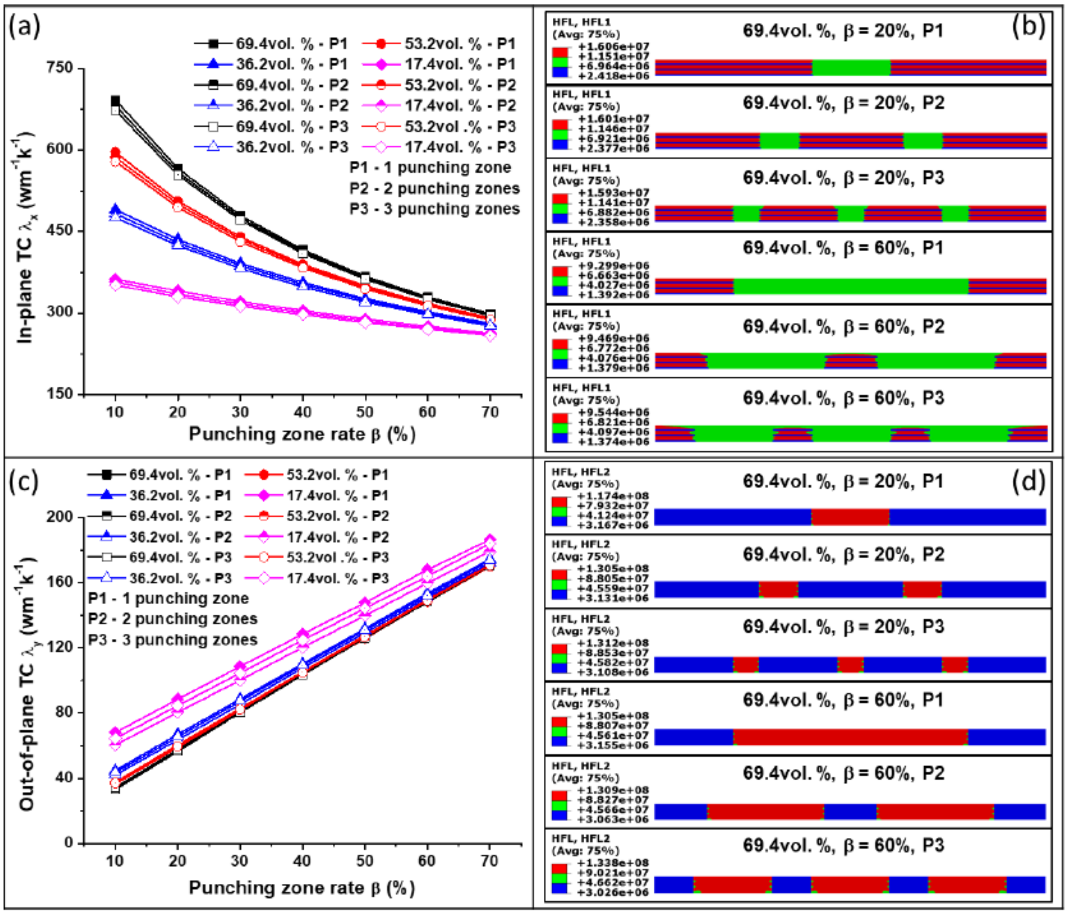

Figure 4: Thermal conductivity and heat flux distributions of Gr film/Al laminated composites with different punching zone rates and numbers: (**a**) in-plane TC and (**b**) heat flux along *x*-axis; (**c**) out-of-plane TC and (**d**) heat flux along *y*-axis.

zone rates are 20%, 60%, respectively. The values of heat flux along *x*-axis in the punched Al zones are located between those of Gr films and Al foils so as to reduce the in-plane TC. Figure **4**(**c**) shows the out-of-plane TC (λ_y) of Gr film/Al laminated composites with varying punching zone rates and punching zone numbers. As punching zone rate increases from 10% to 70%, the out-of-plane TC of Gr film/Al laminated composite with a certain volume fraction of Gr films exhibits nearly linear increases. In respect of the Gr film/Al laminated composites with Gr volume fractions of 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. %, the out-of-plane TC of Gr film/Al laminated composites with the punching zone rate β of 10% and punching zone number of 1 are calculated as 60.3, 41.8, 36.3 and 32.9 W·m^{-1}k^{-1}, while those of Gr film/Al composites with the punching zone rate β of 70% and punching zone number of 1 are presented as 179.3, 172.3, 169.8 and 170.3 W·m^{-1}k^{-1}, respectively. In addition, even though the punching zone numbers are different, the out-of-plane TC of Gr film/Al laminated composites with a certain volume fraction of Gr films (except for the case of 17.4vol. %) and a certain punching zone rate are very close to each other. Figure **4**(**d**) gives the heat flux along *y*-axis of 69.4vol. % Gr film/Al laminated composites, in which the punching zone numbers are 1, 2, 3 and the punching zone rates are 20%, 60%, respectively. The heat flux in punching zone is higher than that in other places, leading to the increase in out-of-plane TC. Based on the analysis above, the TC anisotropy of Gr film/Al laminated composites can be effectively reduced by punching zone method.

