Materials Science & Engineering A 632 (2015) 72–77

Contents lists available at ScienceDirect

Materials Science & Engineering A

journal homepage: www.elsevier.com/locate/msea

Mechanical properties of nanodiamond and multi-walled carbon nanotubes dual-reinforced aluminum matrix composite materials

Hansang Kwon [a,*], Gil-Geun Lee [a], Sung-Gyoo Kim [a], Byung-Woo Lee [a], Won-Chan Seo [a], Marc Leparoux [b]

[a] Department of Materials System Engineering, Pukyong National University, 365 Shinseonro, Nam-gu, 608-739 Busan, Republic of Korea
[b] Empa, Swiss Federal Laboratories for Materials Science and Technology, Advanced Materials Processing, Feuerwerkerstrasse. 39, CH-3602 Thun, Switzerland

ARTICLE INFO

Article history:
Received 6 October 2014
Received in revised form
15 January 2015
Accepted 22 February 2015
Available online 5 March 2015

Keywords:
Nanodiamond (nD)
Multi-wall carbon nanotube (CNT)
Vapor-grown carbon nanofiber (VGCF)
Mechanical properties
Powder metallurgy

ABSTRACT

Nanodiamond (nD) and multi-walled carbon nanotubes (CNT) was used to fabricate reinforced pure aluminum (Al) matrix composites by mechanical ball milling and hot-pressing. Pure Al bulk and each single nanoparticle (nD or CNT) reinforced composites was also fabricated for comparison. Micro-Vickers hardness was measured for single (nD or CNT) and dual nanoparticle (mixture of nD and CNT)-reinforced Al matrix composites and showed maximum values that were approximately six times higher than those of pure Al bulk. Four-point bending behaviors were also discussed and the dual nanoparticles-reinforced Al matrix composites showed highest value of the flexural strength (about 760 MPa). The fracture surfaces that resulted from the bending test were also observed. Moreover, the crystallite size and lattice strain of the Al particles in the composites were described by the Scherrer equation.

1. Introduction

High strength structural materials are required in many industrial fields such as building, automobiles, and aviation technology to produce stronger, more ductile and lighter engineering materials parts. Carbon nanotubes (CNTs), discovered in 1991, have high mechanical (tensile strength: ~150 GPa and Youngs modulus: ~5 TPa) [1,2], electrical (electric current density of 4×10^9 A/cm^2, 1000 times greater than copper) [3], and thermal (Thermal Conductivity at 25 °C (W/mK): 2000–6000 [4]) properties than conventional materials [5]. Therefore, CNTs are attractive candidate materials for the next generation of metal–matrix composites used for high performance structural reinforcement [6,7]. Nanodiamond (nD) is also considered to be useful for composite reinforcement because of its exceptional hardness (10,000HV) [8]. Despite growing scientific interest, CNT or nD reinforced metal–matrix composite materials are not yet scientifically or technically well-characterized to achieve commercialization [9]. Because, nanosized the CNT and the nD materials are very difficult to handle and homogeneously disperse within matrix materials due to their strong van der Waals force and very fine size effect, resulting in highly agglomerated nanoparticle-reinforced composite materials [10–13]. Moreover, there is still a lack of perfect fabrication process for the CNT and/or the nD reinforced-composite materials to use as a commercial engineering parts. Thus, it is necessary to overcome those above mentioned problems through the variety of fabrication and characterization trial.

In this study, we investigated the possibility of fabricating dual (nDs mixed with CNTs) nanoparticles-reinforced aluminum (Al) composite materials using high energy mechanical ball milling followed by hot-pressing. Pure Al bulk and each single nanoparticle (nD or CNT) reinforced composites was also fabricated for comparison. Detonation-synthesized nDs and vapor-grown carbon nanofibers (VGCFs) (i.e., as a kind of multi-walled carbon nanotubes synthesizes by catalytic chemical vapor deposition) were selected as reinforcing materials. Pure Al was used as a matrix material exclude effects from additives. However, mechanical properties of the dual nanoparticles-reinforced composite materials were mainly investigated based on microstructure observation, crystallite size analysis, Vickers hardness and flexural strength measurements.

