

Affecting the morphology of silver deposition on carbon nanotube surface: From nanoparticles to dendritic (tree-like) nanostructures



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ABSTRACT

Chemical reduction was used to synthesize silver crystals on the surface of multiwall carbon nanotubes (MWCNTs) in the presence of acetone, N,N-dimethylformamide (DMF), N-methyl-2-pyrrolidone, and isopropyl alcohol as solvent. DMF and sodium dodecyl sulfate were used as a reducing and a stabilizing agent, respectively. The structure and nature of hybrid MWCNT/silver were characterized by Raman spectroscopy, FTIR spectroscopy, transmission electron microscopy (TEM), and field emission scanning electron microscope (FESEM). The presence of silver crystals on the nanotubes was confirmed by XRD. The results show the formation of silver crystals on the MWCNT surface and indicate that the morphology of silver crystals can be control by changing the solvent. The type of solvent is an effective parameter that affects the particle size and morphological transition from nanoparticles to silver trees.

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1. Introduction

Since the discovery of carbon nanotubes (CNTs) in 1991 by Iijima [1], fundamental research on CNTs and their applications, such as nanoelectronic and photovoltaic devices [2,3], biosensors [4], superconductors [5], electromechanical actuators [6], energy conversion/fuel storage [7], and nanotransistors and composite materials [8,9], has made rapid progress [10]. Unlike carbon materials such as graphite and diamonds, this one dimensional type of carbon allotrope is of interest because of its high aspect ratio, ultra-light weight, high tensile strength, chemical stability, and excellent mechanical, electrical, thermal, and magnetic properties [11].

Nanomaterials, especially metal nanoparticles (NPs) are an emerging class of compounds with unique electronic, magnetic, and catalytic properties [12]. They show completely different properties as compared to their bulk counterparts; although they are composed of the same materials, their different shapes and sizes result in different characteristics. Most research has been focused on NP properties and their applications. The combination of CNTs and NPs has created new hybrid nanostructures that display the properties of nanosized composites and important features for catalysis, energy storage and nanotechnology [13].

The structure and morphology of CNT enable it to serve as a specific template for the deposition of metal NPs or semiconductor NPs [14]. A number of studies have investigated fabrication of metal NPs by decorating CNTs and measuring their unique electrical, magnetic and optical

properties [15–22]. The CNTs appear to provide a suitable substrate on which to deposit silver-NPs, and silver-decorated CNTs have potential applications as advanced nanosized composites, antibacterial agents, catalysts and sensors [23–26].

Several methods have been introduced to prepare silver-CNTs, including vapor deposition [27], thermal decomposition [18,28], surface chemical reduction [29] and gamma-irradiation [30]. Meanwhile, Sahoo et al. used the metallization using an e-beam deposition system, and changed the voltage to grow silver crystal trees on the surface of CNTs [31].

Dispersing CNTs in a solvent is a common step to achieving uniform decoration of CNTs with silver-NPs [32]. CNT suspensions are either macrodispersions or nanodispersions. In macrodispersions, the CNTs disperse in the form of bundles in the solvent whereas in nanodispersions they suspend individually [33].

The strong hydrophobic nature and the high van der Waals force between CNTs result in poor dispersion. Several methods have been suggested to provide uniform and stable dispersion, including covalent and non-covalent methods. Non-covalent functionalization [34] is the physical adsorption of surfactants or polymers onto CNT walls. Surface functionalization improves dispersion of the CNTs [35,36] and forms chemical bonds on the CNT surface, as attaching carboxylic groups in the covalent method [37–39]. The covalent ligands induce dispersion of CNTs because they decrease the van der Waals forces between the CNTs which leads to change of surface characteristics; however, surface modification was reported to cause defects on CNT walls. Meanwhile, the use of organic solvents is another effective method to improve suspension [33].

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Physical methods to coat CNT surfaces include beam-assisted deposition, but chemical reduction is more common and preferable to prepare silver-NPs because it is easy to control and low in cost. In this study chemical reduction was used to synthesize silver-NPs on the CNT surface.

Due to the idea that carbon nanotubes possess unique properties such as huge surface ratio and antibacterial properties, it has been suggested for biomedical application in many literatures. The previous studies show combining antibacterial properties of silver and carbon nanotube by depositing silver on the CNT surface possess strong bactericidal property [40].

Different methods to reduce silver ions have been investigated; however, the role of organic solvent type in decorating CNTs with silver has not been an area of much focus. Understanding the role and efficiency of organic solvents could promote their application and open new avenues in the preparation of nanosized composites. In the present study, silver crystals were grown on the surface of CNTs in the presence of acetone, N,N-dimethylformamide (DMF), N-methyl-2-pyrrolidone (NMP), and isopropyl alcohol (IPA) as solvents. DMF was used as a reducing agent and sodium dodecyl sulfate (SDS) as a stabilizing agent; the decorated CNTs with silver crystals were tested to understand the role of solvent type.

