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Keywords: apparent density, graphite flake/Copper composite, orientation of graphite flakes

Abstract

Due to the anisotropy properties of graphite flakes (G_f), the control of their orientation inside copper (Cu) matrix is strictly correlated with the final properties of such metal matrix composite (MMC). In this study, MMCs are fabricated by powder metallurgy process using three types of Cu powder particles (flake, dendritic, and spherical) as well as different powder filling methods. By using Cu flake powder with a relatively lower apparent density as compared to the two other Cu powder types (dendritic and spherical), high aligned G_f could be easily obtained, after uniaxial hot pressing, in a one-step powder filling approach. For the other two types of powders, a comparable orientation degree of G_f could be achieved only via several steps of delicate powder pressing. In-plane thermal conductivities of Cu/ G_f composites were enhanced by improving the orientation degree of G_f , agreeing with effective medium approximation predictions (Maximum TC up to 540 W mK^{-1}). A modeling, based on the apparent density of Cu powders, was discussed to show the effect of the alignment process on the final thermal property of such MMCs. This model supplies a basic guideline to obtain highly orientated G_f .

1. Introduction

With the advent of artificial intelligence (AI) and the boosting need for information processing capability, the trend for the microelectronic chip's development is toward the fabrication of parts with higher integration, high power density, and high frequency. This trend has induced a drastic increase in the heat generation of chips in a confined space. In order to maintain the electronic devices' performances, heat dissipation ability has become a key challenge in the design of electronic devices [1]. A solution to this issue is to apply thermal management materials with high thermal conductivity (TC) that could effectively cool down the chips through heat transfer.

Recently, due to their high potential in showing tailored thermal properties, graphite flakes (G_f) reinforced MMCs, have been extensively investigated. These investigations have been mainly focused on G_f orientation improvement and interfacial property control [2–7]. It is noted that G_f is a highly anisotropic material as it has different thermal properties between its a axis (high TC close to 1000 W mK^{-1}) and c axis (low TC ranging from 10 to 20 W mK^{-1}). Considering the initial investigation done on G_f reinforced MMCs, the orientation of G_f was regarded as a key parameter affecting the in-plane TC of the composites [2]. Moreover, G_f has poor wettability to

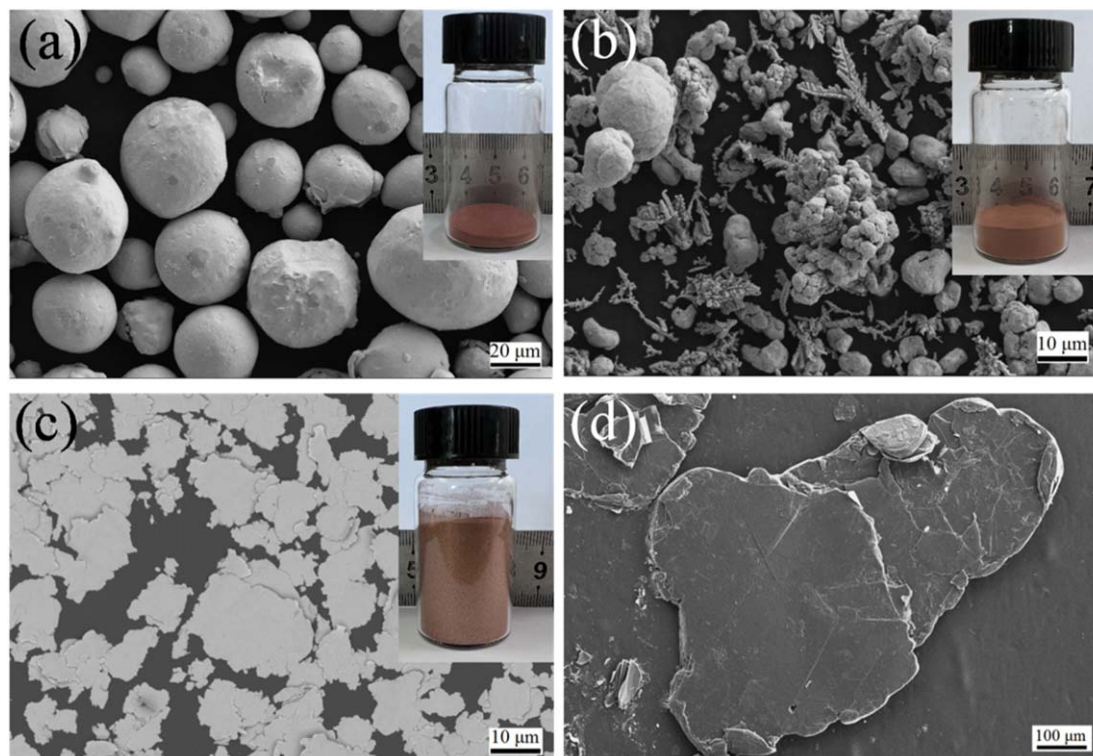


Figure 1. SEM micrographs showing the morphologies of as-received Cu powders and G_f : (a) spherical Cu powder, (b) dendritic Cu powder, (c) flake Cu powder, (d) G_f . Insets showing the transparent bottles with 8 g of Cu powder inside.

copper which is not a favor for heat transfer [8]. Increasing the relative density of final composite materials via hot pressing is also necessary for improving the thermal conductivity.

