Deposition of Silver Nanoparticles on Dendrimer Functionalized Multiwalled Carbon Nanotubes: Synthesis, Characterization and Antimicrobial Activity

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Introduction

Recently, carbon nanotubes (CNTs) have attracted significant scientific interest ranging from nanoelectronics to biomedical applications owing to their unique structure and properties.[1-5] Despite these great promises, many real applications of CNTs have been impeded by difficulties associated with their processing and manipulation. Formation of amino-terminated nanotubes is a critical step in covalently linked nanotubes and biomolecules because amino groups act as chemical bridges. In this paper, we described the supramolecular functionalization of multiwalled carbon nanotubes (MWCNTs) with *p*-phenylenediamine and methylmethacrylate. This is followed by deposition of metal nanoparticles. The structure and nature of *f*-MWCNTs and *f*-MWCNTs-Ag nanohybrids were characterized by UV-vis spectroscopy, Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy, powder x-ray diffraction (XRD), scanning electron microscopy (SEM) transmission electron microscopy (TEM), x-ray energy dispersive spectroscopy (EDS) and thermogravimetric analysis (TGA). The antimicrobial activity of carboxylated MWCNTs (MWCNTs-COOH), amino-functionalized MWCNTs (*f*-MWCNTs) and Ag nanoparticles decorated functionalized MWCNTs (*f*-MWCNTs-Ag) were comparatively studied against different pathogens.

Experimental

Materials. MWCNTs prepared by chemical vapor deposition (CVD) were obtained from Carbon Nanotechnology Laboratory at Rice University, Houston TX, USA. The sulfuric acid, nitric acid and methanol were purchased from Fischer. Toluene, thionyl chloride, *p*-phenylenediamine, tetrahydrofuran, methylmethacrylate and silver nitrate were obtained from Sigma-Aldrich and were used as received. All aqueous solutions were prepared with ultrapure water obtained from the Milli-Q Plus system (Millipore).

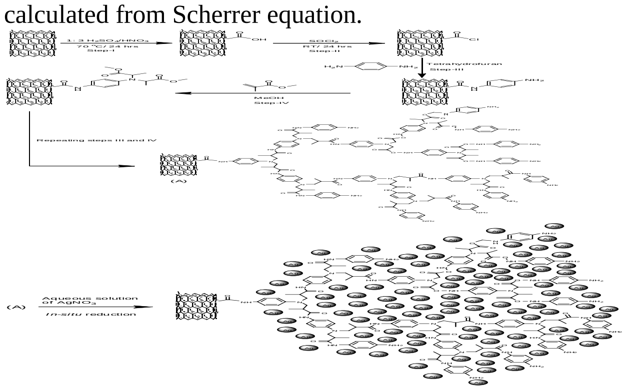
Apparatus and Procedure

The synthesis of functionalized MWCNTs and deposition of metal particles is summarized in scheme 1. The antibacterial activity of MWCNTs-COOH, *f*-MWCNTs and *f*-MWCNTs-Ag nanohybrids were tested at Hygeia Laboratories Inc. Houston, TX USA.

Results and Discussion

Figure 1(a-c) is the XRD pattern of *f*-MWCNTs-Ag nanohybrids, the diffraction peaks at 38.03°, 44.24° and 64.26° can be readily indexed to (1 1 1), (2 0 0) and (2 2 0) Bragg's reflections of silver with face centered cubic (fcc) symmetry.[6,7] This demonstrates the deposition of Ag nanoparticles in their metallic state on the surface of *f*-MWCNTs. The characteristic diffraction peaks of MWCNTs are not clearly visible in *f*-MWCNTs-Ag nanohybrids due to high intense peaks generated from strongly attached Ag nanoparticles to the surface of aromatic polyamide functionalized MWCNTs (*f*-MWCNTs). The estimated domain size of silver nanoparticles using (1 1 1) reflection by the Scherrer equation[8] is ca. 35

The SEM Figure 2a, and TEM images shown in Figures 2(b)-(d) display the morphology of *f*-MWCNTs after deposition of Ag nanoparticles. It could be observed in Figure 6(b) that the *f*-MWCNTs bundles are separated and distributed homogeneously in *f*-MWCNTs-Ag nanohybrids, which suggests the uniform attachment of dendrimers on each nanotube. The average size of silver nanoparticles calculated from TEM is found to be ca. 34±1.2 nm, which is consistent with the particle size calculated from Scherrer equation.



