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Grain size effect on the strengthening behavior of aluminum-based composites containing multi-walled carbon nanotubes

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ABSTRACT

Strengthening efficiency of multi-walled carbon nanotubes (MWCNTs) is investigated for aluminum-based composites with grain sizes ranging from ~250 to ~65 nm. The strength of composites is significantly enhanced proportional to an increase of the MWCNT volume. However, the increment differs depending on deformation mode of the matrix. The strengthening efficiency of MWCNTs in ultrafine-grained composites is comparable with that predicted by the discontinuous fiber model, whereas the efficiency becomes half of the theoretical prediction as grain size is reduced below ~70 nm. For nano-grained aluminum, activities of forest dislocations diminish and dislocations emitted from grain boundaries are dynamically annihilated during the recovery process, providing a weak plastic strain field around MWCNTs. The observation may provide a basic understanding of the strengthening behavior of nano-grained metal matrix composites.

1. Introduction

Nano-grained metals exhibit superior strength as predicted by the Hall–Petch relation, although it may not hold for very fine grain sizes [1,2]. However, their limited ductility, stemming from early plastic instability, has been concerned as crucial factors limiting their structural applications [3,4]. Efforts have been made through microstructural controls to achieve ductility at a minimal expense of strength [5]. Composites have also been reported as a breakthrough [6,7,10–15]. By choosing the right combination of ultrafine- or nano-grained metal matrix and reinforcement, a desired combination of strength and ductility can be achieved. For example, the bulk tri-modal Al 5083-based composite consisting of three phases—coarse-grained matrix, ultrafine- or nano-grained matrix, and B_4C reinforcement—has been proven to have superior mechanical properties [6,7].

Carbon nanotubes (CNTs) have been used to overcome engineering limits of materials by taking advantage of their exceptional properties [8,9] and many researchers have endeavored to develop metal/CNT [10–15]. Ball-milling techniques followed by hot-consolidation processes such as spark plasma sintering [10], spark plasma extrusion [11], hot-extrusion [12], and hot-rolling [13] have often been utilized to develop metal matrix composites containing CNTs. Zhong et al. [14] have developed the aluminum matrix composite containing 5 wt% CNTs, which showed around 2 times higher hardness value. Morsi and Esawi' group have conducted a great deal of studies and the aluminum matrix composite reinforced by 2 wt% CNTs, which was fabricated by a powder metallurgy route followed by annealing, shows ~350 MPa in tensile strength and ~8% in tensile elongation to failure [15]. Furthermore, it has been reported that the ultrafine-grained aluminum matrix composite reinforced with multi-walled CNTs (MWCNTs) exhibits a significant improvement in the overall mechanical performances such as tensile stress and ductility; a superior strength of ~600 MPa with considerable ductility of ~2% tensile elongation to failure [13]. However, the role of the matrix (e.g. ultrafine- or nano-grained metal matrix) on the mechanical behavior of the composites has not been investigated as well.

For composites with the ultrafine- or nano-grained metal matrix, various strengthening mechanisms have been typically considered to be involved; direct strengthening by load transfer from the matrix to reinforcement [16–18], indirect strengthening by affecting dislocation activities [19–27], and grain refinement strengthening [28]. To clarify the load transfer behavior, various continuum mechanics models including the shear-lag model [16,17] and the homogenization methods [17,18] have been suggested over the past few decades. Furthermore, indirect strengthening has been also well-established in the literature; reinforcement additionally contributes to the matrix strengthening by restricting activities of dislocations to bypass the reinforcement (that is, Orowan strengthening [19–24]) or inducing geometrically necessary dislocations from thermal mismatch between the matrix and reinforcement (that is, thermal mismatch strengthening [25–28]). However, these models are valid only when the matrix deforms in the conventional way

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as demonstrated for coarse-grained metals. As grains are refined below the critical size, activities of lattice dislocations become less significant, leading to specific deformation mechanisms [1,2,29–34]. The unique deformation mechanism of the nano-grained matrix might provide an irregular stress distribution around reinforcement and affects strengthening behavior of reinforcement.

The objective of this study is to investigate the effect of a deformation mode on strengthening efficiency of reinforcement. Aluminum and MWCNTs were selected as the matrix and reinforcement. The composite was fabricated by varying grain sizes (i.e. 250, 150, 73, and 65 nm) with regarding the transition in deformation mechanism of aluminum arising at a grain size of ~70 nm [35]. Deformation behavior of the composites with various grain sizes was investigated in the MWCNT volume fraction up to 9 vol%. The experimental data were compared to the theoretical models.

2. Materials and methods

Aluminum and Al/MWCNT composite sheets were fabricated by hot-rolling the ball-milled powder, as described in the previous report [13]. An attrition mill was utilized to disperse MWCNTs (~20 nm in diameter, ~5 μm in length, and supplied from Applied Carbon Nano Co., Ltd.) in aluminum powder (99.5% in purity and ~150 μm in size). Milling was conducted at 500 rpm in a purified argon atmosphere at room temperature. The ball-milled powder was packed in a copper container, degassed for 3 h at room temperature, sealed by welding and then hot-rolled with every 12% reduction; the thickness was reduced to be ~1.5 mm.