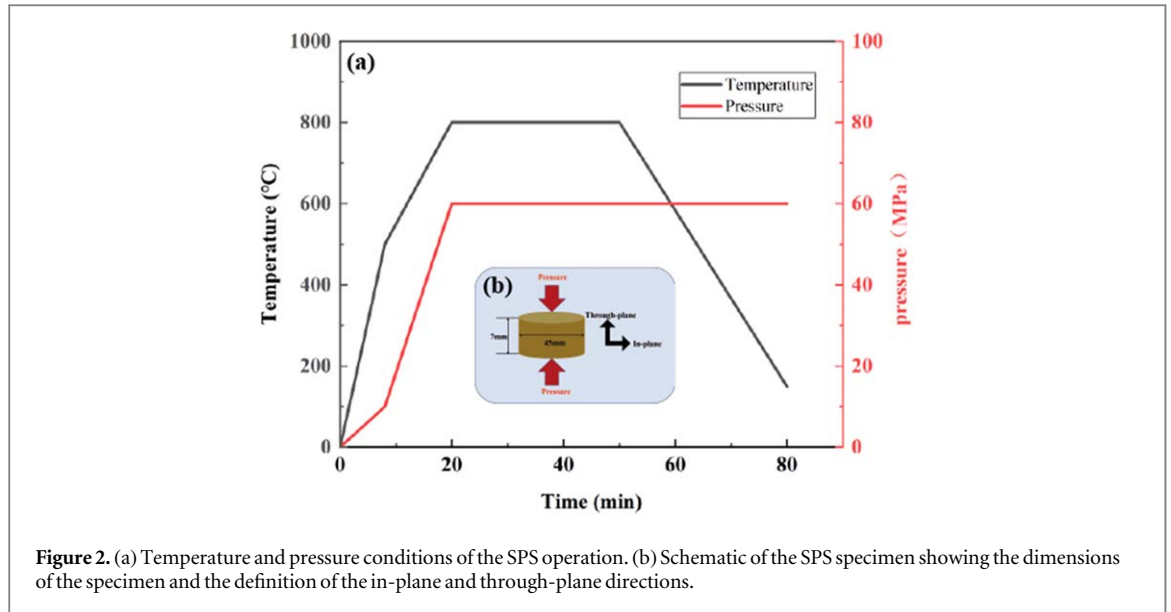
It is thought that due to the flake like geometry of the G_f (in-plane ‘diameter’ ranging from 100 to 600 μm and out of plane thickness in the range of 10 to 30 μm), G_f tend to be aligned on top of each other under uniaxial hot pressing. This is linked to the large ratio between diameter and thickness or flaky morphology. Usually, full oriented graphite flakes are achieved by step-by-step powder filling process or stacking-pressing [3, 4, 9] and so with the control of the thickness of each powder layer. Indeed, misalignments of G_f tend to occur when the powder filling process is done via one-step process [10, 11]. However, it is reported that once flake powder metallurgy is applied, G_f with a high orientation degree could be obtained in composite materials processed through a one-step powder pressing procedure [10, 12, 13]. This effect was attributed to the platelet geometry of the used matrix metal powder which tends to get aligned as the added G_f [10, 12, 13]. It is mentioned that flake metal powders after the ball milling process have a lower apparent density as compared with the spherical one [14]. This can be a key factor for obtaining G_f with a high orientation degree. However, no systematic work has yet been done to understand the critical parameters being required to achieve the composites with highly oriented G_f .

In this work, various Cu powders, such as flake, spherical, and dendritic, were selected as matrix materials corresponding to different ranges of apparent density. One-step and step-by-step powder filling methods were also implemented to investigate the effect of filling step numbers on the alignment of G_f under the application of different Cu powders. The TCs were measured to confirm the relationship between the orientation degree of G_f and the TC improvement. Furthermore, a model was proposed that shows the dependence of the G_f orientation process on the apparent density of Cu powder when using uniaxial hot pressing as the densification approach.

2. Experimental

Commercial Cu powders, including spherical (figure 1(a)), dendritic (figure 1(b)), and flaky (figure 1(c)) types, were manually mixed with G_f (Yanxin-Granphite Co., Ltd., figure 1(d)), respectively, in which the volume fraction of G_f was fixed at 40% for each MMC type. The apparent densities of three types of Cu + G_f mixtures were measured by a standard container and scale.

The G_f + Cu powder mixtures were filled in a graphite mold under both one-step and step-by-step methods consisting of 9 and 15 steps. In each step, the Cu/ G_f powder mixture was put into the graphite mold, spread, and



then pressed using a punch under 1 MPa. The Cu/ G_f composite materials were fabricated by the spark plasma sintering (SPS) technique. The heating and pressing curves of the SPS process are illustrated in Figure 2. In brief, under a vacuum environment, the sample was heated to the temperature of 500 °C for 8 min and meanwhile, an axial pressure of 10 MPa was applied within 10 min [15–17]. The sample was then heated to 800 °C and maintained for 30 min. The uniaxial pressure was also increased to 60 MPa. The procedure was then followed by natural cooling to room temperature. These SPS samples had a dimension of 45 mm in diameter and 7 mm in height. Three types of specimens obtained from pure Cu powder particles were also sintered to measure the physical properties of the matrices without G_f .

