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Functionally Graded Dual-nanoparticulate-reinforced Aluminium Matrix Bulk Materials Fabricated by Spark Plasma Sintering

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Functionally graded (FG) carbon nanotubes (CNT) and nano-silicon carbide (nSiC) reinforced aluminium (Al) matrix composites have been successfully fabricated using high-energy ball milling followed by solid-state spark plasma sintering processes. The CNTs were well-dispersed in the Al particles using the nSiC as a solid mixing agent. Two different types of multi-walled CNTs were used to add different amounts of CNTs in the same volume. The ball milled Al−CNT−nSiC and Al−CNT powder mixtures were fully densified and demonstrated good adhesion with no serious microcracks and pores within an FG multilayer composite. Each layer contained different amounts of the CNTs, and the nSiC additions showed different microstructures and hardness. It is possible to control the characteristics of the FG multilayer composite through the efficient design of an Al−CNT−nSiC gradient layer. This concept offers a feasible approach for fabricating the dual-nanoparticulate-reinforced Al matrix nanocomposites and can be applied to other scenarios such as polymer and ceramic systems.

KEY WORDS: Carbon nanotubes (CNT); Silicon carbide; High-energy ball milling; Spark plasma sintering (SPS); Functionally graded materials (FGM)

1. Introduction

The concept of functionally graded materials (FGM) was suggested in the early 1980s from Japan[1−3]. This concept states that it is possible to control the different properties at each layer within a bulk material by varying the composition design. The coating process can also control the surface properties of materials, but generally, the final coat must be within the micrometer range[4]. One of the common problems with coated materials is that the coating layer and matrix easily undergo delamination under the stress that arises from the environment due to their difference in physical and chemical properties[5,6]. In the case of FGMs, it is possible to reduce delamination because each layer of different compositions within a bulk can be recognised as the same unit by efficient and smooth control of the composition over a range of dozens of centimetres. Since the discovery of carbon nanotubes (CNTs) in 1991[7], they have received significant attention due to their remarkable physical and chemical properties[8−10]. However, production of CNTs reinforced materials for commercialised industrial parts remains distant due to the lack of a suitable fabrication process, control of the interface properties between the CNT and matrix, and homogeneous dispersion of the CNT in the matrix materials[11−14]. Many methods have been applied to overcome these problems, and some solutions are being discovered[15−20]. Recently, an efficient method for CNT dispersion in aluminium (Al) powders was proposed, using a nano-SiC (nSiC) particle as a mixing agent during a ball milling process[21]. It seemed to be physically easy for the nearly spherically shaped nSiC to infiltrate the agglomerated line shape CNTs in the Al powder, resulting in better dispersity.

In this study, we fabricated four functionally graded (FG), Al−CNT−nSiC multilayer reinforced bulk materials with various composition layers dispersed using nSiC particles. In particular, the use of nSiC as the dispersion agent was expected to yield synergistic effects due to its nanosize particle distribution, such as fine particle dispersion strengthening. Spark plasma sintering (SPS) was utilised for fabrication of the FG dual-nanoparticulate-reinforced multilayer bulk materials, and

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samples obtained from SPS were analysed based on their microstructure and micro-hardness properties.

2. Experimental

Two types of multi-walled CNTs (Baytubes C150P, Bayer Material Science, purity 99.5%, mean diameter of 20 nm, length of 10 μm and Hodogaya Chemical Co. Ltd. purity 99.5%, mean diameter of 100 nm, length of 20 μm), gas-atomised pure Al powder (ECKA Granules, purity 99.5%, particle size below 63 μm) and nano-silicon carbide (nSiC) were used as starting materials. The smaller size CNT (20 nm) was selected in order to add more CNTs in a large volume per unit. The nSiC particles used as the mixing agent were produced in an inductively coupled plasma (ICP) reactor by a process described in detail elsewhere[22], producing an average particle size between 20 and 30 nm. The Al powder and average size (100 nm) CNTs with an nSiC mixing agent were mixed in a planetary ball mill (Retsch GmbH, PM400) for 3 h under an argon atmosphere at 360 r/min using Ø10 mm balls at a 10:1 ball to powder weight ratio and 20 wt% heptane as a process control agent. Two compositions of Al−10 vol.% CNT−30 vol.% nSiC and Al−30 vol.% CNT−10 vol.% nSiC were prepared. A fixed 10 vol.% of CNTs with a mean diameter of 20 nm was also prepared under the same conditions.

At the end of the process, the powder blend was transferred to a glove-box with a controlled, inert atmosphere of argon. After passivation in the glove-box, the powders were assembled in a layered structure inside a die of 15 mm in diameter, with compositions ranging from 10 vol.% CNT, pure Al, Al−10 vol.% CNT with 30 vol.% nSiC and Al−30 vol.% CNT with 10 vol.% nSiC, followed by treatment with a spark plasma sintering device (SPS-S515) manufactured by Sumitomo Coal Mining Co. Ltd.

