Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 132 (2014) 594–598

Contents lists available at ScienceDirect

Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy

journal homepage: www.elsevier.com/locate/saa

Preparation, purification and characterization of high purity multi-wall carbon nanotube

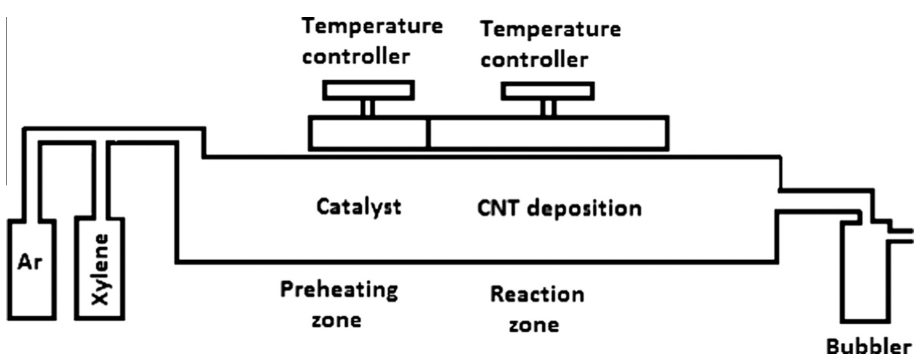
Mohamed Morsy [a], Magdy Helal [a], Mohamed El-Okr [b], Medhat Ibrahim [c,*]

[a] Building Physics and Environment Institute, Housing & Building, National Research Center (HBRC), Dokki, Cairo, Egypt
[b] Physics Department, Faculty of Science, Al-Azhar University, Nasr City, Cairo, Egypt
[c] Spectroscopy Department, National Research Centre, 12311 Dokki, Cairo, Egypt

HIGHLIGHTS

- Chemical vapor deposition (CVD).
- Preparation and purification of multi-wall carbon nanotubes (MWCNTs).
- Characterization tools: SEM, HRTEM, EDX, XRD and FTIR.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 9 March 2014
Received in revised form 14 April 2014
Accepted 22 April 2014
Available online 29 April 2014

Keywords:
CVD
MWCNTs
SEM
XRD
Thermal analyses
FTIR

ABSTRACT

Chemical vapor deposition (CVD) was optimized in order to prepare multi-wall carbon nanotubes (MWCNTs). Preparation of MWCNTs was achieved by the help of ferrocene as a catalyst with continuous flow of xylene. Morphology and structure of as grown and purified MWCNTs were characterized by Scanning Electron Microscope (SEM) and High-Resolution Transmission Electron Microscope (HRTEM). Energy Dispersive X-ray (EDX) spectra for the as grown MWCNTs confirm that the deposits are carbonaceous materials. XRD pattern of purified sample indicates that the Fe peaks at 44.6 and 50.9 have been decreased. This confirms that purification process is effectively reducing Fe component. Further qualitative information on the purification process are indicated and confirmed by the thermal analysis measurements. Finally, FTIR studies have been performed for the identification of the functional group attached on the surface of the MWCNTs. Collecting these results revealed that the optimized CVD is suitable for the production of MWCNTs.

Introduction

Since the discovery of fullerene by Kroto and later on by Krätschmer, it continues to be a topic for research work. Both experimentally as well as through molecular modeling work [1–5]. These classes of compounds show applications in many areas of research [5–11]. Also, it paves the way toward the emerging applications of carbon-based materials. Carbon nanotubes are considered as members of the fullerene structural family. It is a tube-shaped material, made of carbon, having a diameter measuring in the nanometer scale. Multi-wall carbon nanotubes (MWCNTs) can appear either in the form of a coaxial assembly of single wall carbon nanotubes (SWNTs) similar to a coaxial cable or as a single sheet of graphite rolled into the shape of a scroll

* Corresponding author. Tel.: +20 122727636.
E-mail addresses: methat.ibrahim@gmail.com, medahmed6@yahoo.com (M. Ibrahim).

http://dx.doi.org/10.1016/j.saa.2014.04.122
1386-1425/

[12]. Its applications cover many areas of research especially when it is blended with other structure to form nano-composite materials. A novel plasma induced β-cyclodextrin (β-CD) MWCNT/iron oxide is applied for zinc decontamination [13]. It is stated that CD/MWCNT/iron oxide can support longterm use as a cost-effective material in sewage treatment with minimum replacement costs. CNT could also act as an active surface for organo-metallic complexes in order to pre-concentrate heavy metals in environmental samples prior to flame atomic absorption measurements [14]. A composite of ZnONPs/c-MWCNT/PANI/AuE could be used as biosensor [15]. Another composite of MWCNT/TiO_2 core–shell nano-composites is used as photo-catalytic agent for the potential reduction of CO_2 into methane [16]. It is stated that both alumina and zirconia could be blended with MWCNT in order to enhance both the mechanical properties and electrical conductivity [17]. Based upon the above applications for MWCNTs, this work is conducted in order to optimize chemical vapor deposition (CVD) in order to prepare MWCNTs. The prepared samples were purified and characterized with Scanning Electron Microscope (SEM), High-Resolution Transmission Electron Microscope (HRTEM), X-ray Diffraction (XRD), thermal analysis (DTA-TGA) and Fourier Transform Infrared Spectroscopy (FTIR).