Orientations of G_f in the composites were determined by combining both field emission scanning electron microscopy analysis (FE-SEM, Zeiss Gemini 300 system) and image analysis. TCs were calculated by multiplying the thermal diffusivities, the density, and the heat capacity (C_p) of the densified materials. A laser flash method (NETZSCH LFA457) was used to measure the in-plane thermal diffusivity at 70 °C. The density was measured based on Archimedes' method using a densimeter (AND GHe202). The C_p of the powders (Cu and G_f) were measured using the differential scanning calorimeter method (DSC, PerkinElmer DSC8000) with the sapphire as a reference. The C_p of the Cu/ G_f composite materials were calculated according to the rule of mixture.

3. Results and discussion

3.1. Microstructures of Cu/ G_f composites and their orientation analysis

The micrographs of Cu/ G_f composites using different Cu types of powder and powder filling process strategies are presented in Figure 3. In all of the samples, most of the G_f were preferentially aligned in a direction perpendicular to the pressing direction, indicating that G_f tend to be naturally oriented during the uniaxial pressing. It has to be mentioned that some disorientations were observed in the samples fabricated with both spherical and dendritic Cu powders being treated by the one-step powder filling (figures 3(a) and (d)), As noted, this phenomenon was more evident in the spherical ones (figure 3(a)). Using 9-step method, this phenomenon became less obvious, and got almost disappeared after 15-step process (figures 3(c) and (f)). On the other hand, such disorientation behavior was hardly seen in the composite materials fabricated with Cu flake powder (figures 3(g), (h), and (i)), even if the process was done in a one-step route (figure 3(g)).

According to the effective medium approximation (EMA) [18], $\langle \cos^2 \theta \rangle$ is used to represent the alignment degree of reinforcement in the matrix.

$$\langle \cos^2 \theta \rangle = \frac{\int \varphi(\theta) \cos^2 \theta \sin \theta d\theta}{\int \varphi(\theta) \sin \theta d\theta} \quad (6)$$

where $\rho(\theta)$ is a distribution function describing the angle distribution of the reinforcement in the matrix. In this work, θ is the angle between the basic graphite plane and the x-y plane of the composite materials. When the value of $\langle \cos^2 \theta \rangle$ is equal to 1 the G_f reinforcements are exactly oriented in a plane perpendicular to the pressing direction. However, if this value is close to 1/3 the G_f reinforcements are randomly distributed inside the Cu matrix. Image analysis was used to statistically calculate the angle θ distribution of G_f [19, 20]. Figure 4 shows the