3.4. Effect of Orientation Angle on the Thermal Conductivity of Gr Film/Al Composites

In view of large difference between the in-plane and out-of-plane TC of Gr film/Al laminated composites, the relation between thermal conductivity and laminate orientation angle of Gr film/Al laminated composites is very important to extend their potential applications. The angle between the laminate direction and *x*-axis is defined as the laminate orientation angle (θ) to be used to study the effect of laminate orientation angle on the TC of Gr film/Al laminate composites. Figure **5** gives

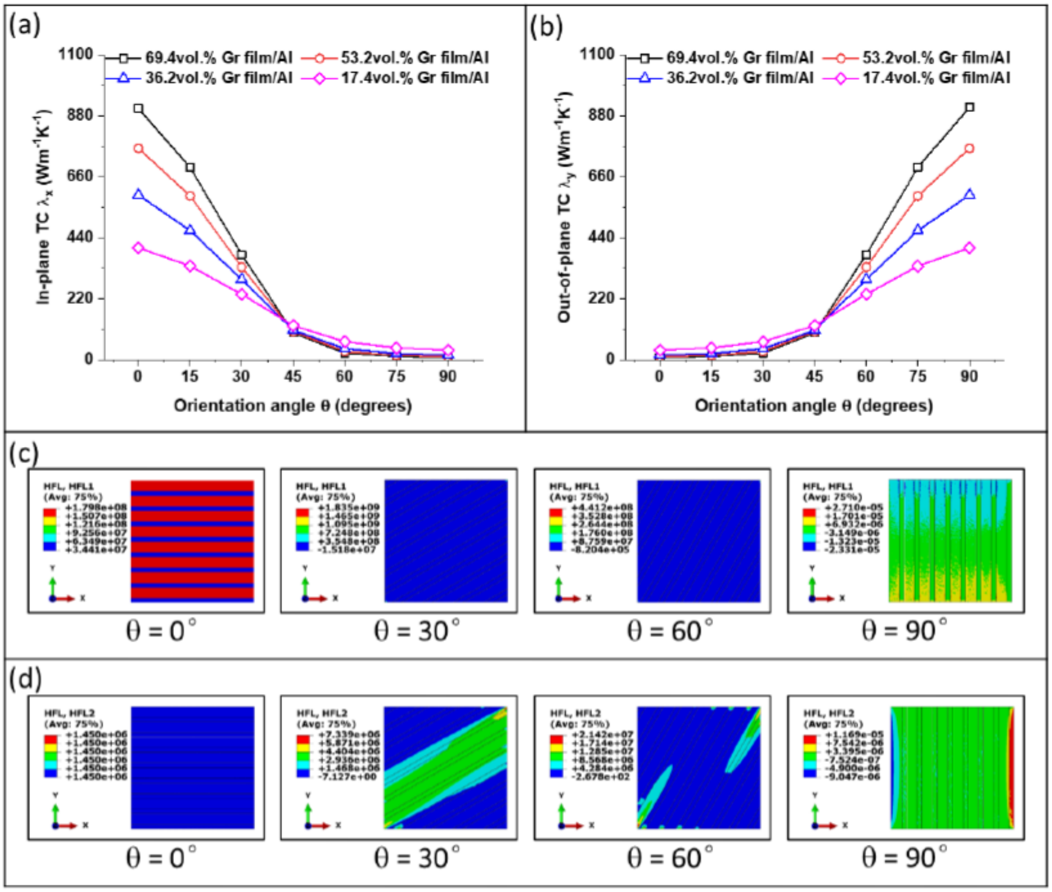

Figure 5: Thermal conductivity and heat flux distributions of Gr film/Al laminated composites with different orientation angles: 0º, 15º, 30º, 45º, 60º, 75ºand 90º: (**a**) in-plane TC and (**c**) heat flux along *x*-axis of 69.4vol. % Gr film/Al, (**b**) out-of-plane TC and (**d**) heat flux along *y*-axis of 69.4vol. % Gr film/Al.

