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Bulk Synthesis and Characterization of Multiwalled Carbon Nanotubes by CVD method

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Abstract

Multiwalled Carbon nanotubes (MWCNTs) were effectively synthesized by Chemical vapor deposition (CVD) using metallic catalyst Fe-MgO with growth temperature 700 °C and pyrolysis time at 30 min. The synthesized MWCNTs were characterized by X-ray diffraction (XRD) pattern confirmed the crystalline structure like hexagonal structure (002) plane. SEM morphology of MWCNTs are having in the diameter range of 50-80 nm and length is up to few μm. From the TEM images shown that the MWCNTs with evidence of clear graphitic walls around 15 layers were clearly visualized in 5nm scale. Fe-MgO as good catalytic growth for MWCNTs and also well supported to enhance the bulk production. In future, pure carbon nanotubes and MWCNTs composite materials have been apply for energy storage applications.

Keywords: Multiwalled carbon nanotubes, catalyst, chemical vapor deposition, energy.

1. Introduction

In 1985 chemists invented a new allotrope of carbon like fullerene related structures [1]. Nowadays, Carbon nanotubes CNTs are attracts significantly interest of the scientific community because of their outstanding functions in all areas. The properties CNTs have been attracted for the large variety of interesting and important applications has been suggested supercapacitors, batteries, fuel cells, solar cells, polymer nanocomposites, sensors and biological applications. Carbon nanotubes were exposed by Sumio Iijima in 1991 by electric arc discharge method [2]. The succeeding discoveries of Carbon nanotubes are a tubular structure and their unique properties, an optical, electronic, thermal, mechanical and chemical characteristics a fine distribution nano size, highly surface area, low resistivity, and high thermal stability research into all aspects of CNTs [3&4]. CNTs are widely classified into single walled carbon nanotubes (SWNTs) and multi walled carbon nanotubes (MWNTs) and double walled carbon nanotubes (DWNTs)[5]. Generally, SWCNTs are MWCNTs synthesis different methods, but most widely used common technique carbon arc discharge, Laser ablation techniques [6]. There is a huge demand for quality CNTs both as research materials and large scale industrial applications with optimizing methods to produce more consistent and higher yields. The CNTs were grown by these methods are some difficulties, with unwanted other impurities, which can make them difficult to purify, size control for convenient applications. Currently, the widely follow the chemical vapour decomposition (CVD) method is a controllable process for the selective synthesis of CNTs either individually or in bulk [7]. CVD method has been believed as the most suitable bulk synthesis method in terms of quality of both SWCNTs and MWCNTs [8,9]. Chemical vapor deposition techniques employing the on supported bimetallic metal catalysts have been demonstrated as viable large scale production methods. CNTs are

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synthesis from the carbon containing gaseous decomposition of hydrocarbon sources acetylene (C_2H_2) and ethylene (C_2H_4) as it decomposes at elevated temperature and passes over a transition metal catalyst (typically Fe, Co and Ni). A high yield of nanotubes can be achieved by this method [10 &11]. In the present work, thermal chemical vapour deposition (T-CVD) in the preparation of high yield and multiwalled Carbon nanotubes (MWCNTs) through the decomposition of acetylene over an MgO supported Fe catalyst at pyrolysis temperatures carried out from 700 °C.

2. Experimental

The Fe-MgO catalyst was prepared by impregnating method. Fe $(NO_3)_3.9H_2O$ (Sigma-Aldrich) were dissolved in millipore water with constant stirring. After ultrasonicating of MgO powder in an iron nitrate aqueous solution well mixed, the resulting gel was dried at 100 °C and then grained into a mortar in fine powder. The resulting material wascalcinationsin a furnace at temperature 500 °C for 3 hours [12]. After the impregnation/ incorporation method Catalyst powder materials was grained into fine powder using mortar.

Preparation of CNTs was synthesized using tubular furnace shown schematic diagram of thermal CVD setup fig.1. The catalyst 50 mg was spread over the quartz boat (length-10 cm). CNTs production in tubular furnace with catalyst (Fe-MgO) was placed in the centre of the furnace.

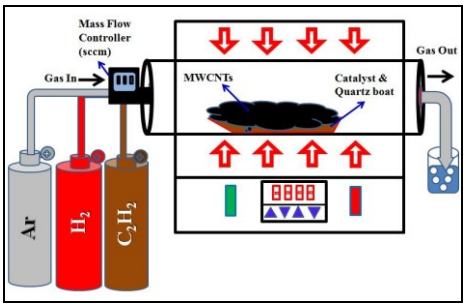

Figure 1. shows the schematic diagram of the experimental setup of CVD method

CNTs growth supported flowing gases sources the argon (Ar) 99.99% gas (gas flow rate 60 sccm) for CNTs growth in inert atmosphere and hydrogen (H_2) 99.99% gas (gas flow rate 60 sccm) was used for the reduction of catalyst. Then the nanotubes growth was started with the introducting acetylene (C_2H_2) (99.9%) gas flow rate 40 sccm with a flow mixed with Ar (Gas) at 700 °C at 30 min CNTs growth. The flow rate of the gases was controlled by a mass flow controller (MFC) in (standard cubic centimeter per minute). After completion the (CNT) Carbon Nanotubes growth reaction the quartz tube was cool to room temperature under Argon (Ar) atmosphere at 60 sccm. The final product was obtained block fine powder from the quartz tube further goes to purification process.