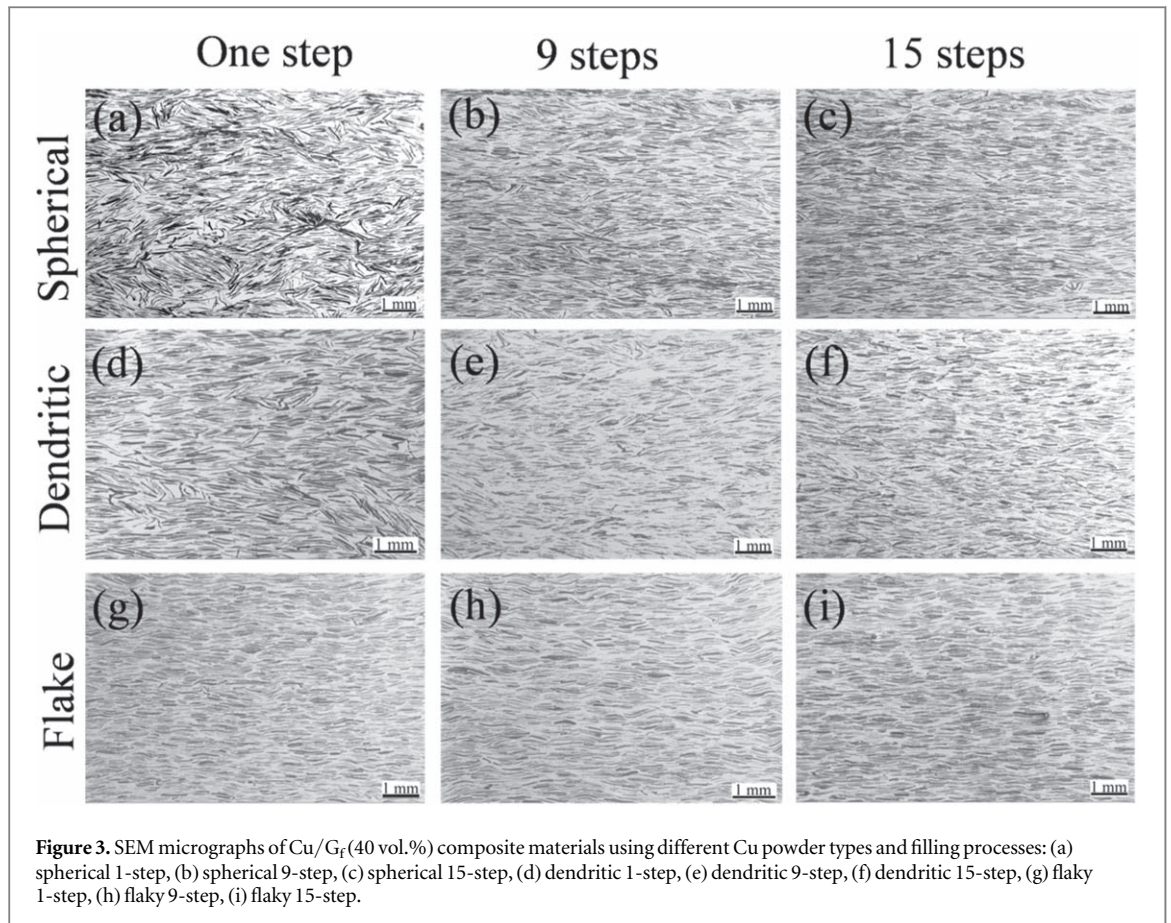


Figure 3. SEM micrographs of Cu/G_f (40 vol.%) composite materials using different Cu powder types and filling processes: (a) spherical 1-step, (b) spherical 9-step, (c) spherical 15-step, (d) dendritic 1-step, (e) dendritic 9-step, (f) dendritic 15-step, (g) flaky 1-step, (h) flaky 9-step, (i) flaky 15-step.

results containing the fittings of angle distribution of G_f existing in the nine samples shown in figure 3. Herein, a function $\varphi(\theta)$ was used to fit the curves of the frequency (the number of G_f at certain θ in all the G_f) versus θ using Origin software [19, 21].

$$\varphi(\theta) = A_1 e^{(-\theta/t_1)} + y_0 \quad (5)$$

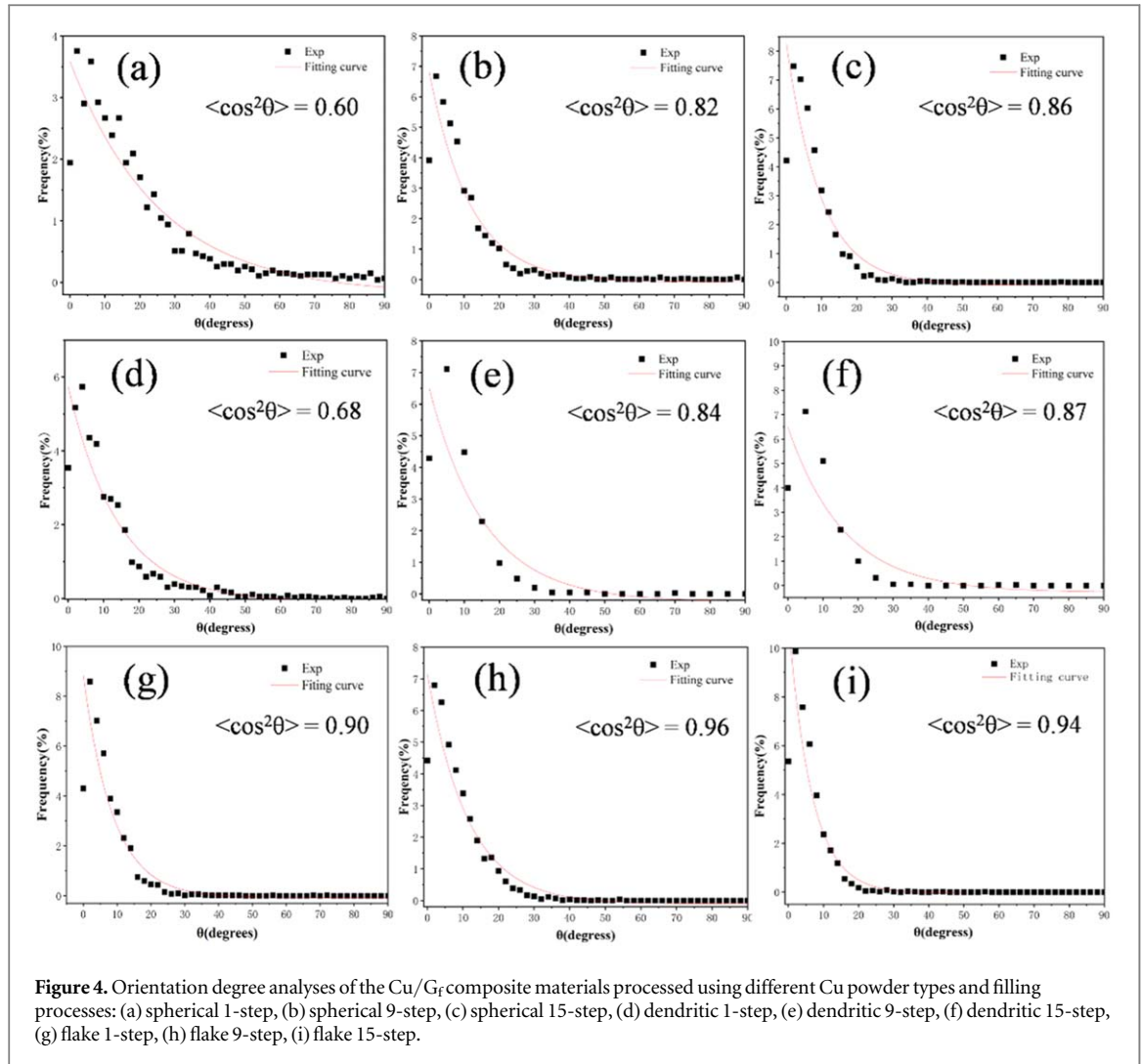
where the symbol $\varphi(\theta)$ denotes the statistical distribution of the reinforcing graphite phase within the copper matrix, with A₁, t₁, and y₀ serving as fitting variables.

