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## Ultrahigh thermal conductivity copper/graphite membrane composites prepared by tape casting with hot-pressing sintering

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### ABSTRACT

Ultrahigh thermal conductivity Cu/graphite membrane composite with continuous well-packed laminate structure is prepared by tape casting with hot-pressing sintering. As-prepared composite material has anisotropic thermal properties with ultrahigh in-plane thermal conductivity and low in-plane coefficient of thermal expansion because of its extraordinary packed laminate structure. Thermal properties of Cu/graphite membrane composite can be designed according to requirements by adjusting volume fraction of graphite membrane in the composite. The maximum value of Cu/graphite membrane composite (50 vol% graphite membrane) in-plane thermal conductivity is 1005 W  $\cdot$  m<sup>-1</sup> K<sup>-1</sup>, while the minimum value of the in-plane coefficient of thermal expansion is 7 × 10<sup>-6</sup> K<sup>-1</sup>. This meets the high requirements for thermal management materials.

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#### 1. Introduction

With the increasing demand of electronic products for higher thermal efficiency, electronic packaging thermal management materials with excellent heat transfer performance have become increasingly important in aerospace and integrated circuit industries [1]. In recent years, metal matrix composites (MMCs) have received significant attention because of their outstanding combination of high thermal conductivity (TC) and adjustable coefficient of thermal expansion (CTE), as well as excellent mechanical properties [2–5]. Copper is a promising matrix material due to its high TC and excellent formability, as well as good corrosion resistance [6]. Graphite membrane (GM) with ultrahigh in-plane TC of 1100–1600 W  $\cdot$  m<sup>-1</sup> K<sup>-1</sup> and low in-plane CTE is a promising thermal management material, which has been widely used in dissipating heat from electronic products and can be made into large area of continuous uniform films [7-9]. Therefore, GMreinforced Cu matrix composite may be a good candidate to meet the requirements of numerous thermal management applications.

In this study, the ultrahigh TC Cu/GM composite with well-packed laminate structure was successfully fabricated by tape casting with hot-pressing sintering without other additives into the raw materials. The aim of the current study was to obtain

\* Corresponding author. *E-mail addresses:* haojunjie@ustb.edu.cn (J. Hao), cgchen@ustb.edu.cn (C. Chen). suitable composition and structure for optimal composite performance. The effects of composite microstructure and GM volume fraction on composite properties were investigated. This work demonstrates a new type of MMCs with ultrahigh TC along with the suitable preparation method.

#### 2. Experimental

Electrolytic copper powder (99.9%, <10  $\mu$ m) with TC of 400 W  $\cdot$  m<sup>-1</sup> K<sup>-1</sup> and CTE of 16.8  $\times$  10<sup>-6</sup> K<sup>-1</sup> and GM (25  $\mu$ m average thickness) with in-plane TC of 1500 W  $\cdot$  m<sup>-1</sup> K<sup>-1</sup> and out-plane TC of 20 W  $\cdot$  m<sup>-1</sup> K<sup>-1</sup>, and in-plane CTE of  $-1.5 \times 10^{-6}$  K<sup>-1</sup> were used as raw materials. The slurry for tape casting process was made by adding solvent, binder, plasticizer and defoamer to the copper powder. Subsequently, slurry was cast into thin films on the GM with a doctor blade, and film thickness was adjusted by the height of the doctor blade [10,11]. After composite film was dried in the vacuum drying oven at 313 K for 1 h, debonding and reduction process of the film proceeded simultaneously at 673 K for 2 h in hydrogen atmosphere. Subsequently, the composites were sintered by hot-pressing sintering at 1273 K under 10 MPa pressure.

Cu/GM composite microstructure was observed by field emission scanning electron microscope (FE-SEM, QUANTAFEG450, USA). The crystal orientation of the composite was analyzed by X-ray diffraction (XRD, RIGAKU TTR3, Japan).  $\lambda$  (W · m<sup>-1</sup> K<sup>-1</sup>) is







thermal conductivity of the composite, which was calculated using the following formula:

$$\lambda = \alpha \cdot C_p \cdot \rho \tag{1}$$

Here,  $\alpha$  (m<sup>2</sup> · s<sup>-1</sup>) is the thermal diffusion coefficient of the composite measured at room temperature by laser flash method (NETZSCH LFA447, Germany) in radial direction of GM with the sample size of  $10 \times 10 \times 4$  mm;  $C_p$  (J · g<sup>-1</sup> K<sup>-1</sup>) is specific heat of the composite, which was calculated according to rule of mixture:

$$C_p = C_{Cu}V_{Cu} + C_{GM}V_{GM} \tag{2}$$

Here,  $C_{Cu}$ ,  $C_{GM}$  is specific heat and  $V_{Cu}$ ,  $V_{GM}$  is volume fraction of copper and GM, respectively. Composite density was measured using the Archimedes principle. Coefficient of thermal expansion of the composite was measured in the 293-473 K range by the thermal dilatometer (NETZSCH DIL 402PC, Germany) in the axial direction of GM.