2. Experimental

VGCFs (Showa Denko, Japan, purity 99%, specific gravity of 2.1 g/cm^3, mean diameter of 100 nm, length less than 15 μm), detonation-synthesized nDs (Reishauer AG, Switzerland, purity 99.5%, specific gravity of 3.5 g/cm^3, mean diameter of 50 nm), and gas-atomized pure Aluminum (Al) powder (ECKA Granules, purity 99.5%, particle size less than 63 μm) were used for

* Corresponding author. Tel.: +82 51 629 6383; fax: +82 51 629 6373.
E-mail addresses: kwon13@pknu.ac.kr, nanocomposites@hotmail.com (H. Kwon).

http://dx.doi.org/10.1016/j.msea.2015.02.057
0921-5093/

fabrication. The Al powder and 1 volume fraction (%) of the nanoparticles (nD or/and VGCF) were mixed in a planetary ball mill (PM400, Retsch GmbH, Germany) for 3 h in an argon atmosphere at 360 rpm using ∅10-mm balls, a 10:1 ball-to-powder weight ratio, and 20 wt% heptane as a process control agent. At the end of the process, the milling jar containing the composite powder blend was transferred into a glove-box containing a controlled inert argon gas atmosphere to passivate the composite powder for almost one week. The ball milled composite powders were in a highly activated energy state and could have oxidized/burned if they were to come in direct contact with air. After passivation, the single and dual nanoparticle powder mixtures were used to fill a high-temperature steel mold. The mold was then heated in vacuum to 550 °C for 1.5 h and pressed. A uniaxial pressure (LFV 400 kN, Walter+Bai AG, Switzerland) of 570 MPa was then applied for 4 s. The resulting samples were 30 mm in diameter and approximately 5 mm in thickness. The densities of the single and dual nanoparticle-reinforced composites were measured using Archimedes' principle according to ISO 3369:1975. Micro-Vickers hardness (Zwick ZHU 2,5 Martens hardness tester, Zwick Roell Group, Germany) was measured according to standard EN-ISO 14577:2003 with a load of 10 kg, a loading speed of 0.2 mm/min, an unloading speed of 0.1 mm/min, a maximum load of 100 N and a dwell time of 15 s. At least five measurements were performed for each sample. A four-point bending test was carried out using a Walter+Bai 150 kN servo-hydraulic test machine with DionSTAT software at a loading speed of 1 mm/min. The sample dimensions were approximately $3 \times 5 \times 22$ mm^3. The microstructures of the composites were observed by high-resolution cold field emission scanning electron microscopy (FE-SEM S-4800, Hitachi, Japan) and high-resolution transmission electron microscope (HR-TEM, Hitachi, Japan) with selected area diffraction pattern (SADP, under 10 nm nano-beam spot size and with camera lens 1.2 m from the specimen). XRD diffraction patterns were measured using an X'Pert Pro diffractometer (PANAlytical, Netherlands) with Cu-K_α radiation ($\lambda = 1.54056$ Å, 45 kV and 40 mA) in the 2θ range of 20–80° using a linear detector (X'Celerator). A step size of 0.0167° and a scan rate of 0.05°/s were used. The crystallite size and lattice strain were calculated using the Scherrer equation [14].

3. Results and discussion

The raw materials of the irregular shape of gas-atomized pure Al powder, the relatively straight morphology of the VGCFs and the synthesized nD particles are shown in Fig. 1. Some of the raw VGCFs were highly bent and contained particle-shaped substances (mainly carbon species or catalytic impurities, see the white arrow in Fig. 1c). According to our FE-SEM observations of the raw materials (Fig. 1a and e), the distribution of Al particle size is much larger than the distribution of nD particle size (mean size of 50 nm). Moreover, the VGCF aspect ratio is much different than the Al and nD particle aspect ratio (Fig. 1a, b, and e). These large differences in particle size and shape lead to difficulties in homogeneously mixing them by low energy mixing process. Therefore, we employed high energy ball milling to physically disperse the materials.

