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The roles of geometry and topology structures of graphite fillers on thermal conductivity of the graphite/aluminum composites

C. Zhou [a], D. Chen [a], X.B. Zhang [a], Z. Chen [a,*], S.Y. Zhong [a], Y. Wu [a], G. Ji [b], H.W. Wang [a]

[a] *State Key Laboratory of Metal Matrix Composites, Shanghai Jiao Tong University, Shanghai 200240, China*
[b] *Unité Matériaux et Transformations, CNRS UMR 8207, Université Lille 1, Villeneuve d'Ascq 59655, France*

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ABSTRACT

Various graphite fillers, such as graphite particles, graphite fibers, graphite flakes and porous graphite blocks, have been successfully incorporated into an Al alloy by squeeze casting in order to fabricate graphite/Al composites with enhanced thermal conductivity (TC). Microstructural characterization by X-ray diffraction and scanning electron microscopy has revealed a tightly-adhered, clean and Al_4C_3-free interface between the graphite fillers and the Al matrix in all the as-fabricated composites. Taking the microstructural features into account, we generalized the corresponding predictive models for the TCs of these composites with the effective medium approximation and the Maxwell mean-field scheme, which both show good agreement with the experimental data. The roles of geometry and topology structures of graphite fillers on the TCs of the composites were further discussed.

1. Introduction

Effective thermal management in power electronic devices requires a heat sink material with high thermal conductivity (TC) and low coefficient of thermal expansion (CTE, <10 ppm/K), being compatible with that of electronic components [1]. Traditional thermal management materials, e.g. pure Cu and Al, Mo/Cu, W/Cu and SiC/Al composites, are insufficient to meet the requirement of heat removal in recently-developed power electronic devices, due to their too high CTE and, in particular, too low TC. For example, it has been reported that TCs of the SiC/Al composites are in the range 170–250 W/m K [2,3], the maximal value being only slightly higher than that of pure Al (around 237 W/m K).

Generally, graphite based materials are promising thermal conductive fillers due to their high TC, low CTE and low cost. Hence, formation of graphite/Al composites by proper incorporation of graphite fillers in the Al matrix offers the possibility for thermal management applications, with the potential for tailoring overall thermal properties (TC and CTE) of the composites. To date, several graphite fillers (graphite particles, graphite fibers, graphite flakes, graphite foam, etc.) have been mixed with Al alloys through various fabrication techniques (e.g. squeeze casting, spark plasma sintering, vacuum hot pressing, etc.) to fabricate highly thermal conductive composites [4–7]. However, in reality, the obtained TCs of these composites are not as high as expected by theoretical predictions. Some believe that interfacial thermal resistance (ITR) is predominantly responsible for this, since undesirable excessive formation of aluminum carbide (Al_4C_3) can hinder effective heat transfer cross the graphite/Al interface, due to a low TC of Al_4C_3 itself [5,7,8]. In contrast, a molecular dynamics simulations study [9] has recently highlighted that the effect of structure factors, like aspect ratio, geometry and topology, of graphite fillers on global TCs of the composites are more significant than that of ITR. Especially, in case when the formed graphite/Al interface is Al_4C_3 free, this effect could be even more important and needs to be systematically investigated. Unfortunately, to our knowledge, not enough attention has been paid to this issue so far.

The aim of this work is to study the influence of geometry and topology structure factors of graphite fillers on the TCs of the composites. Graphite fillers with different forms (i.e. particle, fiber, flake and porous block) were incorporated in an Al alloy matrix by squeeze casting. Microstructure was characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM). Besides, we generalized corresponding predictive models for the TCs of the composites by considering the observed microstructural features.

2. Experimental

The starting materials consisted of graphite fillers (i.e., graphite particles, graphite fibers, graphite flakes, and porous graphite

* Corresponding author. Tel.: +86 21 5474 7597.
E-mail address: zhe.chen@sjtu.edu.cn (Z. Chen).

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Table 1
Densities and TCs of the starting materials used in this work.

Starting materials	Density (g/cm^3)	TC (W/m K)
Graphite particles	2.26	100[a]
Graphite fibers	2.15	*xy*: 12[a], *z*: 700[a]
Graphite flakes	2.26	*xy*: 1000 [7], *z*: 38 [6]
Porous graphite block	1.74–1.85	154–168
Al-7Si alloy	2.68	151
Si particles	2.33	150

[a] From data sheet of manufacturers.

block), an Al-7Si alloy and Si particles. Their thermo-physical properties are given in Table 1. Graphite flakes have a TC of about 1000 W/m K along the basal plane (i.e. *xy*-plane) [7]. However, the TC perpendicular to the basal plane (i.e. *z*-axis) is only 38 W/m K [6]. Graphite fibers have the second highest TC of 700 W/m K along the fiber axis direction (i.e. *z*-axis). Comparatively, graphite particles and porous graphite block have isotropic and relatively low TCs. Fully-dense graphite/Al-7Si composites were produced by squeeze casting. Graphite fillers were first loaded in a steel mould and pressed into a preform at room temperature. Note that this step is not necessary for the porous graphite block. The preform and Al-7Si alloy were then preheated up to 400 °C and 760 °C, respectively. Subsequently, the molten Al-7Si alloy was poured onto the preform, and a pressure of 100 MPa was applied to force the molten alloy to completely infiltrate the preform. Si particles were used as spacers in the case of graphite flakes. Initial mixing of graphite flakes and Si particles was carried out by mechanical stirring for 30 min, which was followed by the same processing process as for the other fillers. More details of the fabrication process can be found in [10].

