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# Interface tailoring to enhance mechanical properties of carbon nanotube reinforced magnesium composites

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ABSTRACT

This study highlights the use of a metallic coating of nanoscale thickness on carbon nanotube to enhance the interfacial characteristics in carbon nanotube reinforced magnesium (Mg) composites. Comparisons between two reinforcements were targeted: (a) pristine carbon nanotubes (CNTs) and (b) nickel-coated carbon nanotubes (Ni–CNTs). It is demonstrated that clustering adversely affects the bonding of pristine CNTs with Mg particles. However, the presence of nickel coating on the CNT results in the formation of $Mg_2Ni$ intermetallics at the interface which improved the adhesion between Mg/Ni–CNT particulates. The presence of grain size refinement and improved dispersion of the Ni–CNT reinforcements in the Mg matrix were also observed. These result in simultaneous enhancements of the micro-hardness, ultimate tensile strength and 0.2% yield strength by 41%, 39% and 64% respectively for the Mg/Ni–CNT composites in comparison with that of the monolithic Mg.



## 1. Introduction

Magnesium is the lightest of all structural metals. It's low density (~1.74 g/cm$^3$) coupled with it's high specific mechanical properties make magnesium-based materials very attractive for weight-critical applications. In recent years, the urgency to cut down energy consumption and greenhouse gas emissions result in a surging interest in energy-efficient materials for automotive and aerospace applications. In view of this, the present study aims to develop new magnesium-based composites with enhanced mechanical performance, to address the ever-increasing demand.

Carbon nanotubes (CNTs) are known for their extraordinary structural, mechanical and electrical performances. These excellent properties are derived from the unique quasi-one dimensional nature and the cylindrical symmetry of nanotubes [1]. The exceptional mechanical properties of CNTs make them good candidate as reinforcement to strengthen metals. However the major challenges in the synthesis of CNT-reinforced metal matrix composites (MMCs) include: (i) difficulty in incorporating and distributing the CNTs uniformly in the metal matrix, and (ii) insufficient wetting between the CNT and the metal matrices.

The interface between the matrix and reinforcement plays a critical role in determining the overall properties of metal–matrix composites. Stiffening and strengthening rely on the load transfer across the interface. Toughness is influenced by crack deflection at the interface and ductility is affected by relaxation of peak stresses near the interface [2]. In a study conducted by Chu et al. [3], it has been shown that it is possible to modify the microstructure and improve the bonding at the interface by matrix-alloying chromium element into CNT/copper composites. Thus, by improving the interfacial adhesion between CNT reinforcements and Mg matrix material, the full potential of CNTs in forming mechanically reinforced metal matrix composites (MMCs) could be realized. While extensive research has been directed towards the development of surface treatment techniques for carbon fibers to improve the fiber–matrix interface bonding [4,5], the use of metallic coatings on nanoscale reinforcement such as CNTs has not been studied in detail. Theoretical studies based on wetting and nucleation theories have shown that low metal-CNT interfacial energies and high diffusion barriers allow the formation of continuous or quasicontinuous nickel (Ni) layers on the CNT surface [6]. This has been proven experimentally, where uniform Ni coatings were formed on CNT surfaces with strong covalent bonding characteristics [7]. In another study, Menon et al. [8] attributed the strong Ni–CNT interaction to curvature-induced rehybridization of carbon $sp^2$ orbitals with the Ni d-orbital. Ni particulates (14 wt.%) addition to Mg has also been demonstrated by Hassan and Gupta [9] to significantly enhance the tensile strength by 80% and 0.2% yield strength by 320%, through the formation of strong interfacial bonding. These previous studies have shown that (i) Ni forms strong covalent bonds with CNT and (ii) Ni addition to Mg enhances its mechanical

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properties. In view of this, it is hypothesized that a tailored functional coating such as Ni with nanometer thickness could result in a strengthened interface while avoiding the formation of excessive brittle intermetallics which might degrade the composite's failure strain.

Accordingly, in this study, Mg composites were fabricated using pure magnesium as the matrix material with bare multi-walled CNT (0.3 wt.%) and Ni-coated multi-walled CNT (0.3 wt.%) as the reinforcement materials. The composites were synthesized using the powder metallurgy route incorporating microwave assisted rapid sintering technique, followed by hot extrusion. The suitability of the Ni coating was also verified through tests which assessed the composites' mechanical properties and analyzed the microstructure of the fiber–matrix interface.

## 2. Experimental procedures

In this study, magnesium powder of 98.5% purity with a size range of 60–300 μm (supplied by Merck, Germany) was used as the matrix material. Pristine multi-walled carbon nanotubes produced by chemical vapor deposition (supplied by Tsinghua University, China) (Fig. 1a) and nickel coated multi-walled carbon nanotubes (Ni–CNTs) (supplied by Chengdu Organic Chemicals Co., Ltd.) (Fig. 1b) were used as the reinforcements in Mg. Both types of CNT have typical diameters of 10–20 nm.

