Materials and Design 142 (2018) 288–296

Contents lists available at ScienceDirect

Materials and Design

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

Design of an efficient flake powder metallurgy route to fabricate CNT/6061Al composites

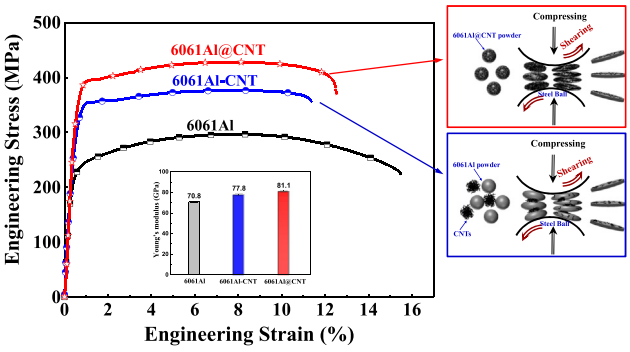
Malin Chen [a], Genlian Fan [a,*], Zhanqiu Tan [a], Dingbang Xiong [a], Qiang Guo [a], Yishi Su [a], Jie Zhang [a], Zhiqiang Li [a,*], Makio Naito [b], Zhang Di [a]

[a] State Key Laboratory of Metal Matrix Composites, School of Material Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China
[b] Joining and Welding Research Institute (JWRI), Osaka University, 11-1 Mihogaoka, Ibaragi, Osaka 567-0047, Japan

HIGHLIGHTS

- CNTs can be de-agglomerated and uniformly coated onto surface of Al powder via the mechanism of dry particle coating;
- High-shear pre-dispersion process promoted the dispersion efficiency, uniformity and structural integrity of CNTs;
- Simultaneous enhancement in tensile strength, Young's modulus and ductility were achieved in the pre-dispersed composite.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 4 September 2017
Received in revised form 22 January 2018
Accepted 22 January 2018
Available online xxxx

Keywords:
Aluminum matrix composites
Flake powder metallurgy
Carbon nanotubes
Pre-dispersion
Dry particle coating
Mechanical properties

ABSTRACT

A flake powder metallurgy route that combines high-shear pre-dispersion process with shift-speed ball milling was designed to fabricate carbon nanotube reinforced aluminum composites. In order to mitigate the dispersion responsibility of ball milling and improve the dispersion efficiency, high-shear pre-dispersion process utilizing the mechanism of dry particle coating was firstly applied to CNTs and 6061Al powder mixtures for a few minutes to de-agglomerate CNTs and uniformly coat them onto surface of 6061Al powder. It was demonstrated that, for the pre-dispersed powders, more rapid and uniform dispersion of CNTs was achieved during following shift-speed ball milling process, which in turn protected CNTs from additional damage and led to better interfacial bonding and higher reinforcing efficiency. As results, compared to counterpart without pre-dispersion, the as-extruded bulk 1.5 wt% CNT/6061Al composites exhibited simultaneous enhancement in Young's modulus, tensile strength and ductility.

1. Introduction

The advanced carbon nanomaterials with extraordinary mechanical and physical properties, represented by carbon nanotubes (CNTs) and graphene nanosheets, are regarded as ideal reinforcements for developing high performance composites [1–9]. During the past two decades, efforts have been made to develop carbon nanotubes reinforced aluminum matrix composites (CNT/Al) [1,10–13], due to the urgent need in transportation and national defense for advanced lightweight structural materials with good stiffness and ductility. However, this kind of composite has not been come into practice yet, owing to the lack of efficient mass production route that can realize satisfying mechanical properties.

* Corresponding authors.
E-mail addresses: fangenlian@sjtu.edu.cn (G. Fan), lizhq@sjtu.edu.cn (Z. Li).

https://doi.org/10.1016/j.matdes.2018.01.044
0264-1275/

Powder metallurgy (PM) via high energy ball milling (HEBM) has become a main approach to prepare CNT/Al composite due to its efficiency and convenience, which resorted to the energetic collision between grinding media during ball milling to disperse CNTs while refining matrix grain. Although the HEBM has been proved effective in breaking the native alumina skin on the Al powder surface and establishing the interfacial bonding between CNTs and Al matrix, the severe impact of the milling balls also caused serious damage to the CNTs structure during the long duration ball milling required to achieve a uniform dispersion of CNTs. To solve the dilemma between the dispersion uniformity, CNT structural integrity and interfacial bonding, many researchers [13–19] investigated the effect of ball milling condition on the structure and properties of CNT/Al composites and tried to optimize the ball milling process with varied milling regimes. Very recently, we proposed a new flake powder metallurgy route via shift-speed ball milling (SSBM) strategy, which allocates the tasks to a long period of low-speed ball milling (LSBM) to obtain Al nanoflakes with uniformly dispersed CNTs firstly and a short period of high-speed ball milling (HSBM) to obtain cold-welded lamellar granulates with good CNT/Al bonding and inter-flake bonding [20]. It is demonstrated that, by combining different powder deformation mechanism under different ball milling speed, the SSBM resulted in comparable strength in 1.5 wt% CNT/Al composites but doubled and tripled ductility as those fabricated by sole LSBM or HSBM. This indicates that the SSBM method could provide quite good coordination between CNT dispersion, structural integrity and interfacial bonding. However, owing to the strong van der waal's force of attraction among CNTs, the starting CNTs are typically in clusters with tens of microns in diameter [21,22]. The common powder mixing method (i.e., 3D mixer) could not tear apart the CNT clusters efficiently. So, it requires a fairly long time to disperse CNTs that rely on LSBM, and some local agglomeration of CNTs may still exist in the final products.

