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Graphite film/aluminum laminate composites with ultrahigh thermal conductivity for thermal management applications

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ABSTRACT

In order to meet the requirements in thermal management, a novel kind of carbon/metal composites, namely graphite film/aluminum laminate composites with excellent thermal properties, was developed. The effects of oxide films on interfacial structure and properties of the composites were identified, and the microstructures and properties of the composites were studied as a function of the volume fraction of graphite films. An ultrahigh thermal conductivity of 902 W/mK for aluminum matrix composites was obtained. The measured in-plane thermal conductivities of the composites are all over 80% of the rule of mixtures prediction. What's more, these composites are shown to have more competitive thermal conductivities as compared to other kinds of carbon/aluminum composites, which makes these composites promising candidates to be used for thermal management.

1. Introduction

Effective thermal management is becoming increasingly important for the reliability of electronic components owing to continuous developments of the electronic industry [1–3]. Therefore, it is essential to develop thermal management materials with high thermal conductivity (TC) and low coefficient of thermal expansion (CTE) [1,4]. Carbon materials, such as graphite, diamond, carbon fibers (CFs), carbon nanotubes (CNTs) and graphene, have proved to be promising thermal management materials due to their excellent thermal properties [2,5–7]. These materials can not only be directly used in thermal management applications, but also combined with other materials, such as metals, to form thermal management composites [8]. These composites offer the possibility of tailoring the properties of a metal by adding an appropriate carbon reinforcement to meet the requirements of thermal management. However, the poor wettability and possible harmful interfacial chemical reactions between carbon materials and metals would decrease the reinforcement effect of the carbon materials [9–11]. Therefore, the fabrication process of these composites should be well-designed and well-controlled in order to obtain carbon/metal (C/metal) composites with fine microstructures and high thermal properties [9–11].

To date, diamond/metal composites, CF/metal composites and graphite flake/metal composites are the mostly studied C/metal composites that can be used in thermal management applications. Diamond/metal composites have high TC ranging from 350 W/mK to 780 W/mK, and have been commercialized [8,9,12]. Unfortunately, their high-cost and poor machining properties strongly limit their application [8]. Due to the anisotropic thermal properties of CFs and graphite flakes, excellent properties are only achieved in one or two axial directions of their composites [8]. For example, if carbon fibers with high TC (also high price) were used, the TC of long carbon fiber (LCF)/metal composites can reach 500–700 W/mK along the axial direction while the TC of short carbon fiber (SCF)/metal composites are 208 W/mK in the in-plane direction [13,14]. Also, as reported by Chen et al. [15], the in-plane TC of the graphite flake/metal composites can reach 324 W/mK to 783 W/mK. However, it is difficult to control the exact direction of the graphite flakes as well as the infiltration process [8,15]. In order to promote infiltration, other reinforcements such as SiC [8,16], Si [10] and CF [8] were often added into graphite flake/metal composites, which, unfortunately, would inevitably lead to a reduction in the TC of the resulting composites. Recently, CNT/metal composites and graphene/metal composites have drawn much attention in thermal management due to outstanding thermal properties of CNTs and graphene, while, unfortunately, it has been found that the improvement of thermal properties of these composites is hindered by many technical problems, such as the control of the distribution and orientation of the reinforcements and high interfacial thermal resistance [11].

Recently, graphite films, such as artificial graphite films fabricated from polyimide films [7,17] and graphene oxide films [2,18], have attracted much attention in thermal management due to their excellent thermal properties. Specially, artificial graphite films with high TC (1100–1600 W/mK) have become commercially available for heat removal in electronic devices containing integrated circuits, such as personal computers and mobile phones [7]. Unfortunately, so far, little attention has been paid to the potential of these graphite films being used as effective reinforcements in composites. Actually, if these

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graphite films are used as reinforcements to form graphite film/metal composites and fabrication processes of the composites are well-designed and well-controlled, high TC and low CTE can be expected. Furthermore, if we use graphite films and metal foils as raw materials to fabricate graphite film/metal laminate composites, we can easily control the distributions and orientations of the reinforcements by controlling the stacking of graphite films and metal foils, which is easier and more effective as compared to CF/metal composites, graphite flake/metal composites, CNT/metal composites and graphene/metal composites, which may simplify the fabrication process and meanwhile obtain composites with higher thermal properties.

In this study, we successfully fabricated these novel graphite film/aluminum laminate composites with high TC and low CTE by a vacuum hot pressing process. The effects of oxide films on the surface of aluminum foils and volume fraction of graphite films on microstructures and properties of the composites were investigated and discussed. The microstructures of the composites were characterized at length scales from the macro down to the nanoscale to investigate the distribution and orientation of the graphite films, and the graphite/Al interfacial structure and the presence of harmful reaction product. Furthermore, the thermal conductivities of the as-fabricated composites were compared with the predicted values calculated by rule of mixture and results of other kinds of carbon/aluminum (C/Al) composites reported in the literatures.