Magnesium was reinforced with 0.3 wt.% of pristine CNTs and 0.3 wt.% Ni–CNTs to form Mg/CNT and Mg/Ni–CNT composites respectively, using the powder metallurgy technique. Pure magnesium powder was blended with the appropriate amount of reinforcements in a RETSCH PM-400 mechanical alloying machine at 200 rpm for 1 h. The homogenized powder mixtures of Mg and reinforcement were then cold compacted at a pressure of 713 MPa to form billets of 40 mm in height and 35 mm in diameter. Monolithic magnesium was compacted using the same parameters without blending. The compacted billets were sintered using an innovative hybrid microwave sintering technique [10,11]. The billets were firstly heated to 640 °C in a 900 W, 2.45 GHz SHARP microwave oven. The sintered billets were then soaked at 400 °C for 1 h and subsequently hot extruded at 350 °C using an extrusion ratio of 25:1. Microwave sintering used in this study, reduced processing time (up to ~90%) and energy savings (up to ~97%) in comparison to conventional sintering techniques. This environmentally friendly and economic heating approach had been adopted in earlier studies to successfully synthesize dense Mg composites with improved mechanical properties [10,11].

Density measurements were performed using the Archimedes' principle on polished samples from the extruded Mg and Mg composites rods. Porosity levels were obtained by area fraction analysis using Scion image analysis software. X-ray diffraction (XRD) analysis was carried out on the Mg and Mg composites samples using an automated diffractometer (SHIMADZU XRD-6000). The samples were exposed to Cu Kα radiation (λ = 1.54056 Å) at a scanning speed of 2 deg $min^{-1}$. The interplanar spacings ($d$) obtained were subsequently matched with the standard values for magnesium, nickel, carbon, their oxide and intermetallic phases using the JCPDS database [12]. Micro-hardness measurements were made on the polished samples of extruded Mg and Mg composites rods in accordance with the ASTM: E384-11e1 using an automatic digital micro-hardness tester (Matsuzawa MXT 50) with a Vickers indenter under a test load of 25 gf and a dwell time of 15 s. The tensile properties of Mg and Mg composites were determined based on ASTM: E8/E8M-13a. The tensile tests were conducted on 5 round tension test specimens of 5 mm in diameter and 25 mm gauge length using an automated servo hydraulic testing machine (MTS 810) with a crosshead speed set at 0.254 mm/min. Microstructural characterization studies were also conducted with the aim of determining: (i) presence of porosity, (ii) grain size, (iii) grain morphology, (iv) distribution of reinforcement and (v) presence of intermetallics. Transmission Electron Microscope (JEOL, TEM 2010F) equipped with Energy Dispersive X-ray Spectroscopy (EDX), Field Emission Scanning Electron Microscope (HITACHI FE-4300) and metallographic optical microscope (OLYMPUS) were used for the microstructural and fracture analysis.

## 3. Results and discussion

### 3.1. Density and porosity measurements

Table 1 shows the density and porosity results of Mg and Mg composites. The densities of the composites were comparable to that of pure magnesium. This showed that although the density of Ni (8.9 g/$cm^3$) is much larger than that of Mg (1.74 g/$cm^3$), the presence of a thin, nanoscale coating of Ni on the CNT surface did not affect the material's overall density. The incorporation of Ni–CNTs and CNTs into the Mg matrix did not contribute to any significant change in the density value. This property is desirable for lightweight applications of Mg composites. The porosity in both composites were higher than that of pure Mg. Mg/0.3 wt.% CNT composites showed higher porosity (0.75 vol%) compared to Mg/0.3 wt.% Ni–CNT composites (0.46 vol%).

### 3.2. Microstructures

Measurements from the etched samples showed that the composites exhibited a relatively finer grain size with lower aspect ratio and roundness when compared to that of the monolithic Mg samples (Table 1). These can be attributed primarily to the coupled effects of: (i) the capability of CNT reinforcements to nucleate

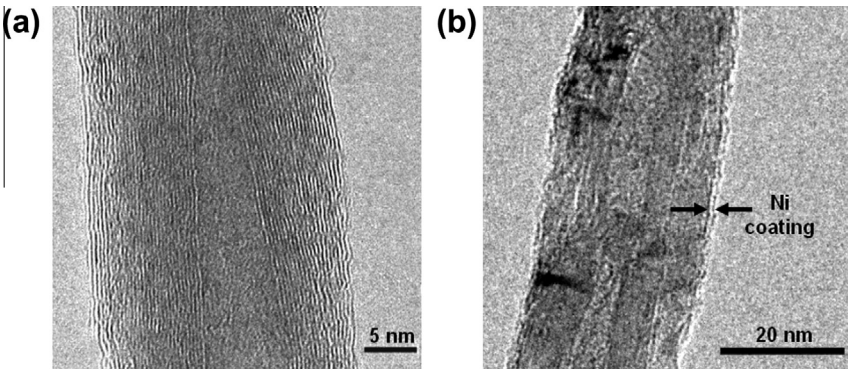


**Fig. 1.** TEM micrographs showing (a) pristine multi-walled carbon nanotubes and (b) Ni-coated multi-walled carbon nanotubes used in this study. The arrows indicate the Ni coating on the CNT.