#### 3. Results and discussion

Fig. 1a shows the FE-SEM image of GM, while the image in the top right corner shows oblique section of GM. GM consists of multiple thin films with many wrinkles, which evidently indicates that GM has excellent flexibility and certain strength. Fig. 1b shows the XRD pattern of ground GM and the in-plane of GM. The two diffraction peaks at 26.52° and 54.72° of both XRD patterns correspond to the diffraction of the (003) and (006) planes of in-plane of GM, respectively. This XRD result indicates that GM grains are neatly arranged. Fig. 1c is an FE-SEM image of typical local structure. Composite has a dual component structure of copper and GM, arranged alternatively in the form of lamellae, which reveals that GMs in the composite are uniform and continuous. XRD pattern of the x-axis of Cu/GM composite is shown in Fig. 1d, where the three peaks at  $43.40^{\circ}$ ,  $50.50^{\circ}$  and  $74.54^{\circ}$  correspond to the (111), (200) and (220) diffraction planes of copper. Diffraction peak intensity of (003) GM plane is decreased significantly, while the intensity of (006) crystal plane is almost invisible, which proves that the as-prepared Cu/GM composite with laminate structure is anisotropic. Thus, composite properties are also anisotropic.

Cu/GM composite was prepared with volume fraction of GM ranging from 10% to 50%. It should be noted that GM is an anisotropic material with in-plane TC of 1500 W  $\cdot$  m<sup>-1</sup> K<sup>-1</sup> and out-plane TC of  $20 \text{ W} \cdot \text{m}^{-1} \text{ K}^{-1}$ . Fig. 2a and b show that the x-y plane TC of the composite increases approximately linearly from 585  $W\cdot m^{-1}\;K^{-1}\;$  to  $\;1005\;W\cdot m^{-1}\;K^{-1}\;$  with the increasing GM volume fraction. The z-axis TC decreases from 40 W  $\cdot$  m<sup>-1</sup> K<sup>-1</sup> to  $12 \text{ W} \cdot \text{m}^{-1} \text{ K}^{-1}$  at the same time, which proves that GM has significant impact on the composite TC. Compared with the highest known thermal conductivity of carbon/Cu matrix composite at 930 W  $\cdot$  m<sup>-1</sup> K<sup>-1</sup> [12], we prepared composite with higher thermal conductivity.

To better understand the thermal conductivity of the Cu/GM composite, series-parallel connection models were used to predict TC of Cu/GM composites without considering interface thermal resistance (ITR), which can be expressed by the following equations:

$$\lambda_c = V_{Cu}\lambda_{Cu} + V_{GM}\lambda_{GM} \tag{3}$$

$$\lambda_c^{-1} = V_{Cu}\lambda_{Cu}^{-1} + V_{GM}\lambda_{CM}^{-1} \tag{4}$$

.

Here,  $\lambda_c$  is TC of the Cu/GM composite,  $V_{Cu}$  and  $V_{GM}$  is the volume fraction of copper and GM,  $\lambda_{Cu}$  and  $\lambda_{GM}$  is TC of copper and GM, respectively. Eq. (3) represents the parallel connection model, which is suitable for predicting TC of the x-y plane, while Eq. (4) is the series connection model, which is suitable for predicting TC of the z-axis. It can be observed that the experimental values of the x-y plane fit well with the predicted values in Fig. 2a, while predicted values of the z-axis have relatively large difference with the experimental values in Fig. 2b. This proves that the series-parallel connection models can predict TC of material relatively accurately

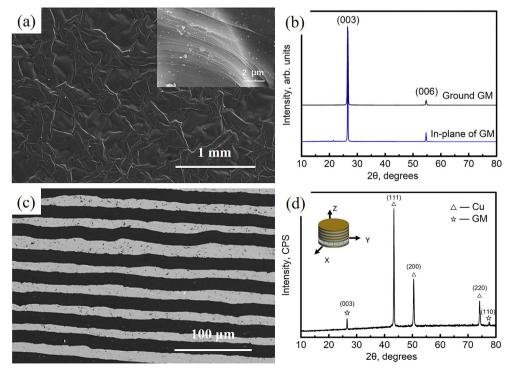


Fig. 1. (a) FE-SEM images of surface and interlayer GM morphology; (b) XRD patterns of ground GM and in-plane of GM; (c) FE-SEM image of local structure and (d) XRD pattern of x-axis of Cu/GM composite. Thumbnail in (a) is oblique image of GM.

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