Synthesis Of Carbon Nanotubes By Electrochemical Deposition using Aluminum Substrate

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الخلاصة

استخدمت تقنية الترسيب الكهروكيميائي لأنتاج أنابيب الكربون النانوية وكانت الخلية الكهروكيميائية مكونة من قطب الأنود (الكرافيت) وقطب الكاثود (الألمنيوم) والألكتروليت المكون من محلول أسيتونيتريل في الماء اللاآيوني بنسبة (1% v/v) وقدرة مجهزة (24V). كان تصرف قطب الألمنيوم في الخلية بأتجاهين، ألأول أرضية لتجميع ونمو أنابيب الكربون النانوية، والثاني مصدر لأنتاج حبيبات الألومينا المائية [Aluminum Oxide Hydroxide(AlOHO)] عالقة في الألكتروليت، هذه الحبيبات هي الأخرى تعمل على تجميع ونمو أنابيب الكربون النانوية. فحص العالق وأرضية الألمنيوم بأستخدام المجهر البصري والمجهر الماسح الالكتروني SEM و تقنيتي XRD و FTIR جميع هذه التقنيات أثبتت نمو الكربون بشكل أنابيب نانوية بقطر حوالي 100 nm على سطح الالمنيوم وقطر أقل من 20nm عالق في المحلول.

ABSTRACT

An electrochemical deposition technique has been employed to produce carbon nanotubes. The electrochemical deposition cell consists of graphite electrode as an anode, aluminum electrode as a cathode and an aqueous solution of organic material as an electrolyte. Carbon nanotube were grown on aluminum electrode using acetonitrile (1% v/v) and water as electrolyte at an applied d.c. potential (24V), The aluminum electrode acts as a deposition surface (substrate) to collect carbon nanotubes and as a source of aqueous alumina [aluminum oxide hydroxide (AlOHO)] particles suspend in the electrolyte which also deposit carbon nanotubes. The suspension and aluminum substrate were characterized by Optical Microscope, Scanning Electron Microscope (SEM), Fourier Transform Infra Red spectroscopy (FTIR) and X-Ray diffraction analysis, all these techniques show the nanotube finger print and the nanotube shape of the deposited carbon.

INTRODUCTION

There are a huge increasing in commercial applications in modern technologies of carbon nanotubes, for example, composite materials, electrochemical devices, hydrogen storage, field emission devices, and nanoscale electronic devices (1). Wide applications of carbon nanotubes are based on their unique physical and mechanical properties, which show the high electrical and thermal conductivities, and high mechanical strength along the tubular axis (2).

There are several methods for producing carbon nanotubes (CNTs) like carbon arc-discharge technique (3), laser ablation, pyrolysis, plasma enhanced, thermal and chemical vapor deposition (CVD) (4). All these synthesis techniques inherently produce carbon nanotubes along with various impurities in the form of amorphous carbon, metal catalysts and many carbonaceous particles, etc. It needed further purification to produce high quality CNTs for device applications (5).

In this study, we described for the first time to the best our knowledge an attempt to synthesis CNTs directly onto the aluminum electrode and in the

electrolyte by a simple electro deposition technique. Aluminum oxide hydroxide has never used yet as a catalyst for preparing CNTs by electrochemical deposition technique, here we used it and had a good results with it.

MATERIALS AND METHODS

Electrochemical deposition cell was fabricated to synthesize CNTs by electrolysis using acetonitrile/de-ionized water (1% v/v) as electrolyte. Electrolysis was carried out at atmospheric pressure and room temperature [6]. Carbon nanotubes were deposited onto cathode (Aluminum sheet Size (0.5x30x100mm)). Graphite was used as the counter electrode (anode). Before mounting the substrates on the cathode, they were thoroughly cleaned and rinsed with de-ionized water and ethanol, respectively. The electrodes were separated by a distance of ~10mm. The applied d.c. voltage between the electrodes was kept ~24 V using DC power supply (HEWLETT.PACKARD 6264B). The deposition was carried out for ~ (4-6) h. One step purification was applied to remove $Al(OH)_3$ from CNT`s, by dissolving impurities in a batch of row material in boiled 2.5N NaOH solution, pure CNT`s filtered and washed with boiled de-ionized water then dried in the oven.

The deposits were characterized by X-ray Diffraction (SHIMADZU XRD-6000), Fourier Transform Infra Red spectroscopy (FTIR) (Shimadzu 8400s), Microscope (NIKON ECLIPSE ME600), Scanning Electron Microscope (SEM).

