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Low Temperature Synthesis of Coiled Carbon Nanotubes and Their Magnetic Properties

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Abstract. In this paper, coiled like structure of carbon nanotubes (c-CNTs) have been effectively grown on bi-metal substituted α-alumina nanoparticles catalyst by chemical vapor deposition (CVD) system. Highly graphitized and dense bundles of carbon product were attained at a low temperature of 550 °C. The coiled carbon nanostructures in very longer lengths were noticed by field emission scanning electron microscope (FESEM) observation. Furthermore, high purity material was achieved, which correlates the energy dispersive x-ray spectroscopy (EDX) analysis. High resolution transmission electron microscope (HRTEM) revealed the diameter and graphitization of coiled structures. The superparamagnetic like behavior was observed at room temperature for the as-synthesized product, which was found by VSM investigation.

INTRODUCTION

Carbon nanotubes (CNTs) are well recognized for their greater properties could benefit prospective applications in the field of science, engineering and technology after their discovery by Sumio Lijima through transmission electron microscopy observations [1]. Tremendous development has been attained during the past 20 years on the yield, nucleation, growth processes and numerous applications in diverse fields. They have outstanding mechanical strength, thermal stability, efficiency in heat conduction, high chemical resistivity, electrical properties and have subsequently been utilized in a wide range of applications [2]. CNTs behaviour or properties powerfully depends on their internal structures because of that shortly, diverse shapes for CNTs have been reported [3,4]. Research has been carried out to build up novel designs for CNTs to facilitate a multiplicity of applications. Among them, coiled carbon nanotubes (c-CNTs) have paying attention due to their unique electrical, magnetic, and mechanical properties [5]. These types of CNTs are created by the construction of paired pentagon and heptagon carbon rings and arrange themselves regularly inside the CNT network [6].

They have promising probable applications include electromagnetic absorbers, electromagnetic nanotransformers, nanocircuitry, nanoactuators, nanoscale mechanical springs, lithium battery electrodes, field emission devices [7,8]. The prominent applications are due to as they have extraordinary properties such as high electrical conductivity, high surface area, high charge transport capability, high mesoporosity, high electrochemical stability, and high electrolyte accessibility [9]. They have the advantages over their counterparts due to the combination of their unusual morphology and coil shape effects in elastic properties as well as the ability to undergo reversible deformation [10]. Lau *et al.* proved that a 19% enhancement in stiffness achieved with addition of 2 wt% straight single-walled CNTs into an epoxy resin while the addition of 2 wt% c-CNTs produced in a 54% enhancement [11]. Recently, Changsoon *et al.* reported weavable, stretchable solid state super capacitors based on coiled carbon nanotubes polymer and showed higher areal energy density on areal power densities [12]. It was suggested, attractively, that a coil could communicate to a series of joints through discontinuous metallic and semiconducting behavior, adding to the possibilities of detecting exciting electronic behavior in nonlinear nanostructures [13,14]. Additionally, Akagi *et al.* designed that when a quick density-of-state peak takes place at the

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Fermi level, c-CNTs can also perform as superconductors. These properties influence the make use of c-CNTs in nano-electronic devices and nano electromechanical systems [15].

The large-scale production of c-CNTs is extremely advantageous due to their fascinating electrical and mechanical properties and many methods have been described to produce c-CNTs [16,17]. Among those approaches, chemical vapor deposition (CVD) method is most attractable for variety of applications and allows pre-defined morphology, alignment, simplicity of growth, cost effectiveness and high yields by adjusting reaction parameters [18]. Hekamt *et al.* reported that coiled carbon nanotubes were directly grown on an anodized aluminium oxide template by using CVD method and obtained material show a high specific capacitance (202 $F.g^{-1}$ at a current density of 10 $A.g^{-1}$) as well as excellent cycle stability even after 6000 cycles [19]. In this CVD technique, catalyst plays a key role in achieving desired CNTs materials. A variety of metal catalysts such as noble metal (Rh, Ru, Pt, and Pd) and non-noble metal (Fe, Co, and Ni) catalysts and others like Cr, Cu, Al, In, Ti, Cs, W, Mo, Mn, Ag, Au have been investigated by many researchers [20,21]. Other hand bimetallic catalyst has been used to grow CNTs at low temperature as it decreases the melting point temperature by forming an alloy. In order to reduce the reaction temperature, Cu has been added to Ni metal in a suitable method to fabricate good quality of c-CNTs is hereby reported.