Materials and methods

Chemicals and reagents

Ferrocene was purchased from Sigma Aldrich, Germany. Sulfuric acid 95–98%, extra pure was produced from Scharlau, European Union. Xylene technical grade was purchased from El-Nasr Pharmaceutical Company, Egypt. Nitric acid 55% was purchased from El Salam Chemical Industry, Egypt.

Optimization of the CVD

MWCNTs were synthesized by the catalytic CVD method using a fixed bed laboratory reactor. The reactor consists of peristaltic pump, heating mantel, two zones horizontal quartz tube furnace (inner diameter 60 mm and length 1000 mm), gas-delivering system (mass flow controller and on-off valves) and gas bubbler. Ferrocene, xylene and argon was used as catalyst, carbon source and carrier gas respectively. All the reagents were used without further purification. Xylene was chosen as carbon source because of the formation of the grapheme planes is promoted by the two carbon atoms which exist in two methyl groups in the xylene molecules [18]. Fig. 1 shows the schematic setup of the optimized CVD system. The peristaltic pump is used to deliver xylene at constant rate (5 ml/min) to three-neck flask placed inside heating mantel at 250 °C to vaporize xylene which then carried out by Ar to quartz tube located inside furnace. The temperatures in the pre-heated zone and in the hot zone were set to be 200 °C and 800 °C, respectively. The outlet of quartz tube attached to gas bubbler.

MWCNTs synthesis and purification

In a typical run, 100 mg of ferrocene catalyst was placed inside a ceramic boat that was loaded into a pre-heated zone. The temperature of hot zone was elevated to 800 °C under Ar gas at flow rate 750 sccm. As soon as the temperature of a hot zone reached 800 °C the temperature of pre-heated zone was raised to 200 °C, which is considered quite above the sublimation temperature of ferrocene (174 °C). At the same moment, the peristaltic pump is switched on to fed xylene continuously to three-neck flask (inside heating mantel) to convert it to vapor. Xylene vapor flow was allowed for 30 min, and then the furnace was cooled to 400 °C under Ar flow. At 400 °C, the carrier gas was switched on to oxygen to burn out amorphous carbon. Carbonaceous material was deposited as a black film onto the walls of quartz tube, which was collected. After the synthesis, a 500 mg of as grown MWCNTs were suspended in a 50 ml of 3:1 mixtures of concentrated H_2SO_4 and HNO_3 solution and sonicated in a water bath for 2 h [19,20]. Subsequently, the purified multi wall carbon nanotubes (MWCNTs) were washed with distilled water several times and dried at 110 °C over night.

As grown and purified MWCNTs were characterized by various analysis techniques including,

- Philips (Inspect S, FEI Company, Holland) Scanning Electron Microscope (SEM) operated at 30 kV.
- Philips (FEI Tecnai G2 S-Twin) High-Resolution Transmission Electron Microscope (HRTEM) operated at 200 kV.
- Philips XRD powder diffractometer (PW3050/60) with Cu Kα radiation (λ = 0.15406 nm).
- DTA-TGA thermal analyzer (STD-Q 600).
- Fourier Transform Infrared Spectroscopy (FTIR) Vertix 70, Bruker Optik, Germany.

Results and discussion

There are some experimental characterizations in order to indicate that the purified MWCNTs as compared to the as grown samples. First, the morphology and structure of as grown and purified MWCNTs is characterized by Scanning Electron Microscope (SEM) and High-Resolution Transmission Electron Microscope (HRTEM). Fig. 2 shows the SEM image of as grown MWCNTs (Fig. 2a) followed by the purified sample (Fig. 2b). The figure indicates that raw soot contains bundles of aligned nanotubes (nanoropes), in addition to a little amount of metal catalyst impeded into MWCNTs.

One of the most interesting results indicated by SEM, there is no evidence of the existence of amorphous carbon because of burning

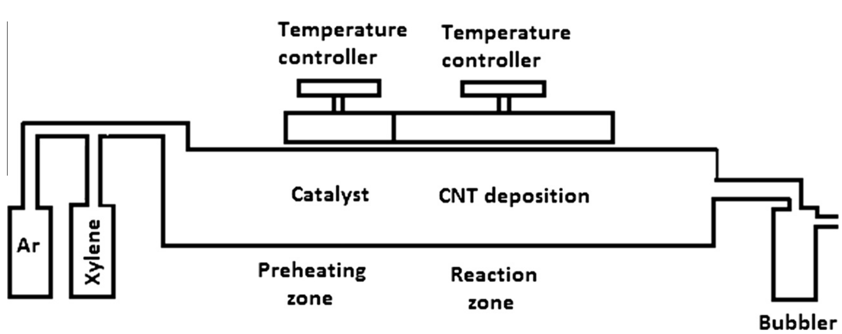


Fig. 1. Schematic diagram for the setup of the chemical vapor deposition (CVD) system which is optimized and then utilized in the preparation of multi-wall carbon nanotubes MWCNTs.