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Preparation of graphite flakes/Al with preferred orientation and high thermal conductivity by squeeze casting

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A B S T R A C T

High thermal conductive graphite flakes/Al composites were prepared by optimized squeeze casting technique with higher molten temperature and lower infiltration pressure. The characterization and thermal conductivity (TC) of graphite flakes/Al composites were investigated. The graphite flakes/Al composite prepared from 70 vol. % natural graphite has a high TC as high as 714 W/m K, as well as a high thermal diffusivity of 388 mm^2/s in the plane parallel to the graphite layers. However, the compressive and flexural strengths in direction perpendicular to the graphite layers are limited to 17.6 and 16.9 MPa, respectively. Microstructural analysis demonstrates that the graphite flakes/Al composites have a typical structural anisotropy derived from the three-dimensional layered arrangement of the highly oriented graphite flakes. Meanwhile transmission electron microscope observations further illustrate that the formation of a tightly-adhered amorphous layer interface between graphite flakes and Al matrix without Al_4C_3 reaction product, which is contributed to the excellent thermal conductive of graphite flakes/Al composites.

1. Introduction

With increased levels of dissipated power, the electronic industry requires smaller, more capable, and more efficient electronic systems, which makes thermal challenges have become critical issues in electronics [1,2]. Since heat removal plays a key role in the continuing progress in the electronic industry, thermal conductivity (TC) of materials is a key parameter for thermal design and management of the electronic components. Thus, it is an optimal choice to identify a material with extremely high TC and ideally light weight for solving thermal management problems [3]. Among the different classes of materials nowadays being considered in electronics, the extreme excellent thermal properties of flake graphite are attractive great attention for applications in high performance thermal management, especially used as an ideal candidate reinforcement for manufacturing composites [3–12].

It has been reported that TC in (002) crystal plane of natural crystalline flake graphite can reaches 2200 W/m K at room temperature and the single crystal graphite has a maximum in-plane TC of 2800 W/m K at 80 K [8,13,14]. Since optimizing the orientation of graphite platelets in composites can greatly improve the in-plane TC, the control of preferred orientation of the graphite flakes is clearly very important [3,4,8,15,16]. Many results for the preparation of high TC graphite flakes/Al composites have been reported. An essential problem for manufacturing graphite flakes/Al composites is that there is almost no space between graphite flakes, which makes liquid metal infiltration under pressure an almost unfeasible task [9–11]. The previous efforts for solving this problem were to introduce a small proportion of SiC or Si particles which acted as spacers between the layers of graphite flakes [10,11]. Prieto et al. added SiC as spacers to prepare 69 vol. % graphite flakes/$AlSi_{12}$ composites with TC of 390 W/m K [11]. Similarly, Cong Zhou et al. manufactured 71.1 vol. % graphite flakes/$AlSi_7Mg_{0.3}$ composites with Si added and improved thermal conductivity to 526 W/mK [10]. However, the existence of spacers acted as impurities hinders thermal transfer and degrades thermal conductivity of graphite flakes/Al. That means there is still great potential to develop preferred orientation graphite flakes/Al materials without separator between layers of flakes. Commercial available graphite flakes/Al composites produced by MMCC Inc. (AlGrp™) are fabricated by net shape gas pressure infiltration and have a very high TC up to 750 W/mK with density of 2.3 g/cm^3 [17]. However, there is not information available about the composition of matrix Al alloy,

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volume fraction of graphite flakes, thermal diffusivities and specific heat capacities. Compared with the density of graphite flakes (2.26 g/cm^3) [10,11], this indicates that AlGrp™ (750 W/mK) has a very high content of fillers. The volume fraction of graphite flakes, as the key parameter to determine thermal properties of composites, could be as high as 90% estimated by Rule of Mixtures density (Al density of 2.7 g/cm^3) [10,11]. Meanwhile, the volume fraction of AlGrp™ with TC of 650 W/mK (2.35 g/cm^3) is also close to 79%.

The composite fabricated by piston-driven squeeze casting exhibits better properties due to the presence of fewer common defects such as porosity and shrinking cavities, and the elimination of segregation of the reinforcement [18]. However, graphite flakes/Al composites without separator fabricated by this technique have seldom been reported, and details of their thermal properties are still lacking. The molten temperature is a key parameter for liquid metal infiltration between graphite flakes, due to its positive effect on liquid metal fluidity. Meanwhile, the control of infiltration pressure is crucial for maintaining the graphite flakes preferred orientation and improving infiltration channel inside the graphite flakes preform. Therefore, it is acceptable to decrease the infiltration pressure to prevent graphite flake sliding and increase infiltration channel.

This work reports the preparation and characterization of highly oriented graphite flakes/Al composites without separator prepared by a simple squeeze casting technique. Compared with the earlier studies, this represents a simplification of the process with higher molten temperature and lower infiltration pressure to maintain the flakes alignment and make liquid metal infiltration between them. The highly oriented structure and interfacial configuration of graphite flake/Al composites were discussed. In addition, the thermal properties and mechanical strength of composites were also investigated.