EXPERIMENTAL

Synthesis of c-CNTs

In order to achieve coiled CNTs, 5,10 wt% Ni-Cu/α-Al_2O_3 nanoparticles was used as a catalytic material. The typical catalyst was prepared by a simple wet impregnation method at room temperature, which was reported in our previous investigation [22]. The c-CNTs synthesis process was carried out by CVD technique. The CVD have a horizontal alumina tube reactor at atmospheric pressure, temperature and gas flow controllers. 100 mg of catalyst material evenly dispersed on a quartz boat, which was then located at the middle of the reactor tube. The reaction temperature was set to 550 °C and heated the catalyst slowly under carrier gas of N_2 flow. Precursor gas of acetylene was purged a flow rate of 50 sccm when the reactor temperature reaches 550 °C for 10 min. Acetylene flow was stopped at the end and N_2 gas flow was endlessly purged to CVD chamber till to the room temperature. The collected black sample was characterized by FESEM, EDX, HRTEM and VSM techniques.

Physical Characterization

Structure of the as-synthesized product was analyzed by field emission scanning electron microscope (FESEM) SU-6600, Hitachi, Japan, operated at an accelerating voltage of 15 kV. Energy dispersive x-ray spectroscopy (EDX), INCA PentaFET-x3 (Oxford Instruments, UK) was used to study the elemental composition and purity of the material. High resolution transmission electron microscope (HRTEM) images were recorded on a TecnaiT20 G2 200 kV, FEI Brand (Netherlands) microscope duly operating at an accelerating voltage of 200 kV. The magnetic properties of the sample are recorded on a vibrating sample magnetometer (VSM 7404 series of LAKESHORE make) with vibrating frequency 8.3 Hz in the magnetic field range -15 kO_e to +15 kO_e at room temperature.

RESULTS AND DISCUSSION

Characterization of c-CNTs

FESEM images of the carbon material synthesized at 550 ° were depicted in Figure1. It clearly displays coiled structure of stretched carbon material is found and confirms the formation of c-CNTs. The product exhibits dense bundles of c-CNTs on alumina nanoparticles, which were formed as a nano ball. Formation of c-CNTs was occurred in the pores of alumina nano structure, where the active sites are presented and also shown in Figure 1(a). Measuring the length of the synthesized material is difficult due to twisting morphology, but the average length is able to be predicted to be more than numerous tens of micrometers. From all the FESEM images, diameter of the c-CNTs estimated to be in the range of 30 – 50 nm and further shows uniform coiled structure of the material. It was also observed that the smooth surface of c-CNTs, even though very few number of the catalyst particles adhered on them.

EDX analysis was performed to estimate the purity of the product by calculate approximately the elemental components of the material. Figure 2 demonstrates the EDX spectrum of the coiled carbon product synthesized at

550 °C. The atomic percentage of C, Al, O, Ni and Cu were given in the spectrum itself as a table and atomic percentage are diminished significantly contrast to that of 5, 10 wt% Ni-Cu/α-Al_2O_3 catalyst. The additional peak for gold (Au) is also found in the spectrum, which comes from the Au sputtering while EDX sample preparation procedure. Furthermore, it is worthy to note that the CVD procedure is very effective in contamination free material synthesis.

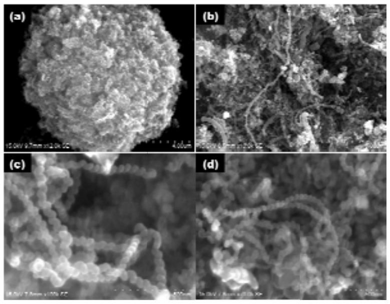

FIGURE 1. FESEM images (a), (b), (c) & (d) of carbon product grown on 5, 10 wt% Ni-Cu/α-Al_2O_3 catalyst at 550 °C.