TCs and heat flux distributions of Gr film/Al laminated composites with different laminate orientation angles of 0º, 15º, 30º, 45º, 60º, 75ºand 90º, respectively. In these simulations, the Gr film/Al layer number is 8 and the length is equal to its thickness. Figure **5**(**a**) provides the in-plane TC of Gr film/Al laminated composites with varying laminate orientation angles. When the laminate orientation angle increases from 0º to 45º and then to 90º, the in-plane TC of Gr film/Al laminated composites with a certain volume fraction of Gr films first sharply and then slowly reduces. As to the Gr film/Al laminated composites with Gr volume fractions of 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. %, the in-plane TC of Gr film/Al laminated composites with the laminate orientation angles θ = 0º and 90º are calculated as 404.6 (θ = 0º), 595.6 (θ = 0º), 763.6 (θ = 0º), 908.1 (θ = 0º), 35.4 (θ = 90º), 18.2 (θ = 90º), 12.7 (θ = 90º) and 9.8 (θ = 90º) W·m^{-1}k^{-1}, while those of Gr film/Al laminated composites with the laminate orientation angle θ = 45º are provided as 123.9, 107.3, 101.9 and 97.6 W·m^{-1}k^{-1}. It means that the laminate orientation angle generates an important effect on the in-plane TC of Gr film/Al laminated composites. Figure **5**(**c**) show the heat flux along the *x* direction (HFL1) of 69.4vol. % Gr film/Al laminated composites, in which the laminate orientation angles are 0º, 30º, 60º and 90º, respectively. For the Gr film/Al laminated composites with the laminate orientation angle increasing from 0º to 90º, the peak values of heat flux along laminate direction gradually decrease.

In Figure **5**(**b**), it presents the out-of-plane TC of Gr film/Al laminated composites with varying laminate orientation angles. When the laminate orientation angles increasing from 0º to 45º and then to 90º, the out-of-plane TC of Gr film/Al laminated composites with a certain volume fraction of Gr films first slowly and then sharply increases. With regard to the Gr film/Al laminated composites with Gr volume fractions of 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. %, the out-of-plane TC of Gr film/Al laminated composites with the laminate orientation angles θ = 0º and 90º are calculated as 35.4 (θ = 0º), 18.2 (θ = 0º), 12.7 (θ = 0º), 9.8 (θ = 0º), 404.6 (θ = 90º), 595.6 (θ = 90º), 763.6 (θ =

90º) and 908.1 (θ = 0º) $W \cdot m^{-1}k^{-1}$, while those of Gr film/Al laminated composites with the punching zone rate θ = 45º are provided as 123.9, 107.3, 101.9 and 97.6 $W \cdot m^{-1}k^{-1}$. It can be seen that the in-plane and out-of-plane TC of Gr film/Al laminated composites with varying laminate orientation angles exhibit strong symmetries. Once again, it indicates that the laminate orientation angle generates an important effect on the out-of-plane TC of Gr film/Al laminated composites. Figure **5**(**d**) give the heat flux along the *y* direction of 69.4vol. % Gr film/Al laminated composites, in which the laminate orientation angles are 0º, 30º, 60º and 90º. For the Gr film/Al laminated composites with the laminate orientation angles increasing from 0º to 90º, the peak values of heat flux along the *y* direction first increase and then decrease. Based on the analysis above, the in-plane and out-of-plane TC of Gr film/Al laminated composites can be turned with the variations of laminate orientation angles, which may be useful for the extensive application of Gr film/Al laminated composites.

4. CONCLUSIONS

In this work, the graphite film/aluminum laminated composites were fabricated by vacuum hot pressing and their anisotropic thermal conductivities are measured. 2D composite structural modeling and thermal conductivity simulations of Gr film/Al laminated composites are performed. Effects of volume fraction, interfacial property, punching zone and orientation angle of Gr films on the thermal conductivities of Gr film/Al laminated composites are brought into light. Main conclusions can be summarized:

1) Continuous Gr film/Al laminated composites were fabricated by vacuum hot pressing and in-plane and out-of-plane TCs of produced laminated composites with varying volumes fraction of Gr films were experimentally measured. For the 17.4vol. %, 36.2vol. %, 53.2vol. % and 69.4vol. % Gr film/Al laminated composites, the in-plane TCs are ranging from 380 to 940 $W \cdot m^{-1}k^{-1}$ and the out-of-plane TCs are changing from 28.7 to 7.2 $W \cdot m^{-1}k^{-1}$, respectively.