Scheme 1. Illustration of Covalent Modification of pristine MWCNTs and *In-situ* Reduction of Aqueous Solution of $AgNO_3$ and Formation of Silver Nanoparticles on the Surface of *f*-MWCNTs

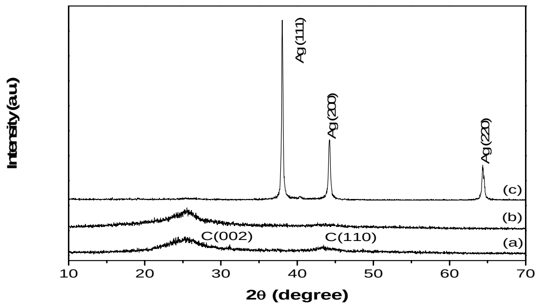


Figure 1. XRD patterns of (a) pristine MWCNTs, (b) *f*-MWCNTs and (c) *f*-MWCNTs-Ag nanohybrids

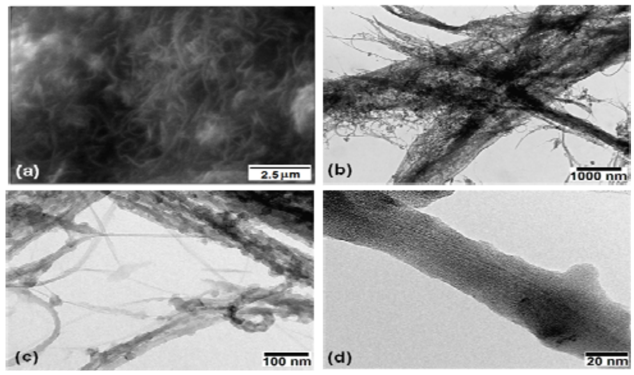


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Figure 2. a) SEM image and (b-d) TEM images of *f*-MWCNTs-Ag nanohybrids

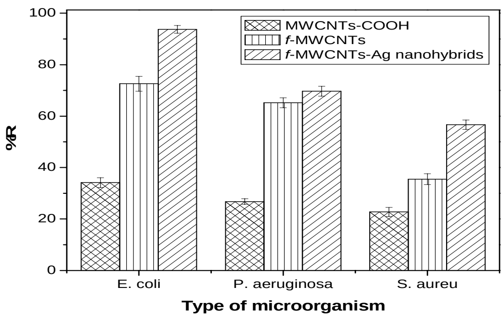

Figure 3. Antibacterial activity of MWCNTs-COOH, *f*-MWCNTs and the *f*-MWCNTs-Ag nanohybrids

The antibacterial activity of MWCNTs-COOH, *f*-MWCNTs and the *f*-MWCNTs-Ag nanohybrids is graphically shown in Figure 3. The *f*-MWCNTs-Ag nanohybrids showed higher antibacterial activity against Gram-negative bacteria, *E. coli* and *P. aeruginosa* compared with Gram-positive bacterium, *S. aureu*. This observed variation in the activity of Gram-positive and Gram-negative bacteria might be due to difference in their membrane structure.

Conclusion

The efficient exfoliation of *f*-MWCNTs and homogeneous distribution of Ag nanoparticles in *f*-MWCNTs-Ag nanohybrids is an indication of the effectiveness of our functionalization method. Our findings revealed that *f*-MWCNTs and *f*-MWCNTs-Ag nanohybrids have higher antibacterial activity than MWCNTs-COOH against against *E. coli*, *P. aeruginosa* and *S. aureu*.

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