Microstructural characterization was carried out using XRD and SEM. A JEOL JSM-7600F instrument was used for SEM examinations. XRD measurements were performed on a D8 ADVANCE X-ray diffractometer, operating at 40 kV/40 mA and using Cu K_α radiation ($\lambda = 0.15406$ nm). TCs of the specimens (Ø 12.6 × 2 mm) were measured by a laser-flash method using a NETZSCH LFA 447 instrument. Measurement accuracy of this instrument was about ±3%. Overall uncertainty of the measured TC values given in this paper consisted of measurement error and geometrical uncertainty.

3. Results and discussion

3.1. Microstructural characterization

Fig. 1 illustrates the different morphologies of the graphite fillers. Graphite particles are irregular in shape and appear agglomerated to some extent (Fig. 1(a)). Graphite fibers have an average diameter of about 10 μm and an average length of about 400 μm (Fig. 1(b)). Graphite flakes, with an average diameter of about 500 μm and an average thickness of about 20 μm (Fig. 1(c)), display a neat platelet morphology. Porous graphite block has interconnected pores visible on its fracture surface (Fig. 1(d)). Fig. 2 shows the spatial distributions of the graphite fillers in the as-fabricated graphite/Al-7Si composites. As shown in Figs. 2(a) and (b), graphite particles are homogeneously embedded in the Al-7Si alloy matrix, while graphite fibers present preferential directions (mostly perpendicular to the pressing direction). The fibers are oriented by the applied pressing pressure during the fabrication process. The elongated dark regions in Fig. 2(c) represent the graphite flakes while the rest bright ones are the Si/Al-7Si matrix. The layers of graphite flakes are stacked mostly parallel to each other and separated by the added Si particles. Fig. 2(d) illustrates the microstructure of the porous graphite block/Al-7Si composite where the light and dark gray contrasts represent the infiltrated Al-7Si alloy and graphite block, respectively. It should be noted that no visible pores are observed (i.e. full density) in all these composites. The inserts in Figs. 2(a)–(d) show no evidence to confirm the formation of Al_4C_3 at the graphite/Al interfaces. The absence of Al_4C_3 is also confirmed by XRD (Fig. 3). Whatever the graphite fillers, only the diffraction peaks of graphite, Al and Si are present.

3.2. Thermal-physical properties of the graphite/Al-7Si composites

The measured thermal-physical properties of the graphite/Al-7Si composites are given in Table 2. The as-fabricated graphite flakes/Si_p/Al-7Si and graphite fibers/Al-7Si composites show remarkable anisotropic TC values, which is attributed to oriented flakes and fibers in the composites, consistent with the SEM observations (see Figs. 2(b) and (c)). Both composites present relatively high TCs in *xy*-plane but low TCs along *z*-axis. For example, the in-plane TCs of the graphite flakes/Si_p/Al-7Si composites are in the range 275–504 W/m K, much higher than the TCs along *z*-axis in

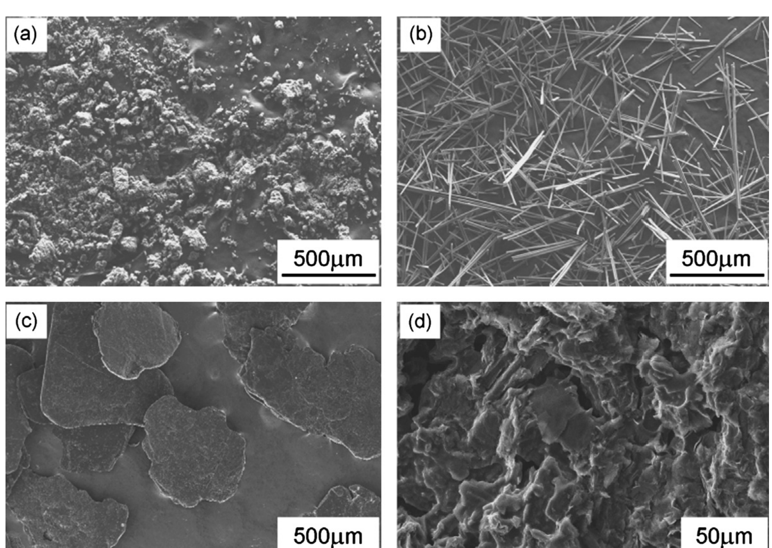

Fig. 1. SEM micrographs showing morphologies of the starting graphite fillers: (a) graphite particles, (b) graphite fibers, (c) graphite flakes, (d) porous graphite block.