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Improving the mechanical properties of carbon nanotubes reinforced pure aluminum matrix composites by achieving non-equilibrium interface

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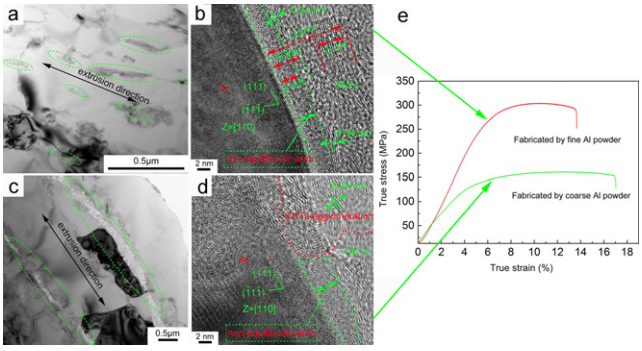
HIGHLIGHTS

- Pure Al powder and CNTs were used to fabricate CNTs reinforced Al matrix composites.
- A non-equilibrium interface was obtained through SPS sintering and hot extrusion.
- The new interface keeps the non-equilibrium state prior to the formation of Al_4C_3.
- The new interface can greatly improve the tensile properties of the composite.
- The fine Al powder is preferred to achieve a high performance of the composites.

GRAPHICAL ABSTRACT



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ABSTRACT

The weak interfacial bonding between carbon nanotubes (CNTs) and Al matrix is a critical issue for the achievement of high strength and good ductility of Al/CNTs composites, and thus hinders the wide application of the composites. Here we obtained a non-equilibrium interface that can provide tight interfacial bonding between the CNTs and Al matrix in the Al/CNTs composites fabricated through spark plasma sintering (SPS) and subsequently hot extrusion. This special interface, accompanied by small grain size, the uniform dispersion and the integrity of the CNTs, can significantly improve the mechanical properties of the Al/CNTs composite. Additionally, the effect of initial Al matrix powder size on the mechanical properties of the composites were investigated. The results indicate that the size of the initial matrix powder affected the dispersion of the CNTs and the interface between the CNTs and Al matrix, and thus influenced the mechanical properties. This work provides a new designation for the high performance of metal based composites with excellent interfacial bonding strength.

1. Introduction

Since the first discovery by Iijima in 1991 [1], both experimental measurements and theoretical calculations have shown that carbon nanotubes (CNTs) possess high Young's modulus and strength, large aspect ratio, and light weight [2–5]. The excellent overall properties combined with low density make CNTs the ideal reinforcements to design nanocomposites with excellent mechanical properties. In the past two decades, a lot of researchers dedicated to develop CNTs reinforced Al based composites [6–9]. However, only a few successful attempts for the high performance of the CNTs reinforced Al matrix composites

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have been reported [10–12]. One of the main reasons is the difficulty in achieving good interfacial bonding. More recently, Kwon et al. [13] claimed that Al_4C_3 was formed on the surface of the tip of CNTs and this helped the stress transfer from the Al matrix to the CNTs. However, it is believed that the interfacial strengthening effect by the formation of Al_4C_3 might be finite due to two main reasons. On one hand, just as Ci et al. [14] demonstrated, the carbide formation is detrimental to the integrity of the CNTs because these reactions tend to form notches along the nanotube surface. On the other hand, the rhombohedral structure of Al_4C_3 with unit cell parameters of a = 0.334 nm and c = 2.50 nm [15] shows relative large difference to the cubic structure of Al matrix ($a = 0.4049$ nm). As a result, the interfacial strains between Al_4C_3 and Al along both *a* and *c* directions of the Al_4C_3 will be large and are detrimental to the interfacial bonding strength.

Previous studies [16–18] indicated that maintaining a special state of the microstructure in materials through the non-equilibrium processing can significantly improve their physical and mechanical properties. That inspired current group that keeping the non-equilibrium state of the interfacial area between the Al matrix and the CNTs, instead of forming stable phase such as Al_4C_3, might lead to a notable property improvement of the composites. In general, the non-equilibrium processing of materials requires an extremely high cooling rate and short processing time. As a novel sintering technique, the spark plasma sintering (SPS) technique can provide extremely fast cooling rates (>500 °C/min), a short holding time at a relative low sintering temperature [19–21], which might be proper to fabricate the desired interface in the present study. In addition to the interfacial bonding strength, it is also necessary to investigate the size effects of initial Al powder on the microstructures and properties of the Al/CNTs composites which is rarely studied in the reported literatures. Eventhough these effects were studied thoroughly in the traditional particle reinforced Al based composites, that in the Al/CNTs composite might be different due to the much larger difference in the aspect ratio between Al and CNTs [22–24].

In this article, a combination of SPS and subsequent hot extrusion was used to fabricate the CNTs reinforced pure Al matrix composites, and the uniform dispersion of the CNTs was realized by flake powder metallurgy technique [25]. One of the goals of the present work was to obtain the non-equilibrium interface to improve the mechanical properties of the composites. In addition, this study also investigated the effects of the Al matrix powder size on the mechanical properties and microstructures of the composites. This work provides a complete new direction for designing a strong interfacial bonding between Al and CNTs.