2) Generally speaking, the volume fraction of Gr films produces the most important effect on the TCs of Gr film/Al laminated composites, while the composite with a larger volume fraction of Gr films possesses the larger in-plane TC and smaller out-of-plane TC. Heat transfer coefficient at Gr films and Al foils hardly produces effects on their in-plane TCs. However, their out-of-plane TCs gradually increase with the raising heat transfer coefficient below 1.0×10^7 $W \cdot m^{-2}k^{-1}$, and their out-of-plane TCs nearly keep constant with the increasing heat transfer coefficient above 1.0×10^7 $W \cdot m^{-2}k^{-1}$. The critical heat transfer coefficient α = 4.5×10^7 $W \cdot m^{-2}k^{-1}$ can be reasonably revealed and indicated to describe the thermal properties between well-bonded carbon-aluminum interfaces.

3) In order to balance the anisotropic TCs of Gr film/Al laminated composites, the punching zone rate and orientation angle of Gr films are well studied. The in-plane TCs gradually go down and the out-of-plane TCs linearly raise up with the increasing punching zone rate. The punching zone number can produce few effects on both in-plane and out-of-plane TCs of laminated composites. Along with the orientation angles of Gr films changing from 0ºto 90º, the in-plane and out-of-plane TCs of Gr film/Al laminated composites exhibits good symmetry to each other, which may generate a large potential to design and fabricate isotropic thermal properties of novel laminated composites.

ACKNOWLEDGEMENTS

The authors acknowledge the financial supports by National Natural Science Foundation (Nos. 51971132, 52192595) and Materials Genome Initiative Center of Shanghai Jiao Tong University.

REFERENCES

[1] Mallik, S.; Ekere, N.; Best, C.; Bhatti, R., Investigation of thermal management materials for automotive electronic control units. Applied Thermal Engineering 2011, 31, (2-3), 355-362. https://doi.org/10.1016/j.applthermaleng.2010.09.023

[2] McNamara, A. J.; Joshi, Y.; Zhang, Z. M., Characterization of nanostructured thermal interface materials - A review. International Journal of Thermal Sciences 2012, 62, 2-11. https://doi.org/10.1016/j.ijthermalsci.2011.10.014

[3] Prieto, R.; Molina, J. M.; Narciso, J.; Louis, E., Fabrication and properties of graphite flakes/metal composites for thermal management applications. Scripta Materialia 2008, 59, (1), 11-14. https://doi.org/10.1016/j.scriptamat.2008.02.026

[4] Moridi, A.; Zhang, L.; Liu, W.; Duvall, S.; Brawley, A.; Jiang, Z.; Yang, S.; Li, C., Characterisation of high thermal conductivity thin-film substrate systems and their interface thermal resistance. Surface and Coatings Technology 2018, 334, 233-242. https://doi.org/10.1016/j.surfcoat.2017.11.021

[5] Bakshi, S. R.; Lahiri, D.; Agarwal, A., Carbon nanotube reinforced metal matrix composites - a review. International Materials Reviews 2013, 55, (1), 41-64. https://doi.org/10.1179/095066009X12572530170543

[6] Han, H.; Zhang, Y.; Wang, N.; Samani, M. K.; Ni, Y.; Mijbil, Z. Y.; Edwards, M.; Xiong, S.; Saaskilahti, K.; Murugesan, M.; Fu, Y.; Ye, L.; Sadeghi, H.; Bailey, S.; Kosevich, Y. A.; Lambert, C. J.; Liu, J.; Volz, S., Functionalization mediates

heat transport in graphene nanoflakes. Nat Commun 2016, 7, 11281.
https://doi.org/10.1038/ncomms11281

[7] Zhong, J.; Liu, D.; Li, Z.; Sun, X., High thermal conductivity materials and their application on the electronic products. 2012 IEEE Asia-Pacific Conference on Antennas and Propagation 2012.
https://doi.org/10.1109/APCAP.2012.6333195