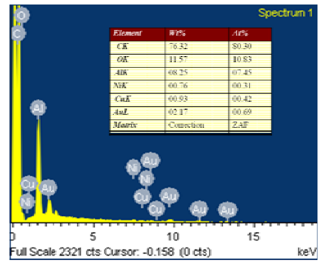

FIGURE 2. EDX spectrum of as grown c-CNTs at 550 °C with atomic percentages.

HRTEM images in Figure 3, clearly shows the formation CNTs with coiled structure from 5,10 wt% Ni-Cu/α-Al_2O_3 catalyst. In all the c-CNTs small hollow core arrangement is not able to be seen due to twisted morphology and number of concentric walls around the central axis are more. It is believed that coil like spring carbon pattern involves the alteration of a transition metal catalyst in order to support non-uniform extrusion of carbon from the active catalyst surface [23]. From the Figure 3, the average diameter of the individual tubes is calculated to be about 40 nm and uniform. It suggested that the Ni-Cu bimetal contribution is very effective in alumina nanoparticles for the growth of c-CNTs at low temperature and ultimately leads to get high quality of the product. We also note that the c-CNTs formation have not been obtained less than the temperature of 550 °C. Thus the temperature also plays a prominent role in achieving in the formation of the coiled structure as well.

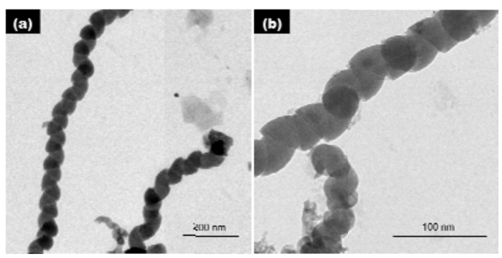

FIGURE 3. HRTEM images (a) & (b) c-CNTs with different magnification.

Magnetic properties of c-CNTs

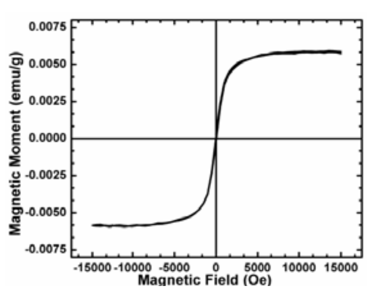

FIGURE 4. VSM plot of c-CNTs obtained at room temperature.

VSM technique was used to evaluate the magnetic properties of produced c-CNTs material at room temperature. Figure 4, shows the VSM plot of magnetic moment versus magnetic field in the range from -15 kO_e to +15 kO_e of the sample at room temperature. The plot displays the measured magnetization curve has no hysteresis loop. It obviously indicates the superparamagnetism nature of c-CNTs at room temperature. The plot also reveals the comparatively low filed saturation magnetization in c-CNTs. The relatively low values of the observed coercivity (H_c) 25 O_e, retentivity (M_r) 0.00014 emu/g and saturation magnetization (M_s) 0.0059 emu/g are exposed the superparamagnetism nature of c-CNTs. This prominent magnetic property is due to the induced magnetism in c-CNTs, which is based on defective sites such as vacancies, structural defects by its distinctive coiled structure and also might be from catalyst impurities. Thus, c-CNTs show superparamagnetism at room temperature which is beneficial for biomedical applications like cell labeling, MRI cell tracking and magnetic manipulationsand also microwave applications.

CONCLUSION

We have successfully established synthesis of coil like structure of CNTs over bimetal impregnated α-alumina nanoparticles catalyst by chemical vapor deposition (CVD) method. The obtained product was characterized by FESEM, EDX, and HRTEM analysis. The result illustrates that the product has highly coiled carbon nanostructures in very longer lengths with good purity. Moreover, the resulted c-CNTs showed soft ferromagnetism at room temperature which is advantageous in AC electrical applications. The magnetic properties of the sample were studied by VSM technique, which reveals the induced superparamagnetism in its structure.

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