2. Experimental

2.1. Composite fabrication

In this study, two series of the composites were fabricated. The classification of the two series was based on the matrix Al powder with a mean particle size of 20 μm and 2 μm, respectively. The fabrication procedures are identical for both series of the composites. Commercial Al powder with a purity of 99% and multi-walled CNTs (purity 98%, O.D. × I.D. × L: 10 nm ± 1 nm × 4.5 nm ± 0.5 nm × 3– 6 μm), supplied by Sigma-Aldrich Co. LLC and produced via catalytic chemical vapor deposition (CCVD) process, were used as the raw materials in the present study. The raw Al powder was pretreated by ball milling with a speed of 300 rpm for 10 h in the liquid medium of pure ethanol and the ball-to-powder weight ratio was 10:1. During ball milling, the spherical Al powder particles with micro size in diameter can be transformed into flake shaped particles, which can promote the attachment of the CNTs to the Al particle surface during mixing. The CNTs was ultrasonic treated in ethanol for 1 h in order to decrease the agglomeration. The pretreated Al and 1 vol.% multi-walled CNTs (MWCNTs) powder were then ball-milled for 5 h in a planetary ball mill under argon atmosphere using the pure ethanol as the liquid medium. The rotation speed and the ball to powder weight ratio was 300 rpm and 5:1, respectively. Then the mixed powders were dried at 75 °C for 5 h in a vacuum oven. The powder mixture was sintered by SPS using an FCT HPD 25/3 spark plasma sintering furnace at a sintering temperature of 630 °C for 0.5 h and a pressure of 30 MPa in a vacuum condition of 5 Pa, with the cooling rate being larger than 500 °C/min. Prior to hot extrusion, the as sintered billets (26 mm in diameter) after SPS sintering was preheated at 500 °C for 0.5 h under argon atmosphere. The extrusion ratio was 5:1 and the cross head speed was 3 mm per second, respectively. The diameter of the extruded rods was 11.5 mm and no crack was observed on the surface of the rods. For each type of the composites, pure Al was fabricated as the comparison material using the same method of preparing Al/CNTs composites in present study.

2.2. Structural characterization and mechanical testing

The morphologies of the raw and ball milling treated Al powders, the CNTs and their powder mixtures with Al, and Al-CNTs interface in the extruded composites were observed by scanning electron microscopy (SEM, Quanta FEG 250, FEI) and transmission electron microscopy (TEM, JEM-2100F, JEOL). The microscopic laser Raman spectrometer (LabRAM, HR800, HORIBA) was used to examine the crystal structural information of the CNTs during processing. The line scan in the Al-CNTs interfacial area was conducted by the Titan G2 60–300 transmission electron microscopy with image corrector under the HADDF-STEM mode. The grain information of the Al matrix in the two types of composites was investigated by the electron backscatter diffraction (EBSD) technique. An energy dispersive spectrometer (EDS) attached to TEM was used to analyze the distribution of the Al and C elements in the interfacial area. The TEM and EBSD samples were prepared through an HELIOS NanoLab 600i Focus ion beam thinner. To measure the tensile properties at room temperature, the specimens were machined to a dog-bone shape with the tensile axis along the extrusion direction. The gauge length and the diameter of the tensile samples was 40 mm and 3 mm, respectively. The average tensile strength was measured from 3 specimens using an Instron 3369 universal testing machine with a strain rate of 5×10^{-4} s^{-1}. To further figure out the effects of initial Al particle size on the mechanical properties and microstructures of the composites, the fractural surfaces after tensile testing were observed by Helios Nanolab G3 UC scanning electron microscopy.

3. Results

3.1. Microstructure of the powders

Fig. 1a and b shows the SEM images of the as-received and the ball milled fine Al powders, respectively. It can be seen from Fig. 1a that the fine Al powder has a spherical shape with a size ranging from 1 to 5 μm. From Fig. 1b, most of the round shaped particles were transformed into irregular shape (flake-like) after ball milling, and the size of the flake-shaped particles was larger than the initial raw particles. The morphologies of the as-received and the ball milled coarse powders were depicted in Fig. 1c and d, respectively. Fig. 1c shows that the coarse Al powder also has a spherical shape with a size ranging from 10 to 30 μm. Fig. 1d reveals that the coarse initial particles were also transformed into flake-shaped particles, but the size of the ball milled particles is smaller than that of the raw particles. Usually, the starting Al powders are spherical particles with micro size in diameter, which means large curvatures, hydrophobic surface properties and small surface areas. On the contrary, CNTs are typical nano fibers of tens of nanometers in diameter, exhibiting quite a large surface area. The ball milling pre-treatment on the raw Al powders was aimed to address the above described incompatibilities between CNTs and the Al powders, and thus to promote the uniform dispersion of CNTs on the Al powder surface through subsequent blending. The CNTs with high elastic modulus was difficult to attach to the original Al powder with spherical