Upon substituting the values obtained from the fitting process, the specific $\varphi(\theta)$ functions were presented, and subsequent numerical integration facilitates the computation of the $\langle \cos^2\theta \rangle$ values obtained from the SEM micrographs of each material. $\langle \cos^2\theta \rangle$ values are also shown in the figure 4. Except for the samples processed using spherical and dendritic Cu powders and under one-step powder filling way, the values of $\langle \cos^2\theta \rangle$ in all the other materials were higher than 0.8. Especially, the $\langle \cos^2\theta \rangle$ values were greater than 0.9 for those samples fabricated using the Cu flake powder. This is in accordance with the observation obtained from SEM micrographs (figure 3).

The interface features of all the samples are shown in figure 5. As the copper became soft at 800 °C, the copper matrix was tightly attached to the graphite flake in a flat interface even in the samples using spherical Cu powder, which is different from the puckered interface using spherical Al powder [10]. 99% of relative densities can be explained by those interface characterizations. In addition, this void-free interface is also favorable for the thermal transfer between the matrix and reinforcement.

3.2. Thermal conductivity of Cu/G_f composite materials

Figure 6 shows the values of the calculated TC and their corresponding $\langle \cos^2\theta \rangle$ values within the in-plane direction of Cu/G_f composite materials fabricated utilizing both the one-step filling and step-by-step filling approaches. The specimen fabricated using Cu flake powder shows the highest TC as compared to the other two types of materials processed under the one-step processing route and using the other powder types. However, this difference in TC values became smaller when the step-by-step powder filling method was applied. It can be claimed that the increase of $\langle \cos^2\theta \rangle$ values or improved orientation degree leads to the increase of TC values. Moreover, the multiple-step filling process had a much weaker effect on the increase of the TC values of the Cu/G_f composites prepared using the Cu flake powder. This is merely because the G_f reached a relatively high orientation degree via the one-step route in these parts and there was not much space left for improvement. It



should be noted that the $\langle \cos^2\theta \rangle$ values of samples processed using dendritic and spherical powders and treated by the 15-step powder filling process were almost identical. These results fit with previous research findings [22]. However, their TC values showed a significant difference. This can be traced back to the noticeable difference observed in the TC values of the sintered pure copper samples (cf table 1).

In this study, given that the size and volume fraction of G_f were fixed and by ignoring the little effect caused by the TC properties of different Cu matrices and their interfacial thermal conductance, it could be concluded that, according to the EMA model, the TC value along the in-plane direction has a strong relationship with the orientation degree of G_f. Here, we present the in-plane TC formula as follows:

$$K_{//} = K_m \frac{2 + f[\beta_L(1 - S_L)(1 + \langle \cos^2\theta \rangle) + \beta_T(1 - S_T)(1 - \langle \cos^2\theta \rangle)]}{2 - f[\beta_L S_L(1 + \langle \cos^2\theta \rangle) + \beta_T S_L(1 - \langle \cos^2\theta \rangle)]} \quad (2)$$

in which

$$\beta_i = \frac{K_i - K_m}{K_m + S_i(K_i - K_m)}, \quad (i = L, T) \quad (3)$$

$$S_L = \frac{\pi t}{4D}, \quad S_T = 1 - \frac{\pi t}{2D} \quad (4)$$

where the subscripts L and T are noted as the in-plane and through-plane directions of G_f, respectively, f is the volume fraction of G_f and S is the geometrical factor, where D and t are the diameter and thickness of the reinforcement, respectively [10], K_m is the TC of the Cu matrix, which was from the measured TC of the as-sintered Cu samples using different copper powders, K_i is intrinsic TC of G_f and K_L, K_T is 1000 W mK⁻¹ [3], and 10 W m⁻¹ K⁻¹ [23], respectively.