2. Experimental

2.1. Materials

Multiwall carbon nanotubes (MWCNTs) with 5% carbon impurity (Neutrino Co.) were used (length = 0.5–2 μm ; average outer diameter = 10–20 nm). Silver nitrate (AgNO_3) with 99.9% purity and a molecular mass of 169.87 g/mol, DMF, acetone, SDS, and nitric acid were supplied by Merck (Germany). NMP and IPA were from Sigma-Aldrich.

2.2. Decoration of silver onto MWCNTs decoration of MWCNT by silver nanoparticles

In this procedure, 0.2 g MWCNTs and 0.05 g SDS were immersed in 50 ml acetone. The solution was stirred at 65 $^{\circ}\text{C}$ and 1400 rpm for 1 h. Nitric acid (0.01 M) was added to adjust the pH to 6.0 [41]. Next, 50 ml AgNO_3 (0.1 M) was added to the solution and ultrasonified for 1 h at 65 $^{\circ}\text{C}$ [42]; 15 ml of DMF was then added and stirred for 1 h at 750 rpm. The solution was kept without stirring at room temperature for 72 h to allow formation of the silver crystals, then centrifuged at 3000 rpm for 5 min and washed with distilled water, acetone, and ethanol sequentially 3 times.

The decorated MWCNTs were obtained after drying in an oven for 2 h at 150 $^{\circ}\text{C}$. This procedure was repeated using IPA, NMP, and DMF in place of acetone. All experiments were carried out in a normal

atmosphere. Fig. 1 shows the mechanism of synthesizing silver crystals on the CNT surface.

2.3. Fourier transform infrared spectroscopy

The chemical structure of the MWCNTs was examined using Fourier transform infrared (FTIR) spectroscopy [Nexus 670, Nicolet] in the region of 500 to 4000 cm^{-1} .

2.4. Raman spectroscopy

The Raman spectra of samples were obtained using a Nicolet Almega XR Dispersive Raman Spectrometer (Thermo Electron, Madison, WI, USA) equipped with a 780 nm laser. The spectra were done at 50 to 4000 cm^{-1} with a scan time of 50 s. Raman data acquisition and data processing were performed using Thermo Electron OMNIC software.

2.5. X-ray diffraction

X-ray diffraction (XRD) (Inel, Equinox 3000) was used to assess the crystalline structure of the silver on the CNT surfaces.

2.6. Electron microscopy

Transmission electron microscopy (TEM) was used to investigate the structure of the silver crystals (Philips, EM208) at 100 kV. Field emission scanning electron microscope (FESEM) images were obtained using a Hitachi S-4100.

3. Results and discussion

3.1. Morphology of silver crystals on MWCNTs and dispersion stability of CNTs

TEM and FESEM images show the surface morphology, and the distribution and size of the silver crystals on the CNT surface (Fig. 2). The results show that the morphology of silver crystals can be controlled by changing the solvent. Also the pictures of the dispersions were exploited for the criteria of dispersion states (Fig. 3). It has been shown that the dispersibility of CNTs was remarkably different at the presence of different solvents.

Fig. 2a shows that the particles were spherical and ranged in size from 36 to 80 nm. There were fewer particles on the MWCNT surface than other samples, which can be attributed to the properties of the IPA. IPA is a weak solvent for MWCNTs; they formed sediment after 30 min (Fig. 3a), leaving little time for the silver particles to form on the surface.

Fig. 2b presents CNTs decorated with silver trees in the presence of NMP solvent. NMP is an intense polar solvent that caused to polar

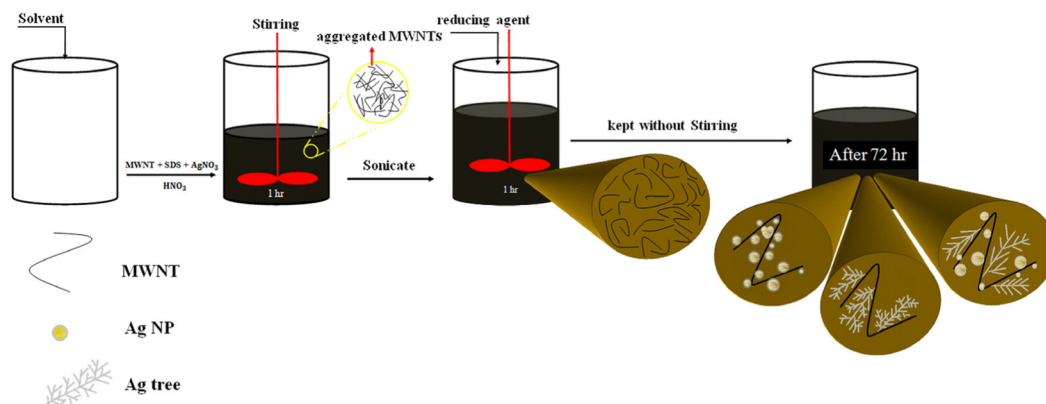


Fig. 1. The procedure of silver crystal growth on the CNT surface.

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