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Interfacial microstructure of graphite flake reinforced aluminum matrix composites fabricated via hot pressing

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ABSTRACT

The microstructure of graphite flake (GF) reinforced aluminum (Al) matrix (Al–GF) composites was observed in detail. Due to thermal mismatch between Al and GF, an inner structure of GF was damaged in proximity to the Al/GF interface, while the unique bridging of the sticky graphite sheets barely connected the Al matrix and GF. This result suggests that the GF interlaminar strength is weaker than the Al/GF interfacial strength; the GF interlaminar strength is thus the dominant determinant of the thermomechanical and mechanical properties of the Al–GF composite. Whereas the thermal conductivity of the Al–GF composite was consistent with that theoretically predicted, the outstanding thermal expansion coefficient (TEC) of the graphite was not reflected in the produced Al–GF composites. The damaged inner structure of GF in proximity to the Al/GF interface contributes to heat transfer but does not bear the load resulting from thermal stress.

1. Introduction

Exponential progress in the development of new electronic components requires the use of high-performance heat sink materials, which strike a balance between high thermal conductivity and a thermal expansion coefficient (TEC) close to that of semiconductors and ceramic substrates in order to minimize thermal stress at the joints [1]. As potential heat sink materials, metal matrix composites have been studied owing to their tunable thermal properties. Due to their low density, Aluminum (Al)-based composites have a great advantage in terms of the fabrication of mobile electronic devices, which have become mainstream in recent years [2–7]. Al-based composites are generally fabricated via solid-phase methods (e.g. powder metallurgy) in order to avoid excess interfacial reaction, although the aluminum oxide (Al_2O_3) layer which covers the surface of Al particles often interrupts densification in solid-phase processes [8–11].

A range of materials have been considered for use as reinforcements in Al-based heat sink composite materials, including silicon carbide (SiC) [2], aluminum nitride (AlN) [3], and various carbon materials [4,12,13]. Carbon materials such as diamond and carbon fiber are promising because of their outstanding thermal conductivity, while they are also reactive with Al. However, although diamond reportedly combines remarkable thermal conductivity ($1000 \sim 2000$ W/m K) with a low CTE (2.1×10^{-6}/K) [14], it is expensive and complicates secondary processing. In contrast, carbon fiber is inexpensive and has an advantage in terms of workability – although its thermal conductivity is lower than that of diamond. It has been reported that carbon fiber exhibits anisotropic thermal conductivity and TEC, with recorded values of $\sim$1100 W/m K and $-1.45 \sim -0.6 \times 10^{-6}$/K in the longitudinal direction [15,16], and 5 W/m K and 12.0×10^{-6}/K in the transverse direction [16].

While the thermal and thermomechanical properties of actual graphite flakes (GFs) have not been widely reported, it is largely assumed that GF combines thermal and thermomechanical properties comparable to those of diamond (in the a–b plane) with a workability similar to that of carbon fiber, according to the reported thermal and thermomechanical properties of highly oriented graphite bulks. Murakami et al. have revealed that the highly oriented graphite block obtained from polycondensation polymer films has a thermal conductivity higher than 1000 W/m K and a TEC of -1.0×10^{-6}/K in the a–b plane [17]. It has also been reported that highly oriented pyrolytic graphite (HOPG) has a

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thermal conductivity of $1600 \sim 2000$ W/m K and a TEC of -1.0×10^{-6}/K in the a–b plane [18,19]. GF therefore appears to be a suitable material for the reinforcement of Al-base composites for use in thermal management applications. Indeed, Chen et al. have already documented the remarkable thermal conductivity and TEC of a graphite flake reinforced Al matrix (Al–GF) composite, by using the GF with an average diameter of 550 μm [20]. However, studies involving the microstructural characterization of Al–GF composites are scarce, with the correlation between composite microstructure and thermal and thermomechanical properties not yet understood.

In the present study a detailed observation is made of the microstructure of an Al–GF composite, especially at the Al/GF interface. The Al–GF composite was prepared via conventional hot pressing. We have reported that a small quantity of aluminum–silicon alloy (Al–Si) effectively helps the densification process in Al-based composites [21,22], a small amount of Al–Si was introduced into the Al matrix powder. Furthermore, we also investigated the thermal conductivity and TEC of the fabricated Al–GF composite, discussing these properties in terms of composite microstructure.

2. Materials and methods

Spherical Al powder (F3731, Hermillon Powders, Fig. 1a) with an average diameter of 8 μm, to which was added 5 vol% of Al–$Si_{11.3at\%}$ alloy powder (Al–Si; F2071, Hermillon Powders, Fig. 1b) with an average diameter of 100 μm, was prepared as a matrix powder. Graphite flakes (GF; Yanxin-Graphite Co., Ltd., Fig. 1c) at 32 mesh and an average thickness of $30 \sim 50$ μm were prepared as a reinforcement, and mixed with the matrix powder for 5 min to obtain an Al + GF powder.