RESULTS AND DISCUSSION

It should be possible to deposit carbon nanotube by electrochemical deposition technique by suitably choosing the electrolyte and the deposition parameters (7), the possibility of the formation of carbon nonotube structures by electrolysis at an applied voltage of ~24V using aluminum cathode, graphite anode and acetonitrile as the organic precursor at atmospheric pressure and room temperature is demonstrated in this communication (8).

The FTIR spectra of graphite, aluminum oxide hydroxide, untreated CNT and CNT after Sodium Hydroxide treatment are presented in Fig. 1. The FTIR spectrum for untreated CNT (Fig.1-C) indicates many peaks at 1070– 1653 cm^{-1}. The peak at about 1070 cm^{-1} is a characteristic to Al-O-Al stretching vibration. The other peaks 1404 - 1653 cm^{-1} may be attributed to the vibrational modes of carbon nanotubes (6). The broad band centered at about 3450 cm^{-1} could be attributed to the presence of –OH groups [Al-(OH)] (9). The FTIR spectrum for CNT after Sodium Hydroxide treatment (Fig. 1-D) indicates the disappearance of many peaks (1070, 3450 cm^{-1}) and appearance of new peaks which were covered or prevented by the impurities, 2850-3000 cm^{-1} which may be attributed to the C-H aliphatic symmetrical and asymmetrical vibrational modes(10), the peak at about 1456 cm^{-1} is a characteristic to CH_2 absorption, 1338, 3444 cm^{-1} could be attributed to the presence of C-N and N-H groups

respectively and that means the π bonds of acetonitrile was cracked during tailoring process of CNT, The peaks 1404 - 1745 cm^{-1} was appeared which attributed to the vibrational modes of carbon nanotubes (6).

In comparison between the band around 2360 cm^{-1} in Fig.1 (A and D) we found that the band changed from sharp strong absorption band (transparency 24%) for graphite (Fig.1-A) to sharp strong transmission band (transparency 90%) for CNT (Fig.1-D), thereto, the transparency of the IR spectra changed from less than 27% (Fig.1-A)to more than 75% (Fig.1-D) , this is a good argument for changing graphite to CNT(11).

Fig. 2(A-F) shows the reflection micrographs of prepared aluminum oxide hydroxide/CNT deposited on Aluminum sheet electrode with magnifications (A=50X, B=100X, C=200X, D=500X, E=1000X, F=1000X). The micrographs (E&F) clearly indicated the formation of carbon nanotube structures. The CNTs produced on the aluminum sheet seemed to be twisted and the diameter of nanotubes is of order of 100 nm.

Fig. 3(A&B) shows the scanning electron microscope of suspended aluminum oxide hydroxide/CNT after filtration from the electrolyte. The CNTs produced in the electrolyte seemed to be straight and the diameter of nanotubes is of order of less than 20 nm.

We expect that twisty CNTs are the results of very complex change of catalytic activity. The difference between diameters of CNTs on the electrode and CNTs in the electrolyte is due to the size of the catalyst particles.

The XRD spectra (Fig. 4, 5) of the suspension and the deposited film showed (002) (004) reflections which could be assigned to the hexagonal ring structure of the graphite sheets forming the carbon nanotube. Additional peaks for graphite carbon for the reflections from (102) and (105) planes could also be observed. All the peaks are slightly shifted to lower angle from that of graphite indicating the wider interlayer spacing. The (002) peak position of the CNT deposits on Al was located at $2\theta = 26.54^{o}$ while for the suspended CNT was located at 2θ-28.2^{o} (6, 9).

Using an electrochemical deposition method, carbon nanotubes have been synthesized from organic solutions at room temperature. The formation and growth of carbon nanotubes are catalyzed by aluminum oxide hydroxide catalyst formed immediately during electrochemical process on the aluminum electrode and in the electrolyte. SEM characterization shows that the diameter of nanotubes is of order of 100 nm deposited on the aluminum sheet and is of order less than 20 nm suspended in the electrolyte depending on the size of catalyst particles.

