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Microstructure evolution during fabrication and microstructure–property relationships in vapour-grown carbon nanofibre-reinforced aluminium matrix composites fabricated via powder metallurgy

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ABSTRACT

Microstructure evolution of vapour-grown carbon nanofibre (VGCF)-reinforced aluminium matrix composites during fabrication and microstructure–property relationships of these materials were studied. Composites were fabricated using powder metallurgy, i.e. by mixing VGCFs and aluminium powder via ball-milling followed by sintering or hot extrusion. The mixing condition was selected to achieve small powder particle size and homogeneously dispersed VGCFs. Aluminium grains and VGCFs were elongated along the longitudinal direction of aluminium particles in the mixed powder. Detailed observation of the aluminium grains in the composites found grain size and morphology dominated by recrystallization. Apparently, grain growth was inhibited by VGCFs. Theoretical models considering strength increment due to grain refinement resulting from VGCF addition, load bearing of VGCFs, thermal mismatch of VGCFs and aluminium and Orowan effect were developed. Theoretical values coincided well with hardness, yield strength, and ultimate tensile strength of the composites, and thus the models could precisely explain the microstructure–property relationships.

1. Introduction

Aluminium matrix composites reinforced by vapour-grown carbon nanofibres (VGCFs) are attractive materials due to their expected high strength, ductility and thermal conductivity [1–10]. These properties originate from the superior properties of the VGCFs. In addition, VGCFs have larger diameters than multi-walled carbon nanotubes (MWCNTs) [11] and can thus be relatively easily dispersed in aluminium matrices. Recently, various methods for the fabrication of aluminium matrix composites reinforced by VGCFs have been proposed. Powder metallurgy, i.e. mixing the VGCFs and aluminium powder followed by sintering or plastic forming, such as hot extrusion [8] or hot rolling [9], is an effective method for obtaining composites with superior properties, because the fabrication process is not affected by the wettability of the VGCFs and the aluminium [4]. Xu et al. [1] fabricated VGCF-reinforced aluminium matrix composites by wet mixing of VGCFs and aluminium powder of different sizes followed by spark plasma sintering (SPS) for consolidation of the mixed powder. A high Vickers hardness and a low coefficient of thermal expansion were achieved using smaller aluminium powder particles due to the excellent dispersion of the VGCFs in the aluminium matrix. However, poor bonding at the VGCF/aluminium matrix interface resulted in composites with low strength. Choi et al. [8] fabricated MWCNT-reinforced aluminium matrix composites via mechanical mixing of MWCNTs and aluminium powder followed by hot extrusion. The composite strength was dramatically improved by mixing the aluminium and MWCNTs using a ball-mill due to refinement of the aluminium grains. However, the reduction in the length of the reinforcement and the variation of the aluminium grains due to extrusion were not fully investigated. Past studies have revealed that the properties of reinforced aluminium composites are strongly dominated by their microstructures, i.e. aluminium grains; the length, distribution and orientation of the reinforcing material; the characteristics of the interface between the reinforcing material and the aluminium matrix [12–16]. However, variation of the microstructures in the aforementioned aluminium matrix composites (using VGCFs or MWCNTs as reinforcements) during fabrication has not been sufficiently investigated, and the relationships between the microstructures and the properties of these composites are also not yet fully understood. In particular, while several investigations of MWCNT-reinforced aluminium matrix composites have appeared in the literature

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[17,18], few studies have been reported on microstructure–property relationships for VGCF-reinforced aluminium matrix composites.

Herein, we describe the results of a study of VGCF-reinforced aluminium matrix composites prepared via the ball-milling of VGCFs with aluminium powder followed by SPS or hot extrusion. The evolution of the microstructure, i.e. the variations of the aluminium grains and the VGCF length, due to mixing, sintering and hot extrusion were investigated. The microstructure–property relationships and the mechanism for reinforcement of the composites are also discussed.

2. Experimental

2.1. Mixing of VGCFs and aluminium powder

First, the conditions for mixing VGCFs and aluminium powder for the preparation of the composites were investigated. The VGCFs (Showa Denko Co. Ltd, Japan; average diameter: 150 nm, average length: 15 μm) and aluminium powder (Kojundo Chemical laboratory; average diameter: 30 μm) were mixed using a planetary ball-mill (Fritsch Pulverisette 5). Table 1 lists the mechanical and thermal properties of the VGCFs and aluminium. The VGCFs and aluminium powder (total weight: 40 g) were placed in a stainless-steel jar containing stainless-steel balls (800 g). Therefore, the weight fraction of the stainless-steel balls to the mixture of VGCFs and aluminium powder was 20:1. To prevent excessive cold welding of the aluminium powder, stearic acid ($C_{17}H_{35}COOH$, 0.6 g) was added as a process control agent. The VGCF volume fraction in the powder was 0% or 3%. The stainless-steel jar was filled with argon gas and rotated at 200 rpm for 0.5, 1, 2, 3, or 10 h. Rotation for 20 min followed by suspension for 40 min was repeated when the milling duration was greater than or equal to 1 h. During mixing, an argon atmosphere inside the jar was maintained. The mixed powders were recovered after milling, and their morphology was observed via scanning electron microscopy (SEM) using JSM-6060 (JEOL). Subsequently, each powder was embedded in a commercial epoxy resin in order to observe its interior. The resins were polished and their surfaces were etched using nitric acid in order to study the distribution and the length of the VGCFs inside the powders. The aluminium grain sizes in the powders were evaluated via X-ray diffraction (XRD) analysis using Rint-Ultima III (Rigaku). The accelerating voltage and filament emission were 40 kV and 40 mA, respectively, and a copper (Cu Kα) radiation source was used. The Williamson–Hall method [7] was employed to estimate the grain sizes, which were calculated by fitting the following equation to the experimental results:

$$B' \cos\theta = \frac{\lambda}{L} + \eta \sin\theta \tag{1.1}$$

$$B' = (B_T^2 - B_I^2)^{\frac{1}{2}} \tag{1.2}$$

Table 1
Properties of the aluminium powder and VGCFs.

Property	Aluminium	VGCF
Geometry	Average diameter 30 μm	Average diameter 150 nm Average length 15 μm
Density	2.7 g/cm^3	2.0 g/cm^3
Young's modulus	76.0 GPa	516.5 GPa [20]
Tensile strength	100 MPa	3100 MPa [20]
Coefficient of thermal expansion	$23.6 * 10^{-6}$ K^{-1} [10]	$4.0 * 10^{-6}$ K^{-1} [10]

where B_T is the broadening, i.e. the breadth at half maximum intensity, of each peak in the X-ray diffraction patterns for the powders, B_I is the instrumental broadening of the peaks in the XRD pattern of the standard sample (aluminium powder annealed at 500 °C for 10 h), B' is the corrected broadening, λ is the wavelength of the X-rays, θ is the Bragg's angle, L is the grain size and η is the lattice strain stored during the milling process. The optimum mixing condition for the fabrication of the composites was determined from the morphologies of the mixed powders, their sizes, the VGCF distributions in the powders, and the lengths of the VGCFs. Using the optimized condition, we then prepared composites with VGCF volume fractions of 0, 0.5 and 1 vol.%. After mixing, these powders were embedded in epoxy resins (G1 epoxy; Gatan) in order to directly observe the aluminium grains. The resins were also polished and their surfaces were finished using a cross-section polisher (CP; JEOL). The finished surfaces of the resins containing the embedded powders were then observed via field emission scanning electron microscopy (FESEM) using JSM-7001F (JEOL).

2.2. Fabrication of composites

Some of the mixed aluminium and VGCF powders were consolidated using SPS (Dr. Sinter; SPS Syntex) in a vacuum. A carbon die and a punch were used for sintering. The powder was placed in the die, pressed with the punch, heated to 600 °C at the rate of 20 °C/min, and then, sintered under an applied pressure of 50 MPa for 0.5 h (total heating time = 1 h). The volume fraction of the VGCFs in the sintered composites was 1 vol.%.

For the composites that were prepared using extrusion, the mixtures of aluminium and VGCF powders were encapsulated in aluminium A1050 containers (outer diameter: 40 mm, inner diameter: 24 mm, height: 40 mm, depth: 37 mm) in order to avoid oxidation. Each powder was first placed in the container and then compressed using a steel punch, and subsequently, the container was evacuated using the SPS system and an aluminium lid was installed by applying a force. The container was then machined to a diameter of 30 mm and served as an extrusion billet. A 60° conical die was used for the extrusion step, which was performed at 550 °C at an extrusion ratio of 9. It should be noted that the temperature was raised to 500 °C over 0.5 h and then to 550 °C over another 0.5 h in order to avoid overheating. The temperature was then held constant at 550 °C for 0.5 h prior to initiation of the extrusion process. The volume fraction of VGCFs in the extruded composites was 0.5 vol.%. In order to evaluate the changes in the microstructure and mechanical properties of the different samples, aluminium-sintered products and aluminium-extruded rods were also fabricated for comparison.

2.3. Characterization of composites

The aluminium grains in the composites were observed as follows: The composites were cut into 1.5-mm-thick, half-moon-shaped pieces, which were polished and finished using the CP. The composites fabricated via SPS or hot extrusion were observed via FESEM. Several analyses were also performed in order to evaluate the mechanical properties of the composites. The Vickers micro-hardness was determined for the samples by using an indentation load of 5 N and a loading time of 5 s. The tensile strength of the hot extruded composites was also determined. Samples were machined to create tensile specimens in accordance with Japanese Industrial Standard (JIS) Z 2241, and tensile testing was performed on a tensile testing machine (Shimadzu; AG-100KNC) by using a 0.5 mm/min crosshead speed. The strain in the composites was also measured using an ultra-high-elongation foil strain gauge (Kyowa). After tensile testing, the fracture surfaces of the tested samples were observed via SEM.