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Fabrication, characterization and mechanical properties of hybrid composites of copper using the nanoparticulates of SiC and carbon nanotubes

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ABSTRACT

Copper based hybrid composites containing nano-sized silicon carbide and carbon nanotubes reinforcements with minimal porosity were fabricated via mechanical milling followed by hot pressing technique. Microstructures of the powders and consolidated materials were studied using scanning electron microscope, X-ray diffraction, Raman spectroscopy, and scanning transmission electron microscope. Microstructural characterization of the materials revealed that the addition of nanosized silicon carbide reinforcement lowered the grain growth rate and enhanced the homogenization during mechanical milling. Microhardness measurements and compression test showed considerable improvements in mechanical properties of the composites due to the addition of nanoparticulates and the grain refinement. The strength of the composite materials was discussed using theoretical models of the Hall-Petch, Orowan, and thermal mismatch mechanisms to determine the contribution of each mechanism in total strength.

1. Introduction

Carbon nanotubes have created tremendous expectations as strengthening additives for metallic, ceramic and polymer composites due to their high strength and stiffness [1–2]. Carbon nanotubes are reported to have an elastic modulus comparable to that of diamond (1.2 TPa) and a strength 10–100 times that of high strength steel [3]. The high strength is a result of the near perfect structure and the strong sp2 bonding between the C–C bonds. Since the last decade, a number of investigations have been focused on incorporating CNTs in polymer matrices [4,5], ceramics [6–8], and metals [9,10]. Research on metal matrix composites has been increasing after the first article appeared in 1998 on Al/CNT composite [11]. Using CNT in MMC manufacturing is very promising because this could be the way for producing lightweight, ultra high strength, and stiff products made out of metal matrix-nanotubes. In recent years, a lot of papers on the mechanical properties of bulk metal matrix/CNT composites have been reported. However, the improvement in the mechanical properties of metal matrix/CNT composites is not commensurate with the extraordinary high strength of carbon nanotubes. The degradation of mechanical properties in metal matrix/CNT composites is severe at higher CNT content (> 2 vol%). Carreno-Morelli et al. [12] emphasized that there are two major problems that face scientists and researchers in manufacturing CNT reinforced composites which are (i) achieving a homogeneous and uniform dispersion of CNT in the matrix and (ii) forming a strong bond at the CNT-metal interface. A lot of papers have been published related to these problems. Reports on Cu-CNT systems deal with improvements in mechanical properties. Dong et al. [13] prepared Cu/CNT composites by simple blending and sintering method. They reported approximately 20% increase of hardness by 15 vol% CNT addition. Spark plasma sintering of Cu-10 vol% CNT composites improved the hardness by 79% with a further improvement up to 207% resulted from rolling of the SPS composite [14]. This improvement is the result of homogenous dispersion of CNTs. Chu et al. [15] used particles compositing system (PCS) for blending the powders and consolidated the blended powders by SPS to produce Cu/CNT composites. They could achieve the homogeneous dispersion of CNTs in the copper matrix by the CNT volume fraction of 10%. At higher volume fraction of CNT, they showed CNTs clustering in the matrix. Some researchers also used molecular level mixing [16] to prepare composite powders with better dispersion of CNTs. Coating of CNT reinforcement by Ni improved bonding with the Cu matrix, resulting in approximately 80–100% increase in the hardness by 9–12 vol% CNT addition [17]. Also, Chu et al. recently prepared high strength Cu–Cr/CNT composites by introducing Cr as an alloying element to improve the Cu-CNT interface strength via ball milling [18]. They ascribed the high strength to the formation of thin intermediate Cr3C2 transition layer between

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the CNTs and Cu–Cr matrix. Beside different methods used for dispersing CNTs in the Cu matrix and strengthening the interface bonding, these issues are subject of many researches to reach the high strength Cu/CNT composites. Recently, Kwon et al. [19] have prepared new Al/(SiC+CNT) hybrid composites via the mechanical milling and hot pressing method. They showed that using small quantity of SiC nanoparticles leads to better dispersion of the highly agglomerated CNTs into the Al matrix. Also, the Vickers hardness of the obtained composites was reported up to eight times higher than that of pure Al bulk samples.

In most of the published papers, mechanical milling has been used to manufacture homogenously dispersed CNT strengthened alloys [20]. During mechanical milling, the homogenization and grain refinement happen simultaneously. Although long time mechanical milling results in high degree of grain refinement and homogenization, it introduces many defects on CNTs structure [21]. Since the structural and chemical stability of CNTs in the matrix is an important factor determining strengthening achieved by CNT reinforcement and improvement in composite physical properties, decreasing the mechanical milling time and its energy is important.

On the other hand, many experimental data show that the structural stability of the fine grained materials is poor [22]. Ultrafine-grained Cu is usually unstable when exposed to moderate temperatures as a result of recovery processes and grain growth. For instance, the 50% recrystallization temperature in 98% cold-rolled 99.999% pure Cu is as low as 360 K [23]. Particle strengthening is one of the methods to increase thermal stability [22]. When small amount of second phase is added to Cu/CNT, it can promote grain boundary pinning during consolidation stage. It has been shown that stability of microstructure, namely, invariability of the mean and maximum grain sizes, can be achieved under the influence of fine and immobile disperse particles [24].

