Journal of Alloys and Compounds 756 (2018) 8–18

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Effect of interfacial microstructure on the thermal-mechanical properties of mesophase pitch-based carbon fiber reinforced aluminum composites

Risheng Pei, Guoqin Chen*, Yaping Wang, Ming Zhao, Gaohui Wu

School of Materials Science and Engineering, Harbin Institute of Technology, Harbin, 150001, China

ARTICLE INFO

Article history:
Received 22 October 2017
Received in revised form
18 April 2018
Accepted 30 April 2018
Available online 5 May 2018

Keywords:
Metal-matrix composites (MMCs)
Microstructures
Thermal properties
Mechanical properties

ABSTRACT

Four unidirectional carbon fiber (67.9%–70.0% volume fraction) reinforced aluminum composites (P100/1199, P100/6063, P120/1199, and P120/6063) were fabricated by pressure infiltration and thermo-mechanical properties of these composites were investigated. We found interfacial microstructures, especially free of carbide, precipitations and interfacial gaps, are important for the properties of the composites. With proper addition of Mg and Si element, P100/6063 composite possessed moderate interface. Thus, thermal conductivity (TC), bending strength and elastic modulus of that were improved prominently. TC along fiber direction of P100/6063 was (391±1)W/mK, which was close to 95% of the role of mixtures (ROM) model. The coefficient of thermal expansion values (CTE) along fiber direction of all the specimens varied between -1×10^{-6}/K and 1×10^{-6}/K. The bending strength and the elastic modulus of P100/6063 composite exhibited a 31.6% and 14.3% increase respectively in comparison to those of P100/1199 composite. High TC, low CTE value, and excellent mechanical properties make this material promising for microelectronic chips production.

1. Introduction

With the power level and density of microelectronic chips increasing, thermal management has attracted great interest over the past decades [1,2]. In order to assure these devices reliable and long-life performance, heat dissipation is of great importance [3]. Traditional metal matrix composites (e.g., SiC/Al, Cf/Cu, diamond/Cu et al.) have been widely used in this field because of the devisable coefficient of thermal expansion (CTE) values and low density [4]. However, the thermal conductivity (TC) of SiC/Al composite and the machinability of diamond/Cu composite are limited, which become the biggest barriers for their extensively use. Recently, graphite has been proved to exhibit promising thermal and mechanical properties and widely used as reinforcement in metal matrix composites [5–7]. But because of the high cost and difficulty for production of high-quality graphite, such composites have not been widely investigated and employed. Mesophase pitch-based carbon fiber (MPCF) exhibits ultrahigh TC and low CTE along the fiber axis as well as high workability, which is of great benefit for the unidirectional dissipation of heat and satisfy the special requirements of heat sinks. Therefore, taking TC, CTE values, as well as the machinability and workability, into consideration, MPCF reinforced metal matrix composites are promising for electronic packaging materials. Although quantities of researchers have paid attention to the PAN-based carbon fiber reinforced aluminum or copper composites [3,8–13] and MPCF reinforced copper composites [14], MPCF reinforced aluminum composites have been rarely studied [15,16].

The biggest obstacle for carbon fiber reinforced aluminum composites production is the low wettability between carbon fiber and aluminum. In order to minimize the effect of insufficient wettability on the interfacial microstructure and the properties of composites, three ways are widely used, namely applying fiber coating, modifying matrix materials and adjusting fabrication parameters (e.g., infiltration time, temperature, pressure) [13,17,18]. However, fiber coating is not feasible for the carbon fibers with high modulus since it is easy to destroy the surface of carbon fiber during the coating process [14]. Many researchers have paid attention on matrix alloying to adjust interfacial microstructure and improve properties of composites. Wang et al. [19] has studied the effect of

* Corresponding author.
E-mail addresses: pei@imm.rwth-aachen.de (R. Pei), chenguoqin@hit.edu.cn (G. Chen).

https://doi.org/10.1016/j.jallcom.2018.04.330
0925-8388/

different contents of Mg on the microstructure and properties of composite and found that wettability between carbon fibers and aluminum improved and the formation of carbides were also restrained. Hashim et al. [20] have found that 5 wt% Si addition could decrease the viscosity of aluminum and improve the wettability between the reinforcement and aluminum. Pech-Canul et al. [21] and Aghajanian et al. [22] concluded the function of Mg as decreasing the viscosity and surface tension of aluminum. However, when the addition of Mg exceed 3 wt%, the fluidity of aluminum can reduced [20]. For fabrication methods, although gas pressure infiltration [3] and vacuum diffusion [15] have been successfully used to manufacture carbon fiber reinforced aluminum composites, squeeze casting is easier to adjust the fabrication parameters to improve the wettability.

Thus, in this work, MPCFs reinforced pure aluminum (1199) and Al-Mg-Si aluminum alloy (6063) has been fabricated by pressure infiltration. Microstructures both of the MPCFs and composites were characterized by X-ray diffraction (XRD), scanning electron Microscopy (SEM), and transmission electron microscopy (TEM). Thermo-mechanical properties (TC, CTE values, elastic modulus, and bending strength) were also investigated.