As shown in figure 7, the theoretical in-plane TC values increase as the $\langle \cos^2\theta \rangle$ value changes from 0.5 to 1 covering the range for the samples processed using three types of powders. As mentioned above, a small deviation between the three curves is mainly caused by the different TC values of the pure Cu powders. The

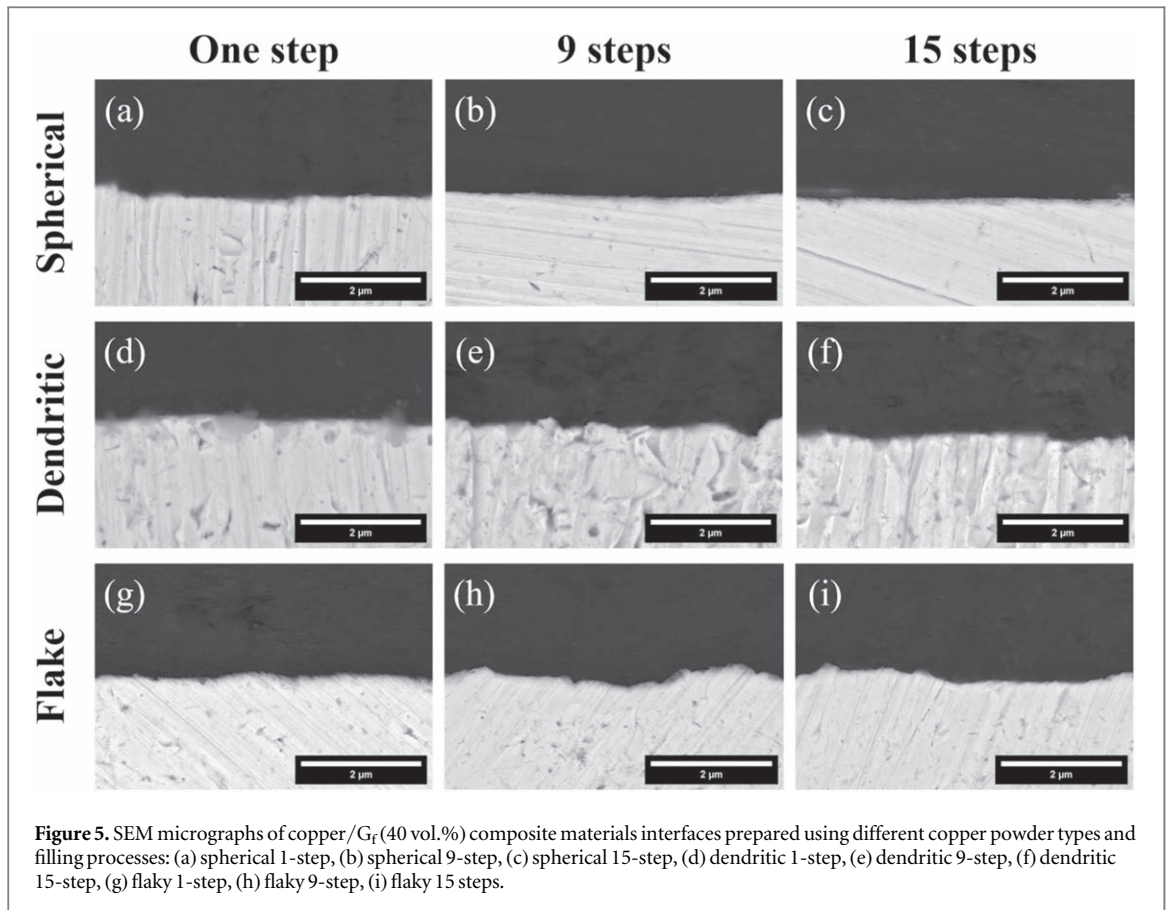


Figure 5. SEM micrographs of copper/ G_f (40 vol.%) composite materials interfaces prepared using different copper powder types and filling processes: (a) spherical 1-step, (b) spherical 9-step, (c) spherical 15-step, (d) dendritic 1-step, (e) dendritic 9-step, (f) dendritic 15-step, (g) flaky 1-step, (h) flaky 9-step, (i) flaky 15 steps.