Size of the Al–1 vol%nD–1 vol%VGCF composite particles was larger than the raw pure Al particles after mechanical ball milling (Figs. 1a and 2a). The larger particles are formed by agglomeration of several finer particles as shown in Fig. 2b. A very few VGCFs were found in the ball milled composite particles and it was also very difficult to distinguish the nD particles. It is estimated that the low concentrations (1 vol%) of nD and VGCF were contributed to difficult observe and they were also trapped by the soft Al matrix during the high energy ball milling process. In some cases, this result is preferable because if we could find the large amount of nanoparticles by the SEM resolution level that would indicate that they were not well-dispersed in the composite powder. However, the distribution behavior of nDs and VGCFs in the Al particles will be carefully investigated in subsequent studies.

The hot-pressed Al–1 vol%nD–1 vol%VGCF composite had a relative density of over 99% (Table 1). Fig. 3 shows FE-SEM micrographs of the chemically etched cross-section of the hot-pressed composite. It was not possible to observe clear boundaries in the composite without deep etching. As shown in Fig. 3a, some of the grains appear elongated; this morphology was mainly induced after application of pressure in the vertical direction. Many pores were created after chemical etching and the pores were found in the every grain. Etchant grooves or pores are usually formed at the interface between phases or some of within particle (see the white arrow in Fig. 3b and c), in areas containing impurities or in regions where stress accumulates [15]. The presence of pores within the grains (Fig. 3b) implies that the grains are composed of multiple phases (Al-nD-VGCF by mechanical ball milling) that induce residual stress in the material. Because the composite is containing 1 vol% nD and 1 vol% VGCF. The VGCFs were found along the grain boundaries and maintained their original shapes and sizes (Fig. 3e and f).

Further microstructural analysis was conducted via TEM. We observed many nanocrystallites in the Al–1 vol%nD–1 vol%VGCF composite, as shown in Fig. 4. The nanocrystallites are mainly attributed to the presence of nD and VGCF (black arrows in Fig. 4b). Furthermore, the relatively well dispersed state seen in the Al matrix means the employed high energy ball milling process was useful. Some nanosized aluminum carbide (Al_4C_3) was also observed and confirmed using SADP, as shown in Fig. 4b. It was suggested that nanosized Al_4C_3 formation helped load transfer from matrix to CNT using a trust chemical link [16,17]. However, Al_4C_3 induction of the load transfer effect should be very carefully considered because of its hygroscopic and brittleness properties.

The crystallite size of Al in pure Al, the single nD or VGCF nanoparticle-reinforced Al bulk matrix, and the nD and VGCF dual nanoparticle-reinforced Al bulk matrix was determined using the parameters from the XRD patterns according to the Scherrer equation [14], as shown in Eq. (1),

$$d = 0.9\frac{K \cdot \lambda}{B \times \cos\theta} \tag{1}$$

where d is the crystallite size and λ, θ, and B are the X-ray wavelength, the Bragg scattering angle, and the full width at half maximum (FWHM), respectively. K is a constant that depends on the crystallite shape (0.9). The FWHM values were measured from the XRD patterns and were calibrated using a standard sample (CeO_2 NIST SRM-674b). The crystallite size and lattice strain were three times larger and two times smaller, respectively, for pure Al as compared to the Al–1 vol% nD–1 vol%VGCF bulk composite. However, the Al bulk composites reinforced with either nD or VGCF had smaller crystallite sizes and larger lattice strain values than the pure Al bulk material (Table 1). This was due to the pinning effect [18] of the nanoparticles. In general, particles or some impurities could restrain grain growth and also induced high stress accumulation that is why we could achieve relatively high lattice strain values as indicated in Table 1 [18]. The spherical nDs were more easily dispersed than the VGCFs. The crystallite size and lattice strain values for the bulk dual nanoparticle-reinforced Al composite were is larger and smaller, respectively than the powder due to grain growth and the release of residual stress at the processing temperature. Slight increases in the lattice strain values were attributed to lattice defects such as vacancies, substitutions, and interstitial atomic impurities [19]. In other words, the presence of two types of nanoparticles caused the crystallite size to decrease and the lattice strain to increase slightly. According to the Hall–Petch equation [20], materials hardness improve as the crystallite (grain) size