2. Material and methods

2.1. Preparation of composites

In this study, four kinds of composites (P100/1199, P100/6063, P120/1199, and P120/6063) were fabricated by squeeze casting method. P100 and P120 MPCFs were used as reinforcements, whose nominal properties given by the manufacturer are shown in Table 1. As for the matrix, pure aluminum (1199) and Al-Mg-Si alloy (6063) were utilized and the compositions (wt.%) of different alloys are illustrated in Table 2. Before the fabrication, the MPCFs were twined unidirectionally as performs firstly. Then, the alloys were melted at 900 °C and the mold, as well as the fiber perform, were preheated at 600 °C. During the infiltration process, a pressure of 5 MPa was carried on for 10 min, followed by the solidification in the air. After the solidification, the composites sheets were machined from the perform and further annealed under 340 °C for 30 min to reduce the residual stresses between carbon fiber and Al alloy.

2.2. Characterization of microstructure of MPCFs and the composites

The morphology of MPCFs parallel and perpendicular to the fiber axis were observed by SEM (Helios Nanolab600i). The crystal parameters and degree of graphitization of carbon fibers were determined by XRD (Xpert, Philips). Before XRD measurement, samples were ground in an agate mortar, sieved through a 140-mesh screen and then mixed with 10 wt.% Si as an internal standard. Each sample was scanned from 10° to 90° with a speed of 0.25°/min. The peak positions of (002) and (110) crystal planes, as well as half maximum height (FWHM) of these peaks, could be gotten with the help of internal standard after correction (e.g. absorption, Lorenz polarization and atomic scattering factors).

In order to analyze the microstructures of the composites, especially the interface between carbon fibers and matrix, XRD(Xpert, Philips), SEM (Helios Nanolab600i) and TEM (Tecnai G F300), as well as selected area electron diffraction (SAED), were employed. And the energy dispersive X-Ray detector (EDX) was also used to identify the distribution of different elements.

Table 2
The nominal element compositions of aluminum alloys (wt.%).

Alloy	Si	Cu	Mg	Zn	Mn	Ti	Fe	Al
1199	≤0.0025	≤ 0.05	–	–	–	–	≤0.03	Bal.
6063	0.2∼0.6	≤ 0.10	0.45∼0.90	≤0.10	≤ 0.10	≤0.10	≤0.35	Bal.

2.3. Thermo-mechanical properties test of the composites

Cylindrical specimens with a dimension of $\phi 6 \times 3$ mm were cut for the test of thermal diffusivity along fiber direction using laser flash method (Netzsch LFA447). The densities of the composites were measured by Archimedes method. The specific heats of the composites were calculated by Eq. (1). Then corresponding TCs along fiber direction could calculate by Eq. (2). CTE values alongside fiber direction measurements were carried out on Netzsch DIL-402C. The dimension of the specimens was $\varphi 6 \times$ 25 mm and tested temperature ranged from 20 °C to 500 °C with a heating rate of 5 °C/min in an argon atmosphere.

$$C_c = \frac{1}{\rho_c}\left(V_f C_f \rho_f + V_m C_m \rho_m\right) \quad (1)$$

Where, V is the volume fraction; subscripts c, f, and m represent composite, carbon fiber and matrix, respectively.

$$\lambda = \rho C \alpha \quad (2)$$

Where,λ is thermal conductivity, W/mK; ρ is the density of composites, g/cm^3; C is the specific heat of composites, J/gK; α is the thermal diffusion of composites, mm^2/s.

Elastic modulus of composites was measured by a dynamic method using the bending specimen with a dimension of 60 (fiber direction) × 10 × 2 mm on the EMT-01. INSTRON-5569 was continuously employed to measure the bending strength with a span of 40 mm at room temperature.

3. Results

3.1. Microstructures of the composites

Secondary Electron (SE) images of four composites along transverse and longitudinal directions are shown in Fig. 1. Carbon fibers in these composites aligned parallelly, which is the prerequisite for the high thermal-mechanical properties of composites along fiber direction. On these images, the diameter of P100 was obviously larger than P120 and no obvious cast defects were found.

To detect the distribution of Mg and Si in P120/6063 composite, EDX mapping was employed and the results are shown in Fig. 2. Mg and Si elements were solute homogeneously in aluminum, as shown in (b) and (c). No obvious precipitations were found either on the interface or in the matrix with the addition of Mg and Si

Table 1
Nominal properties of carbon fibers.

Properties	σ MPa	E GPa	*Ductility* %	ρ g/cm^3	*Diameter* μm	*CTE* 10^{-6}/K	*TC* W/mK
P120	3530	920	0.3	2.19	7	−1.5	600
P100	3430	860	0.4	2.19	10	−1.5	500