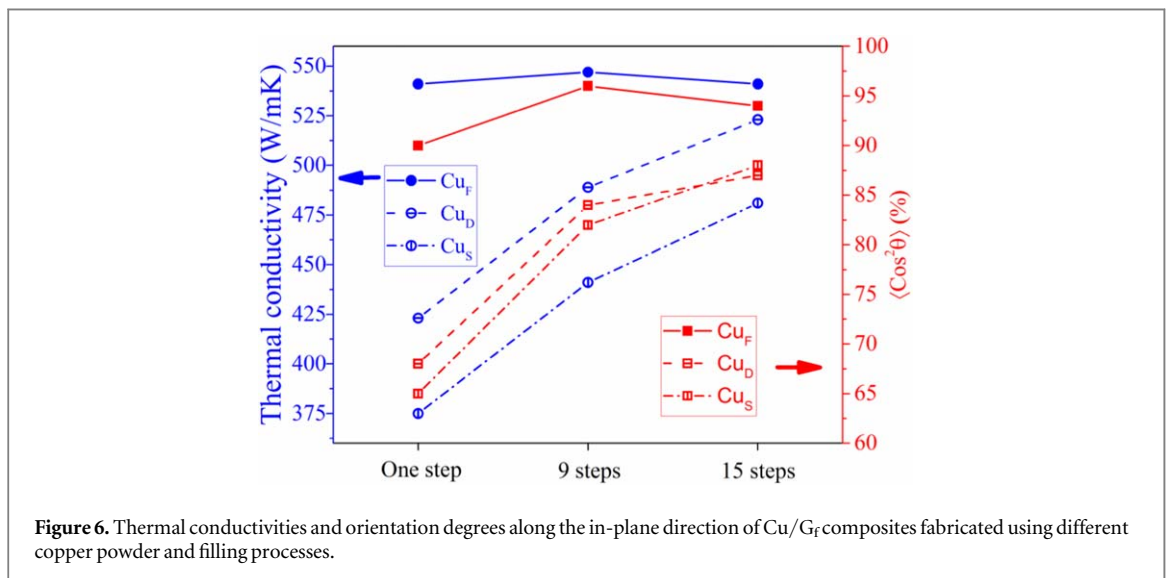
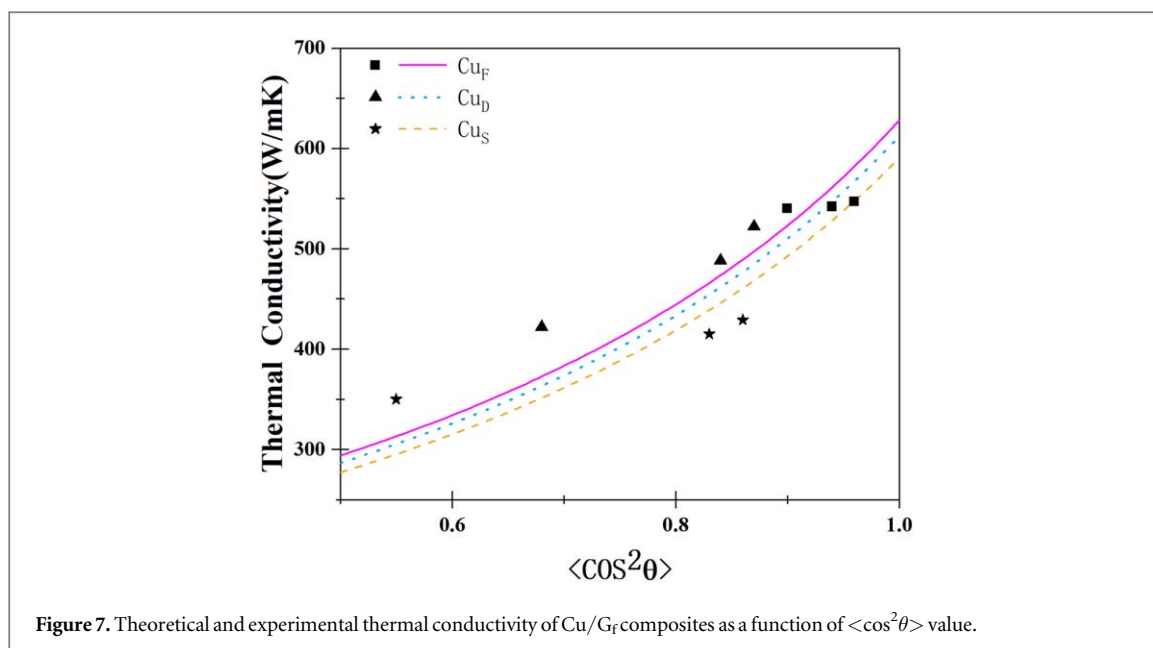


Figure 6. Thermal conductivities and orientation degrees along the in-plane direction of Cu/ G_f composites fabricated using different copper powder and filling processes.

Table 1. Physical properties of sintered Cu samples using as-received pure Cu powders.

Materials	ρ (g/cm ³)	Apparent density (g/cm ³)	Cp (J/kg·K)	TC (W/mK)
Cu _F	8.86	0.5	299	370
Cu _D	8.90	2.3	283	366
Cu _S	8.89	3.7	222	342



measured TCs of Cu/G_f composites are well consistent with the predictions. It demonstrates that an efficient way to maximize the in-plane TC is to tailor the $\langle \cos^2\theta \rangle$ value or orientation degree.

3.3. An interpretation of achieving high in-plane TC of Cu/G_f composites

As shown in table 1, the major difference existing among the three Cu powders is the difference in the apparent densities of the powders. The apparent density value of spherical Cu powder is seven times than that for flake Cu one, (cf the inset photos of figure 1). It is worth noting that the G_f can get distributed in a larger space after getting mixed with Cu powder particles with a lower apparent density. As illustrated in figure 8, regardless of the type of the used Cu powders, the same mass of G_f are distributed in the same mass of Cu powder particles. As Cu powders have different apparent densities, diverse volumes of Cu + G_f mixtures can be observed inside the graphite mold for the same quantity of each of these Cu powders. It was assumed that G_f are randomly distributed inside the copper powder after the mold filling. During the uniaxial hot densification step, G_f can move inside of the Cu + G_f mixture and tend to orient in a plane perpendicular to the pressure direction. From the non-dense packing state to the dense state, during the pressing step, G_f would travel a greater distance to be aligned in Cu flake + G_f mixture, as indicated by the yellow arrows in figure 8.