In this paper, (i) the possibility of production of a new Cu/(SiC+CNT) hybrid composites by introducing small amount of SiC nanoparticles as an active mixing agent for dispersing the CNTs in the Cu powder during a short mechanical milling time using a new approach proposed by Kwon et al. [19], and (ii) the efficiency of SiC nanoparticles for retarding grains growth during hot condensation are investigated. Cu based hybrid composites containing nano-sized silicon carbide and CNT reinforcements were fabricated via mechanical milling and hot pressing method. CNTs dispersion and grain refinement and CNTs structural change during mechanical milling and hot pressing were evaluated. Mechanical properties of the composite materials were evaluated by compressive and micro-hardness tests. The mechanical properties of the composites are discussed in accordance with grain size and homogeneity of the microstructure. Finally Hall-Petch, Orowan and thermal mismatch theoretical models are used to estimate the strength of the composite materials.

2. Experimental

Pure copper powder (99.7% purity and $<20\,\mu m$ size range) obtained from Merck, Germany, Silicon carbide powder (Fig. 1a) (supplied by Nabond Co., Shenzhen, China, with purity of +99%) as one of the reinforcements with average particle size of 40 nm and multi-walled carbon nanotubes (Fig. 1b) with greater than 95% purity and with average outer diameter of 60 nm and length of 3–4 μm (Supplied from Nanotech Port Co., Shenzhen, China) were used as raw materials. Cu powder and the nanoparticulate materials (CNT and SiC) were mixed and mechanically ball milled in a planetary ball mill for 3 h under an argon atmosphere at 200 rpm, using; 10 mm balls, a 10:1 ball to powder weight ratio and 0.5 wt% of stearic acid (Merck, Germany) as a process control agent (PCA). At the end of the process, the tight bowl containing the powder blend was transferred to a glove-box with a controlled inert atmosphere of argon. The mechanically alloyed powders were consolidated by a hot pressing (HP) method using a steel die with the sample size of $6\,mm \times 6\,mm \times 30\,mm$ at a load of 150 MPa, temperature of 973 K and the soaking time of 30 min. The density of the composites was measured by the Archimedes' method.

The specimen surfaces were polished mechanically with emery papers down to 1200 grade, and then with 0.05 μm wet polishing diamond pastes. The microstructure of the composites was observed by scanning electron microscopy (VEGA-TESCAN-XMU) and scanning transmission electron microscope (Cs-corrected STEM (JEM-2100F) operated at 200 kV).

The XRD patterns were measured using a D8 Advance Bruker diffractometer with a Cu Ka radiation ($\lambda=0.154$ nm) at 40 kV and 50 mA by the step size of 0.02°.

The change of MWCNTs structure after mechanical milling and hot consolidation were characterized by a Laser Confocal Raman Microscopy (SENTERRA-2009, BRUKER, Germany), using a laser excited at 785 nm.

The Vickers micro-hardness test of the composites was carried out with a load of 50 g for 10 s (Olympus micro-hardness tester: FM-700). At least 20 measurements were made per sample.

Compressive tests were performed using an Instron 5583 apparatus under a strain rate of $2 \times 10^{-4}\,s^{-1}$ using cylindrical disk-shaped specimens 7.5 mm thick and 5 mm in diameter. For each material at least 3 compression tests were taken and averaged. During the compression tests, precise strains were measured by an optical strain gauge system (ARAMIS 5M).

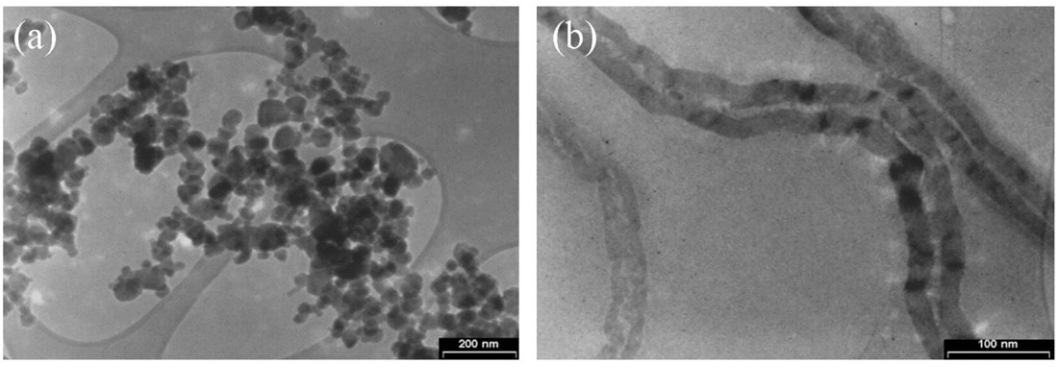

Fig. 1. TEM micrograph of used materials: (a) SiC nanoparticles and (b) MWCNTs.