Four fabrication routes were employed for a variation of grain sizes. The first group (group A) represents nano-grained (the smallest grain size in this study) composites; aluminum powder was solely ball-milled for 18 h to reduce the grain size to the nano regime, and then a mixture of the ball-milled aluminum powder and MWCNTs was further ball-milled for 6 h, followed by hot-rolling. The second group (group B) also has nano-sized grains; aluminum powder was solely ball-milled for 12 h, and then a mixture of the ball-milled aluminum powder and MWCNTs was further ball-milled for 6 h, followed by hot-rolling. The third group (group C), with an intermediate grain size, was ball-milled for 6 h and then hot-rolled at 480 °C. The last group (group D), with the largest grain size, was fabricated at a higher rolling temperature than the third group (group C); the mixture was ball-milled for 6 h and hot-rolled at 530 °C. Finally, diverse specimens were fabricated by varying the volume fraction of MWCNTs (i.e. 0, 1.5, 3.0, 4.5, 6.0, 7.5, and 9.0 vol%) and grain size of the matrix.

The grain size of the specimens was analyzed using X-ray diffraction (XRD, Rigaku, CN2301) with a Cu Kα radiation source through the Scherrer formula [36]. For the specimens with grains larger than 100 nm, grain size was estimated using a transmission electron microscopy (TEM). Nanotube structure evolution upon milling was studied with Raman spectroscopy using a Jobin–Yvon microspectrometer (LabRam HR, Jobin–Yvon Co. Ltd., France). Density of the sheets was estimated using a gas-pycnometer (Ultrapycnometer 1000, Quantachrome Co. Ltd., USA).

Quasi-static uniaxial tensile tests were conducted at a constant crosshead speed with an initial strain rate of 1×10^{-4}/s at room temperature according to ASTM B-209. Tensile specimens (thickness: 1 mm, gauge length: 12.5 mm, gauge width: 6 mm, and effective grip length: 160 mm for both upper and lower grips) were prepared from the hot-rolled sheets and loaded in the rolling direction. The strain was measured using a clip-on extensometer. Before tests, the specimens were heat-treated at 250 °C for 2 h to remove the residual stress. Microstructural variation after deformation was observed using high-resolution TEM (HRTEM, JEOL 2000).

3. Results and discussion

3.1. Characteristics of the composite

Fig. 1a shows the XRD spectra for the powders being milled for 18 and 24 h and their composite sheets. Any notable peak that indicates the presence of carbides is not observed in the spectra. For the specimens in groups C and D, grain size was statistically measured using TEM analysis because the grain size is larger than 100 nm. Average grain sizes of specimens as a function of milling time are plotted in Fig. 1. The grain size of ball-milled powder rapidly decreases with increasing milling time of up to 12 h; powders ball-milled for 6 and 18 h exhibit ~135 and ~70 nm in grain size, respectively. However, this trend slowed down in further milling to reach an apparent steady-state; powder ball-milled for 24 h exhibits ~50 nm in grain size. To avoid the grain growth at high temperatures, rolling was conducted at a relatively low temperature of 480 °C for less than 1 h. Although the grain growth is found to be minimal after rolling at 480 °C, grains grow somewhat after rolling at 530 °C as plotted in Fig. 1. Average grain sizes of the specimens in groups A–D are estimated to be ~250, ~150, ~73, and ~65 nm, respectively. On the other hand, the effect of MWCNTs on grain refinement process is found to be negligible, contrary to the previous reports [37,38]. At the early stage of milling, CNTs could stimulate the grain refinement process by generating forest dislocations within grains [38]. As grain size is reduced to hundreds of nanometers, however, dislocation multiplications are rarely observed within grains, and the role of CNTs in grain refinement loses its significance.

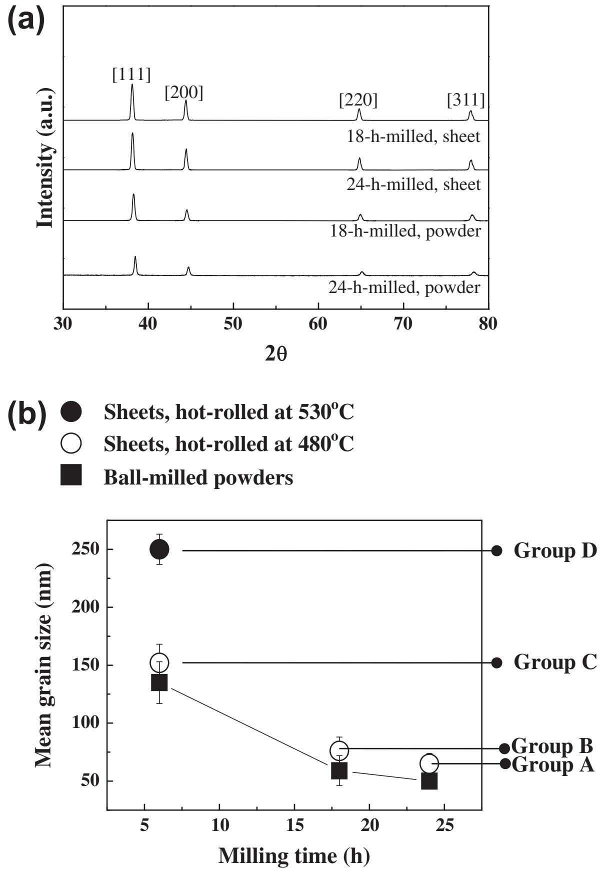

Fig. 1. Grain size of ball-milled powders (solid rectangle) and composite sheets hot-rolled at 480 °C (open circle) and 530 °C (solid circle) as a function of milling time.