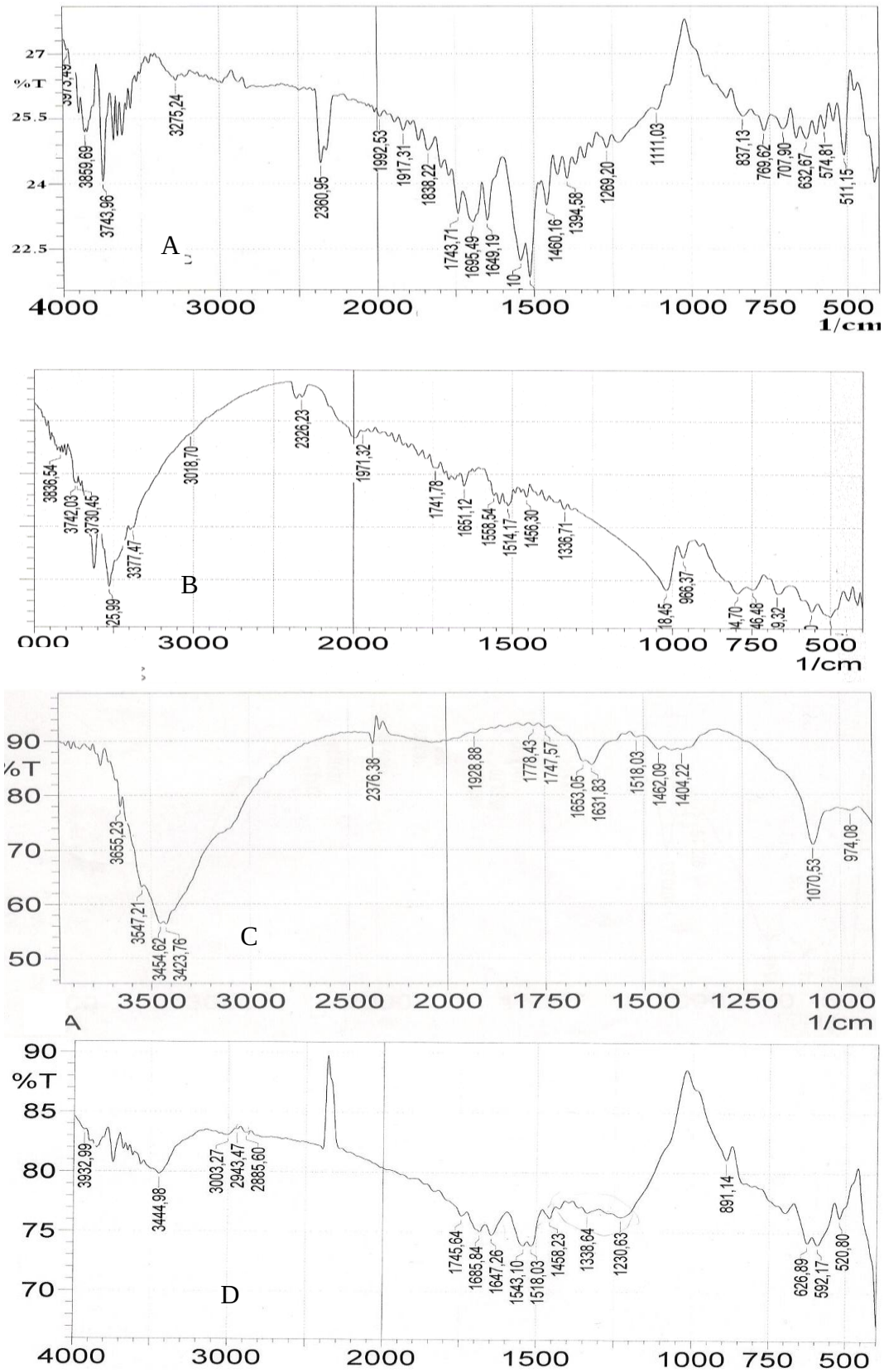

Fig.-1:FTIR absorption spectra of (A) Graphite, (B) Aluminum oxide hydroxide, (C) Untreated CNT and (D) CNT after Sodium Hydroxide treatment.