2. Experimental

2.1. Raw materials

The raw materials consisted of reinforcement of graphite flakes and pure Al. Graphite flakes were supplied by Qingdao tianshengda Graphite Co. Ltd., China, with the mean particle size of 495, 270 and 150 μm (99.9% purity). The matrix alloy was pure Al, with the chemical composition (wt. %): 0.12%Si, 0.16%Fe, 0.03%Cu, 0.01 Mg, 0.001Ca and the balance Al. The following discussions refer to the microstructural analysis of 495 um graphite flakes/Al, except in Section 3.4.

2.2. Preparation of the graphite flakes/Al composites

The composites were fabricated by squeeze casting and Fig. 1 shows the processing schematic. Firstly, the graphite flakes were put into a mold, naturally tend to lie on top of each other [9], and vibrated to possess denser stacking structure before pressed to be a graphite preform. Then, the preform was preheated at 973 K, meanwhile the metal Al was melt, degassed and cleaned in an Al_2O_3 crucible at 1073 K. Finally, the molten Al was poured into the preheated mold containing graphite perform and a pressure of up to 45 MPa was applied to vertically force the molten Al to completely infiltrate the graphite flakes perform. The pressure was maintained for 300 s until the solidification was finished. Concerning the infiltration pressure is the key factor of influencing infiltration channel inside the graphite flakes preform, it is reasonable to cover a graphite sheet on the graphite preform in order to reduce the impact force and make liquid metal infiltration easily. The graphite flakes/Al composites we fabricated were with the size of Φ130 × 50 mm.

As mentioned previously, this work focuses on developing preferred orientation graphite flakes/Al materials without separator between layers of flakes. As in conventional squeeze casting processes, the dies are initially preheated around 200–300 K below the liquidus or melting point of Al to induce matrix rapid solidification [19]. For example, Cong Zhou et al. selected 400 °C and 760 °C as the optimized preheat temperature and infiltration temperature, respectively, for fabricating graphite flakes/Si/Al composites [10]. Considering the poor fluidity of pure Al, that elevating the preheat temperature is beneficial to improving molten Al fluidity during infiltration. That is what led us to use 973 K as the optimal preheat temperature.

It has been reported that a relatively high infiltration temperature can induce the formation of the interfacial Al_4C_3 phase which could hinder thermal transfer and degrade mechanical properties [10,20–22]. Conversely, the fluidity of molten aluminum can be improved by elevating the infiltration temperature, which could be helpful to complete liquid metal infiltration between graphite flakes. The formation of Al_4C_3 phase has been thought to be thermodynamically favorable, and the amount of Al_4C_3 is determined by the kinetics [23]. For this reason, the high cooling rate in piston-driven squeeze casting can also suppress the formation of the interfacial Al_4C_3 phase. Hence, it is acceptable to use a relatively high infiltration temperature 1073 K.

Generally, in order to promote infiltration of molten Al alloy into preform, the applied pressures in squeeze casting are typically in the range of 50–100 MPa, which can provide for good composite soundness and good tolerance values [19,24]. However, concerning the graphite flakes preform without binder added stacked a three-dimensional microstructure, these high applied pressures may cause preform deformation and graphite flake sliding, which is the reason for existence of veins (preform cracks filled with metal that weakens the material properties locally) in metal matrix composites. Meanwhile, these high applied pressures can also make the infiltration channels too small so hinder the infiltration process [10]. For these reasons, an optimized lower infiltration pressure of 45 MPa was used to avoid graphite flake sliding, maintain the

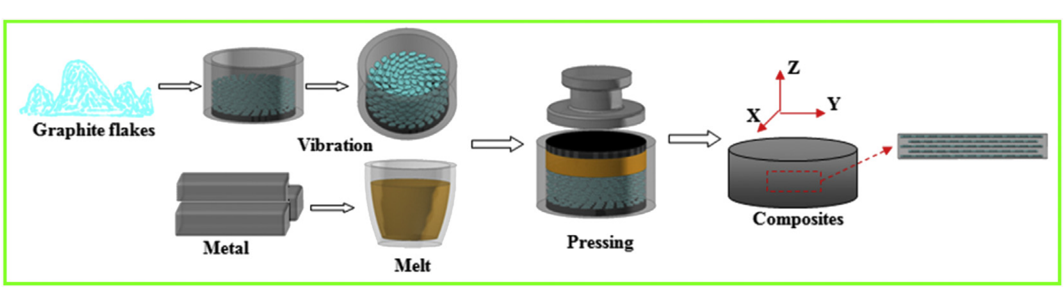

Fig. 1. Schematic of preparation process of graphite flakes/Al. (A colour version of this figure can be viewed online.)