[8] Lin, S.; Ju, S.; Zhang, J.; Shi, G.; He, Y.; Jiang, D., Ultrathin flexible graphene films with high thermal conductivity and excellent EMI shielding performance using large-sized graphene oxide flakes. RSC Advances 2019, 9, (3), 1419-1427.
https://doi.org/10.1039/C8RA09376H

[9] Peng, L.; Xu, Z.; Liu, Z.; Guo, Y.; Li, P.; Gao, C., Ultrahigh Thermal Conductive yet Superflexible Graphene Films. Adv Mater 2017, 29, (27).
https://doi.org/10.1002/adma.201700589

[10] Hu, D.; Gong, W.; Di, J.; Li, D.; Li, R.; Lu, W.; Gu, B.; Sun, B.; Li, Q., Strong graphene-interlayered carbon nanotube films with high thermal conductivity. Carbon 2017, 118, 659-665.
https://doi.org/10.1016/j.carbon.2017.04.005

[11] Gspann, T. S.; Juckes, S. M.; Niven, J. F.; Johnson, M. B.; Elliott, J. A.; White, M. A.; Windle, A. H., High thermal conductivities of carbon nanotube films and micro-fibres and their dependence on morphology. Carbon 2017, 114, 160-168.
https://doi.org/10.1016/j.carbon.2016.12.006

[12] Murakami, M.; Tatami, A.; Tachibana, M., Fabrication of high quality and large area graphite thin films by pyrolysis and graphitization of polyimides. Carbon 2019, 145, 23-30.
https://doi.org/10.1016/j.carbon.2018.12.057

[13] Hutsch, T.; Schubert, T.; Schmidt, J.; Weißgärber, T.; Kieback, B., Innovative metal-graphite composites as thermally conducting materials. PM2010 World Congress, PM Functional Materials-Heat Sinks 2010, 1-8.

[14] Li, W.; Liu, Y.; Wu, G., Preparation of graphite flakes/Al with preferred orientation and high thermal conductivity by squeeze casting. Carbon 2015, 95, 545-551.
https://doi.org/10.1016/j.carbon.2015.08.063

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

[16] Chang, J.; Zhang, Q.; Lin, Y.; Wu, G., Layer by layer graphite film reinforced aluminum composites with an enhanced performance of thermal conduction in the thermal management applications. Journal of Alloys and Compounds 2018, 742, 601-609.
https://doi.org/10.1016/j.jallcom.2018.01.332

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

[18] Huang, Y.; Ouyang, Q.; Guo, Q.; Guo, X.; Zhang, G.; Zhang, D., Graphite film/aluminum laminate composites with ultrahigh thermal conductivity for thermal management applications. Materials & Design 2016, 90, 508-515.
https://doi.org/10.1016/j.matdes.2015.10.146

[19] Landry, K.; Kalogeropoulou, S.; Eustathopoulos, N., Wettability of carbon by aluminum and aluminum alloy. Materials Science and Engineering A 1998, 254, 99-111.
https://doi.org/10.1016/S0921-5093(98)00759-X

[20] Cao, H.; Tan, Z.; Lu, M.-H.; Ji, G.; Yan, X.-J.; Di, C.; Yuan, M.; Guo, Q.; Su, Y.; Addad, A.; Li, Z.; Xiong, D.-B., Graphene interlayer for enhanced interface thermal conductance in metal matrix composites: An approach beyond surface metallization and matrix alloying. Carbon 2019, 150, 60-68.
https://doi.org/10.1016/j.carbon.2019.05.004

[21] Qian, L.; Pang, X.; Zhou, J.; Yang, J.; Lin, S.; Hui, D., Theoretical model and finite element simulation on the effective thermal conductivity of particulate composite materials. Composites Part B: Engineering 2017, 116, 291-297.
https://doi.org/10.1016/j.compositesb.2016.10.067

[22] Huang, Y.; Su, Y.; Li, S.; Ouyang, Q.; Zhang, G.; Zhang, L.; Zhang, D., Fabrication of graphite film/aluminum composites by vacuum hot pressing: Process optimization and thermal conductivity. Composites Part B: Engineering 2016, 107, 43-50.
https://doi.org/10.1016/j.compositesb.2016.09.051

Received on 03-11-2022 Accepted on 30-11-2022 Published on 07-12-2022

DOI: https://doi.org/10.31875/2410-4701.2022.09.04