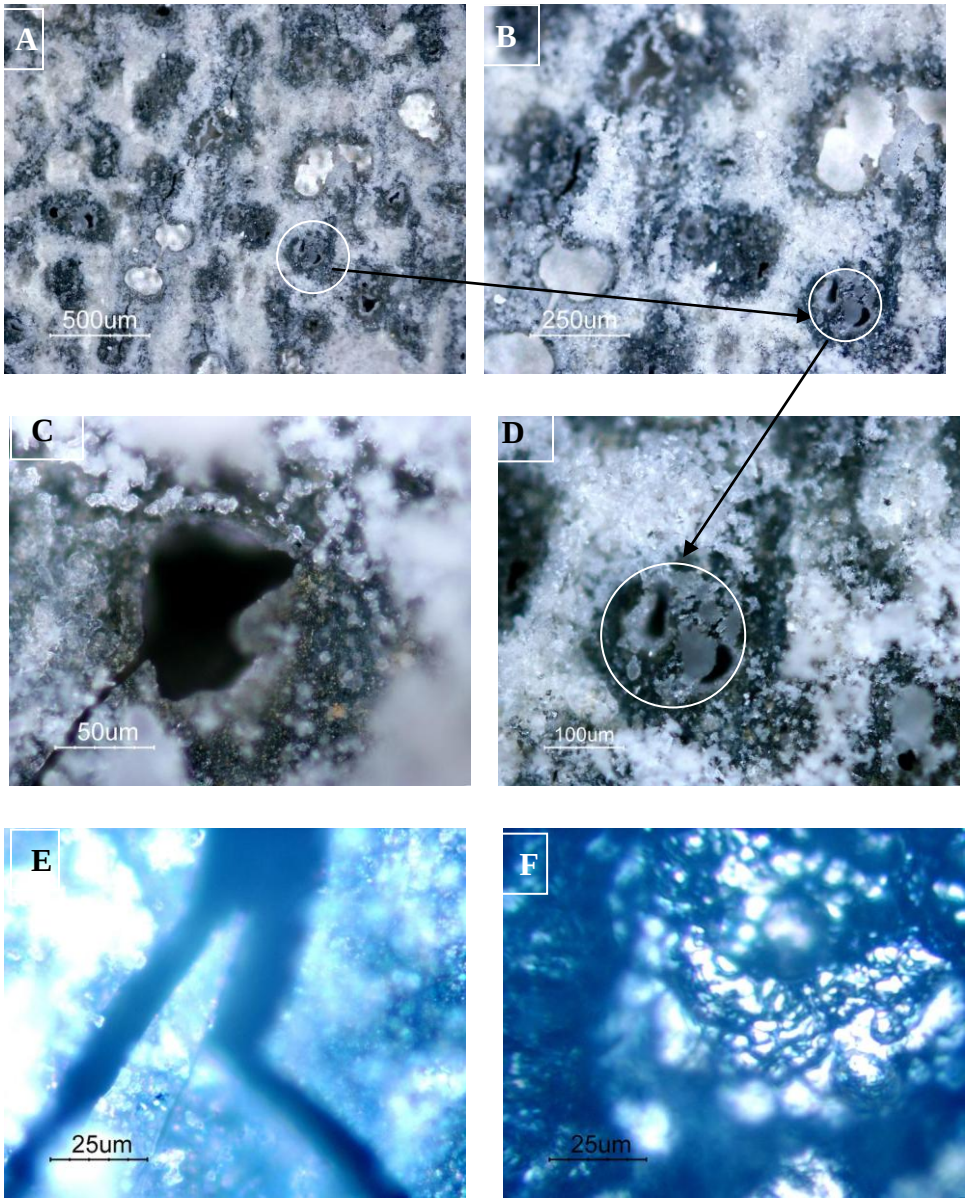

Fig.-2:Reflection micrographs of prepared Aluminum Oxide Hydroxide /CNT deposited on Aluminum electrode.