The sintering conditions were: a maximum temperature of 600 °C, a holding time of 20 min, a heating rate of 40 °C/min, and a pressure of 50 MPa. The resulting functionally graded (FG) composites had a diameter of 15 mm and a thickness of approximately 10−20 mm depending on the number of layers. The density of the FG-composites was measured by the Archimedes' principle according to ISO 3369:1975. The micro-Vickers hardness of the FG-composites was measured according to EN ISO 6507-1 with loads of 20 and 0.02 kg for 15 s (220, GNEHM Härteprüfer AG and Paar MTH4 microhardness-tester). At least five measurements were made per sample.

The microstructure of the composites was observed by high-resolution cold field emission scanning electron microscopy (Hitachi, HRCFE-SEM S-4800) and high-resolution transmission electron microscopy (HR-TEM, Hitachi, Japan) using selected-area diffraction patterns (SADP). X-ray diffraction (XRD) patterns were measured using an X'Pert Pro diffractometer (PANAlytical) with a Cu-$K\alpha$ radiation source ($\lambda = 0.15148$ nm, 35 kV and 40 mA) in the 2θ range of 20°−80° using a linear detector (X'Celerator). A step size of 0.02° and a scan rate of 0.05°/s were used. The crystallite size was calculated by the Scherrer equation[23]. Raman spectroscopy was performed using a red He-Ne ion laser with a wavelength of 633 nm (Leica) to evaluate the disorder in the CNTs.

3. Results and Discussion

FE-SEM micrograph of the raw Al particles shows irregular shape and several size distributions (Fig. 1(a)). The nSiC mixing agent shows an aspect ratio close to 1 and an overall spherical morphology (Fig. 1(b)). The CNTs of 20 nm in mean diameter were extremely agglomerated, and the thickness of one-side of the multi-wall was approximately 2 nm, as shown in Fig. 1(c).

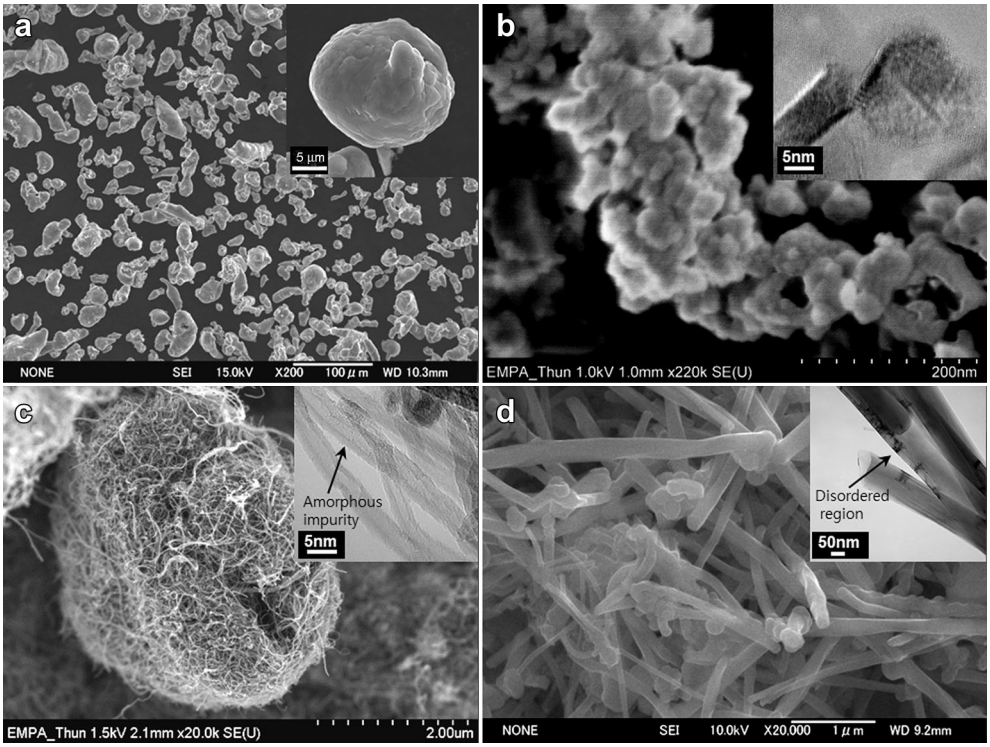

Fig. 1 FE-SEM micrographs of (a) raw Al particles, (b) nSiC, and CNTs with a mean diameter of (c) 20 nm and (d) 100 nm.