The above obtained CNTs were some impurities and catalytic particles. Hence, the CNTs powder was purified by using acid and heat treatment. These CNTs were stirred with Hydrochloric acid (HCl) (about 35% pure). The CNTs solution was washed continuously and centrifuged. Finally, the filtered material was dried at annealing the 300 °C for in 4 hrs in muffle furnace to remove the impurities amorphous carbon and catalyst. Finally the pure CNT fine powder was materials characterized studies.

2.1. Characterization

Purified MWCNTs crystallographic structures and planes was determined by a powder X-ray diffraction (XRD) system by X'PERT-PRO model with Cu Kα radiation (λ = 0.15406 nm). The Surface morphology structure was analyzed by Scanning electron microscope (SEM) VGA-3 TESCON (30 Kv) instrument and Transmission Electron Microscopy (TEM) model (JEOL 3010) operated at 300 Kv.

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3. Result and Discussion

3.1. X-Ray Diffraction (XRD)

The X-ray diffraction pattern of purified (MWCNTs) is shown in the Fig. 2. The pattern shows the broad diffracted peaks appears at 2θ=26° was assigned correspond to (002) plane. The diffraction peak corresponds to (002) plane and d-spacing value 3.39 Å. From XRD pattern shows for MWCNTs the results reveals that the carbon nanotubes with hexagonal structure with carbon phase and diffraction patterns typically well graphitized.

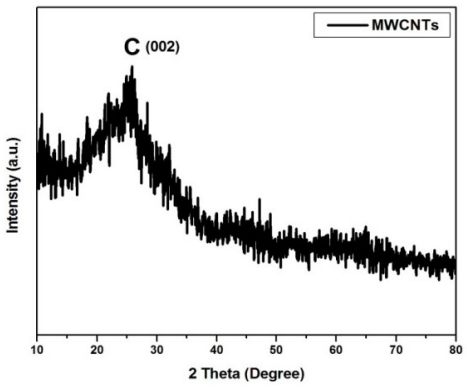

Figure 2. XRD pattern of the MWCNTs

3.2. SEM and TEM

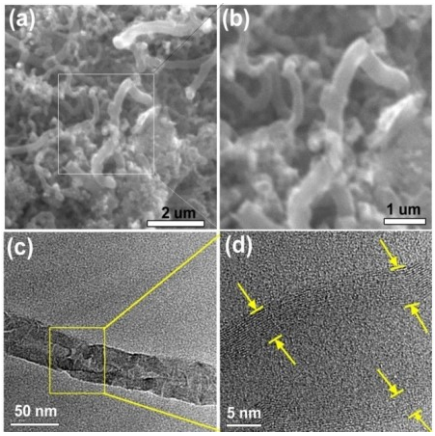

Figure 3 (a) (b). SEM morphological images of MWCNTs and (c) TEM images of MWCNTs. (d) TEM images of MWCNTs in multi walls pattern (scale bar = 5 nm).

The Scanning electron microscope for purified MWCNTs micro scale images are shown in Fig 3 (a) and 3(b). In Fig (a) SEM micrograph are entangled clear morphological view for few CNTs. Fig. 3 (a) shown an magnified view of fig. 3(b) the bended MWCNTs are having in the diameter range of 50-80 nm and length is up to few μm in different magnification. SEM images are clearly indicates the formation of CNTs on the surface of the (Fe) Feric loaded MgO catalyst.

The Transmission Electron Microscopy (TEM) nanoscale images are shown in fig. 3(c) and (d) for purified the MWCNTs. Fig 3(c) shows the individual MWCNTs focused in TEM analysis was confirms that walled that the purifications acid treatments and does not affected the nanotubes structure. Fig 3(d) shows the highest resolution for TEM images can reveal fine details of nanotube structure, including the number of carbon (graphene) layers in their walls, and multi walled carbon nanotubes and fine graphene layers with around 15 graphene walls were clearly visualized (yellow colored aero mark) indicated by TEM image in 5nm scale. The diameter of the Multi walled carbon nano layers nanotubes was found to be few nm and it shows multi layers formed over the MWCNTs during decomposition of acetylene carbon source over the Fe -MgO by Thermal Chemical Vapour Deposition method.

In order to conclude that the SEM and TEM strongly reveals that the MWCNTs obtained over Fe-MgO at 700°C were pure and thermally stable due to the constructions of well graphitized walls due to (Fe) catalyst supported. Which is clearly indicates the MWCNTs were uniformly distributed without any contamination with nano tubular morphologies. The SEM and TEM good morphology and high yield for used Fe-MgO catalyst. Furthermore, Fe catalyst support is enhanced CNTs growth. So the better high

yield preparation defects free CNTs from catalytic-Thermal CVD.

4. Conclusion

Multiwalled Carbon nanotubes (MWCNTs) have successfully synthesized by thermal chemical vapour deposition (TCVD) and catalytic decomposition of acetylene over the Fe metallic nanoparticles catalyst loaded in porous support MgO at 700°C. The XRD result confirms the carbon peak at broad diffracted patterns as graphitized in hexagonal structure. From XRD there are no peaks related to Fe metals used for catalyst due to the purified MWCNTS. The SEM and TEM images showed the morphology of MWCNTs were confirmed the purity of multiwaled CNTs structures and graphitic mutilayers reveals that few nanometer scales. In Future, industrial scale possible production of large scale for energy storage increased the specific capacitance value to develop the symmetric supercapacitor, CNTs to use to electrical energy storage device applications.

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