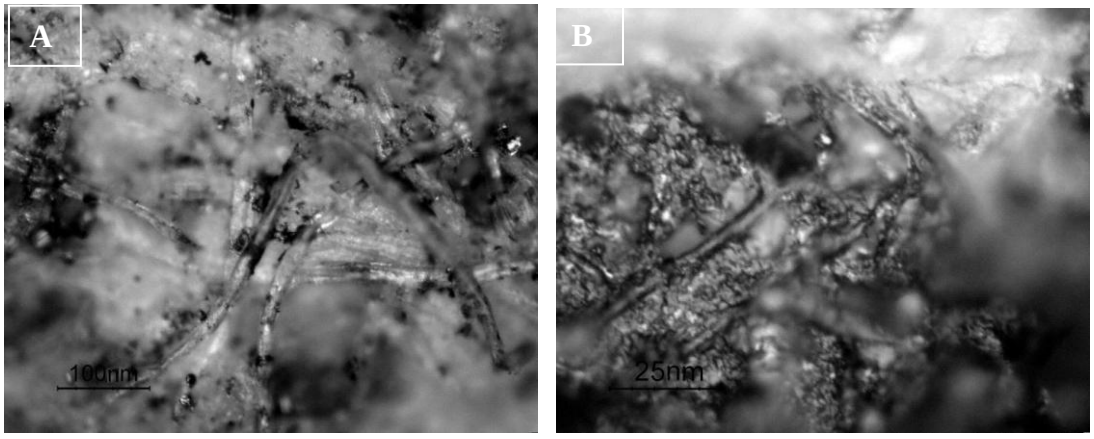

Fig.-3.SEM micrographs of suspended Aluminum Oxide Hydroxide /CNT after filtration from the electrolyte.

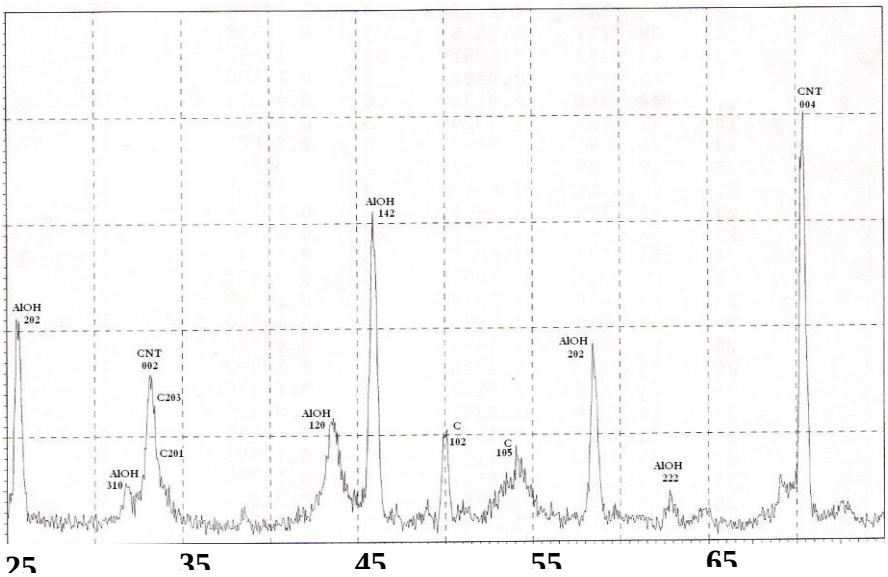

Fig.-4: XRD analysis of suspended Aluminum Oxide Hydroxide/CNT after filtration from the electrolyte.

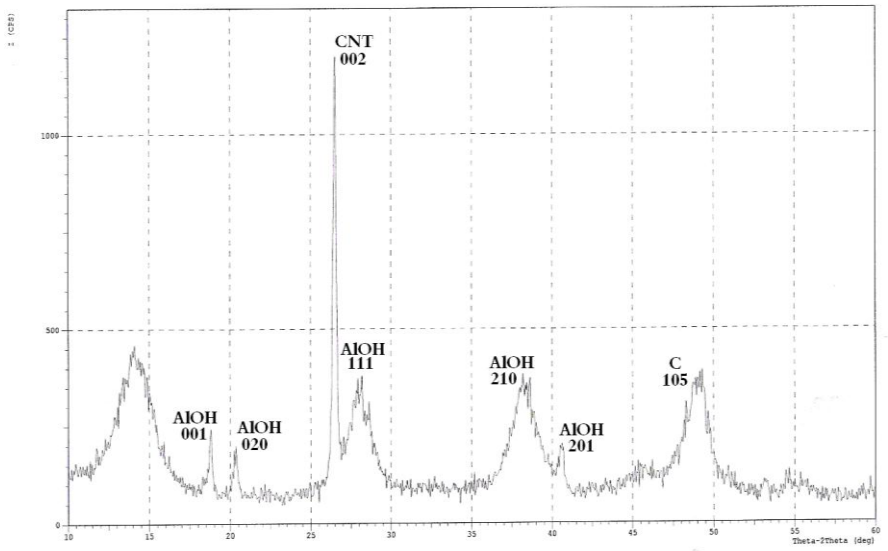

Fig.-5: XRD analysis of Aluminum Oxide Hydroxide/CNT deposited on Al sheet.

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