De-agglomerate CNTs before ball milling seems to be a good strategy to relieve the responsibility of ball milling dispersion, which could improve the dispersion efficiency during ball milling as well as reduce the CNTs structural damage. In several works [23–25], CNTs were pre-dispersed in alcohol ultrasonically, and then they were slurry mixed with Al powders before ball milling. Although the improved dispersion was achieved than that of merely ball milling, simple slurry mixing process was usually not sufficient due to the easy re-aggregation of CNTs during drying processes. Some researchers tried to combine several different methods together to disperse CNTs homogeneously and maintain the structural integrity simultaneously. Chen et al. [21] developed an approach with the combination of solution coating, ball milling and Al-flake producing into a simple organic unity, and it was found that much better mechanical properties can be obtained compared to each single method. Yang et al. [26,27] shortened the ball milling time by directly growing CNTs on the Al particle surface and it was proved to be efficient in improving strength and ductility simultaneously. However, whether using solution coating or in-situ growth as a preliminary CNT dispersing method is time and energy consuming, which are hardly compatible with industrial production routes. Therefore, developing an efficient pre-dispersion method is essentially important for producing high performance CNT/Al composites by ball milling.

In this study, a smart and efficient flake powder metallurgy route via the combination of high-shear pre-dispersion process and the shift-speed ball milling was designed to fabricate CNT reinforced 6061Al based composites, in which CNTs were firstly de-agglomerated mechanically and uniformly coated onto surface of spherical 6061Al powder via the mechanism of dry particle coating [28–30]. It was demonstrated that the high-shear pre-dispersion process not only promoted rapid and uniform CNT dispersion during the following SSBM process, but also provided better protection for CNTs from additional damage. As a result, strong and ductile bulk CNT/6061Al composites with CNTs homogenously distributed in aluminum matrix were obtained.

2. Experimental

2.1. Fabrication of CNT/6061Al composite powders

Fig. 1 illustrated the flake powder metallurgy fabrication procedures of the CNT/6061Al composite powders used in this investigation. In a typical experiment, two steps were involved:

(1). **The high-shear pre-dispersion process of CNTs and 6061Al powders**. The near-spherical 6061Al powder (about 30 μm, 99.8% in purity, Bai Nian Yin (Zhejiang) co., Lt., China) and 1.5 wt% multi-walled CNTs (~20 nm in diameter, ~1–2 μm in length, Cnano (Zhenjiang) Technology Ltd., China) were firstly homogenized for 30 min at the speed of 300 rpm and then processed in a high-speed mechanical powder processor (Nobilta, Hosokawa Micron Corporation, Japan) at the speed of 1800 rpm for 15 min. As shown in Fig. 1a, the mixtures were processed in a container in which a rotating four-way blade was designed such that it was almost touching the inner wall of the container with a 3 mm gap [28]. A severe but tailorable shear stress was induced to de-agglomerate the clustered CNTs and coated them onto the surface of 6061Al powder. The high-shear pre-dispersed CNT/6061Al composite powder was denoted as 6061Al@CNT composite powders, hereafter.

(2). **Shift-speed ball milling process of 6061Al@CNT powders**. As shown in Fig. 1b, the pre-dispersed powders were ball milled with 1 wt% stearic acid (used as processing control agent) in a stainless steel jar using a planetary ball mill at speed of 135 rpm (LSBM) for 6 h and at 270 rpm (HSBM) for 1 h at room temperature. The initial ball-to-powder weight ratio was 20:1. Then, the as-prepared composite powders were heated to 653 K at a heating rate of 10 $K \cdot min^{-1}$ and kept in a vacuum for 2 h to remove the stearic acid.

2.2. Consolidation of CNT/6061Al composite powders

The CNT/6061Al composite powders were cold pressed into $\phi 40 \times 50$ mm billets under 500 MPa pressure, and then they were consolidated by vacuum pressureless sintering at 783 K for 2 h and hot extruded into $\phi 8$ mm rods at 693 K with an extrusion ratio of 25:1 at a speed of 12 $mm \cdot min^{-1}$.

For comparison, a CNT/6061Al composite which was only homogenized for 30 min by low speed mixing at the speed of 300 rpm (denoted as 6061Al-CNT composite), and an unreinforced 6061Al specimen were fabricated following the same ball milling and consolidation condition. The density of extrude rods were over 99.5%.

2.3. Material characterization

The distribution of CNTs and microstructure of CNT/6061Al powders was characterized by a field emission gun scanning electron microscope (FE-SEM, SIRION 200) and a high-resolution transmission electron microscopy (HR-TEM, JEOL 2100F). Raman spectroscopy (SENTERRA R200-L) was performed by using the 532 nm line of an Ar^+ laser as the excitation source to analyse the structure of the CNTs. The extent of CNT/6061Al interfacial reaction was quantitatively characterized by using the method of measuring the CH_4 gas contents released by Al_4C_3 hydrolysis [31]. Tensile specimens of 5 mm in diameter and 25 mm gauge length were machined form the extruded rods, and tensile test was carried out in a universal testing machine at an initial strain rate of 5×10^{-4} s^{-1} at ambient temperature (Zwick Z100 100KN, Zwick Roell Group Ltd., Germany), which use a use a mechanical contact extensometer to measure the ductility. For each material, three individual tests were run. The Young's modulus of the composites was measured by a resonance method, in which high frequency vibration with a small load was applied to the sample to measure its resonance