2. Experimental

Graphite films and pure aluminum foils (99.9% in purity) were used as raw materials. The graphite films were acquired from Dasen Electronic Material Co., Ltd. in China. The morphologies of the graphite films are shown in Fig. 1 (a). These graphite films have a rough surface with many ripples on it. The inset shows the cross-section of the graphite films. The average thickness of the graphite films was measured to be 29.5 μm. Fig. 1 (b) shows the X-ray diffraction (XRD) pattern of the graphite films, where only peaks of graphite can be detected from this pattern, and the $K_{\alpha1}$ and $K_{\alpha2}$ peaks of (006) peaks are separated very well, which indicates that the graphite films have high purity and crystallinity [19]. The in-plane and out-of-plane TC values of the graphite films were measured to be 1222.6 (±30.3) W/mK and 17.5 (±2.5) W/mK, respectively.

Graphite film/aluminum laminate composites were fabricated by a vacuum hot pressing process. Four sets of composite samples with different volume fractions of graphite films were obtained by controlling the thickness of the aluminum foils. The volume fractions of graphite films of sample #1, #2, #3 and #4 are 17.4%, 36.2%, 53.2% and 69.4%, respectively. The detailed fabrication processes of these laminate composites are listed as following: 1). Graphite films and aluminum foils were washed by acetone and then dried. 2). In order to eliminate the oxide films on the surface of the aluminum foils, they were first washed by 15 g/L NaOH solution, then washed by HNO_3 with volume fraction of 25%, and finally washed by alcohol and dried. 3). The pre-treated graphite films and aluminum foils were carefully filled into a graphite mold layer-by-layer and then heated to 655 °C in high vacuum condition (less than 5×10^{-3} Pa) and kept for 100 min, while an uniaxial pressure of 22 MPa was applied. In order to figure out the effect of oxide films on microstructures and properties of the composites, a composite sample named #3′ was fabricated without the oxide film eliminating process (process 2)). Since the thickness of the oxide films (1–5 nm) [20,21] is much smaller than that of the aluminum foils (26 μm), the effect of them on volume fraction can be neglected. Thus, the graphite volume fraction of sample #3′ is considered to be the same with sample #3.

The in-plane thermal diffusivities of the graphite films (Φ25.4 mm) and the composite samples (10 × 10 × 4 mm) at room temperature were measured by a laser flash technique using a NETZSCH LFA447 thermal analyzer. The specific heat capacities of the samples at room temperature were measured by PerkinElmer differential scanning calorimeter (DSC) 8000. A speed of 20 °C/min was used in the temperature range of 15–35 °C. The densities of the samples were measured by the Archimedes method. The in-plane TC values of the samples were calculated by the product of the density, thermal diffusivity and specific heat capacity. The in-plane CTE values of the specimens (10 × 2 × 25 mm) were measured by a dilatometer (NETZCH DIL 402C) from room temperature up to 300 °C at a speed of 5 °C/min. XRD patterns of the graphite films were obtained by a D/max-2550 instrument (Cu Kα). A scan speed of 4°/min was used in the range of 10–90°. Microstructures of the graphite films and composite samples were characterized by scanning electron microscopy (SEM) operated at 20 kV using a FEI Quanta FEG 250 electron microscope and transmission electron microscope (TEM) using a JEOL 2100F Field Emission Electron Microscope. The energy dispersive spectroscopy (EDS) element line scanning across the aluminum-graphite interface was characterized using the aforementioned SEM.

3. Results and discussion

3.1. Effect of oxide films on microstructures and properties of the composites

According to the results reported in graphite fiber/aluminum composites [22] and diamond/aluminum composites [23], during the fabrication process of the composites, diffusion may happen between aluminum and carbon materials to form a strong interfacial bonding. However, in this study where aluminum foils are used for raw material, there are thin oxide films with thickness of 1–5 nm on the surface of aluminum foils due to their exposure to air [20,21], which would generally

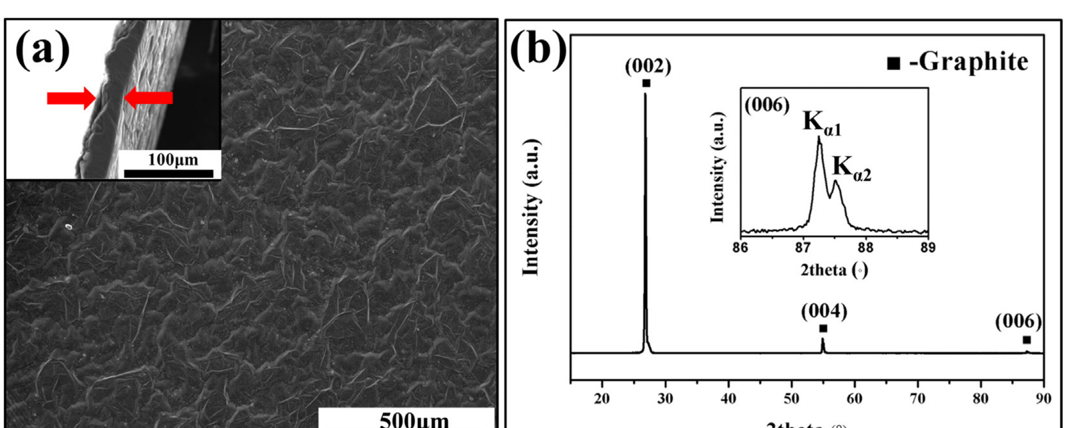

Fig. 1. (a) Morphologies of the surface and cross-section (inset, between the two arrows) of the graphite films; (b) XRD patterns of the graphite films.