After compacting the Al + GF powder in a carbon mold, columnar Al–GF composite bulk ($\phi 10 \times 8$ mm^3) was fabricated by hot pressing for 30 min at 600 °C (between the melting points of Al–$Si_{11.3at\%}$ alloy (584.6 °C) and Al (660 °C)) under a uniaxial compressive stress of 60 MPa. The volume fraction of GF in the final Al–GF composite was controlled at 10, 30, and 50 vol%. The hot pressing temperature was monitored via a K-type thermocouple located 2 mm from the sample in the carbon mold. Here the carbon fibers tend to be aligned in the in-plane direction due to the uniaxial compressive stress in the hot press [4,16]. Therefore, a number of fabricated Al–GF columns were vertically machined to ϕ 6×4 mm^3 in order to prepare the specimens for thermal conductivity and TEC investigations in the in-plane direction with graphite orientation (in a–b plane of GF).

The relative density of the Al–GF composites was measured using the Archimedes principle. Microstructural characterization of the Al–GF composite was carried out via scanning electron microscopy (SEM; Tescan, VEGA©) and high-resolution transmission electron microscopy (HR-TEM; JEOL 2000-FX). For TEM observation, thin specimens of Al–50 vol% GF composites were prepared using the ion milling system (GATAN PIPS Model 691), after mechanical polishing with waterproof abrasive silicon carbide papers (#600, #1200, #2000, #4000) to less than 50 μm thickness.

The thermal conductivity of the Al–GF composites (K_c) was estimated using the following equation:

$$K_c = \alpha \times \rho \times C_p \tag{1}$$

where α is the thermal diffusivity of the Al–GF composites, which was measured via the laser flash method (NETZSCH LFA 457, MicroFlash®) at room temperature. The thermal diffusivity of the Al–GF composites was measured parallel and perpendicular to the stress axis (i.e. the transverse and in-plane directions of the GFs). ρ and C_p are the measured density and the heat capacity of the Al–GF composite, respectively. C_p was calculated from the heat capacities of graphite and pure Al by the rule of mixture.

The TEC of the Al–GF composites was measured perpendicular to the stress axis in the hot press (i.e. the in-plane direction of the graphite flakes), under an argon gas flow in two thermal cycles between room temperature and 250 °C with a heating/cooling rate of 2 °C/min, using a TEC measurement system (NETZSCH DIL 402, PC®). TEC values were estimated from the averages obtained in 2 thermal cycles between 100 and 180 °C.

3. Results and discussion

3.1. Microstructure of the Al–GF composites

Fig. 2 shows the relative densities of the Al–GF composites. The relative density of the Al–10 vol% GF composites was 99%, regardless of the addition of Al–Si. For composites with more than 30 vol% GF, the relative density remained higher than 98% with 5 vol% Al–Si, but decreased without the addition of Al–Si. SEM observation revealed that GFs were effectively oriented in the in-plane direction due to the uniaxial compressive stress in the hot press (see Fig. 3a and c) [4,16]. Voids were mainly observed between the GFs and the Al matrix, with the number of voids greater in Al–GF composites fabricated without Al–Si than in those with 5 vol% Al–Si. This result is consistent with the relative density of the Al–GF composites. Without Al–Si, the Al/GF interface seemed intimate and rectilinear (see Fig. 3b), i.e. a similar microstructure to that reported by Chen et al. [20]. In contrast, with 5 vol% Al–Si the Al/GF interface was intricate, as shown in Fig. 3d. It seems that this intricate Al/GF interface is formed by the incorporation of detached graphite fragments into the Al particle boundaries around the Al/GF interface, a process which is thought to be furthered by the transformation of Al–Si to a liquid

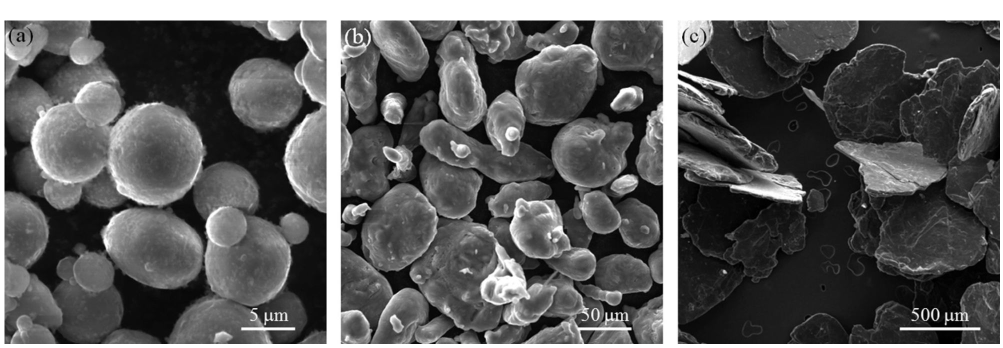

Fig. 1. SEM micrographs of starting materials; (a) Al powder, (b) Al–$Si_{11.3at\%}$ alloy powder, and (c) graphite flakes.