For the step-by-step method, the thickness of the powder layer for each step was determined by the amount of powder mixtures filled in the graphite die. When the thickness of the powder layer was controlled under the average value of the diameter of G_f, the G_f was constrained in a laying state. Thus, almost all the G_f lie flat in a single powder layer. When the Cu/G_f composites are stacked by this powder layer, a highly oriented G_f can be obtained. However, this process is still unclear, and further investigation via *in situ* industrial X-ray computed tomography can be a proper approach.

4. Conclusions

Flake, dendritic, and spherical Cu powders were used to fabricate Cu/G_f composite materials. Those three powders show various apparent densities. Flake Cu powder has the lowest apparent density as compared with the others. High orientation of G_f was successfully achieved using flake Cu powder and a one-step powder filling method. This could be obtained in parts processed by the others only under multiple-step processes. Under the one-step approach, disordered G_f in which G_f were not oriented fully, were easily observed in the samples processed by dendritic or spherical copper powders. Increased orientation degree of G_f enhances in-plane TC of Cu/G_f composites (Maximum TC up to 540 W mK⁻¹). An interpretation based on the apparent density and multiple-step powder filling approach was proposed to explain the mechanism of obtaining highly oriented G_f in the copper matrix under powder metallurgy. The lower the apparent densities of the used metal powder, the fewer steps of powder filling are required to obtain G_f with high orientation.

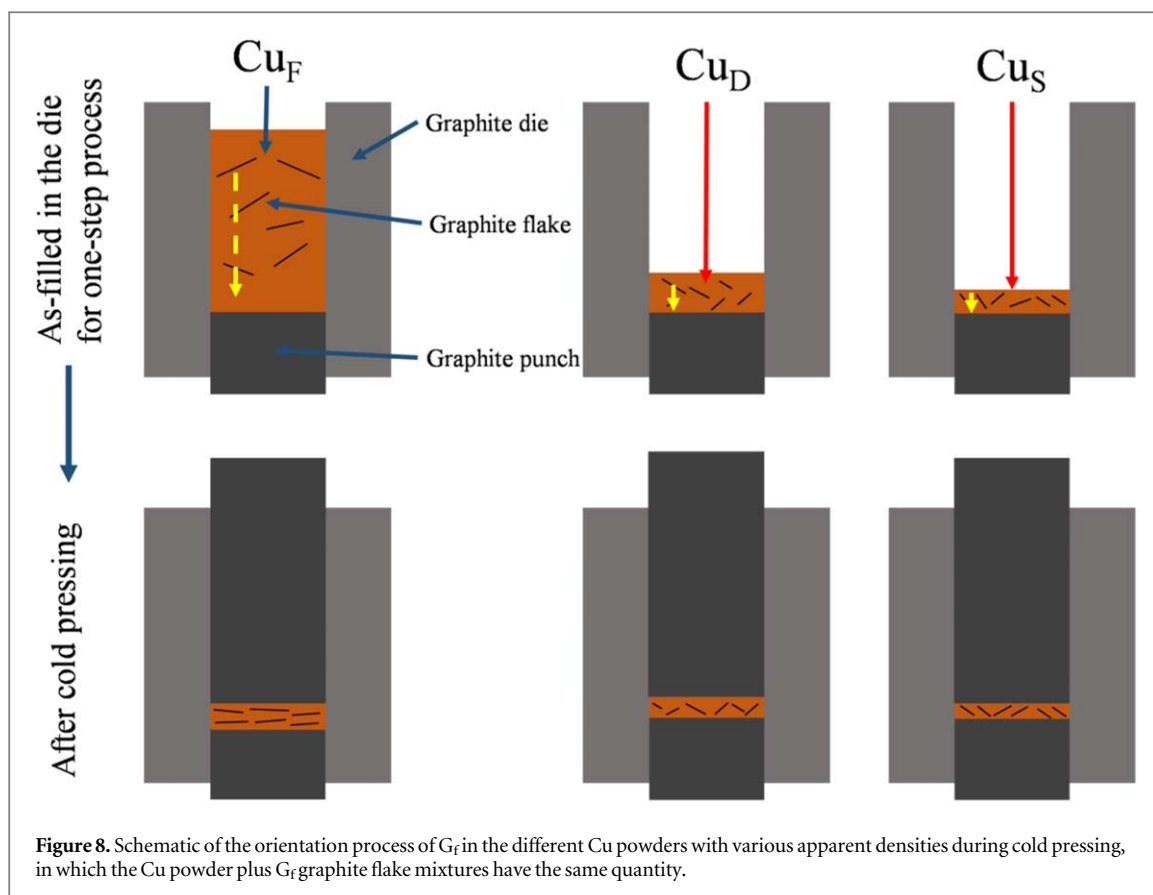


Figure 8. Schematic of the orientation process of G_f in the different Cu powders with various apparent densities during cold pressing, in which the Cu powder plus G_f graphite flake mixtures have the same quantity.

Acknowledgments

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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