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## Enhance the Growth of Multi- walled Carbon Nanotubes from Coal by Catalytic Chemical Flame Deposition

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## Enhance the Growth of Multi-walled Carbon Nanotubes from Coal by Catalytic Chemical Flame Deposition

Safa A. Yassin, Abdulqadier H. Al khazraji , and Firas H. Abdulrazzak  
Chemistry Department, College of Education for Pure Science, Diyala University  
Corresponding author's (firas\_habeb2000@yahoo.com)

### Abstract

Multi-walled carbon nanotubes were synthesized by catalytic chemical flame deposition CCFD by magnesium as a catalyst for growth from coal/kerosene/sulfur with ratios 4 / 1.3 / 1 respectively. The CFD was more efficiency with the magnesium as a catalyst, which led to increasing MWCNTs growth as compare with growth without a catalyst. The synthesized MWCNTs were characterized by X-ray diffraction, Raman spectroscopy and scanning electron microscopy which showed high dense with length growth. The diameters of tubes were more homogenous when precipitation on the surface of Mg with diameter 39-57nm while 31-64 nm without a catalyst.

**Keywords:** CCFD, MWCNT, coal, kerosene, high dense

### 1.Introduction

Synthesized of nanomaterials NMs witness particular attention from researchers all over the world which deals with synthesis, purification, characterization, and attempts to use in many applications [1]. Actually, carbon nanotubes CNTs represent the distinctive mark for a 20th century due to an unusual form and properties of the tubular structure identified in a famil of nanomaterials. CNTs commonly define as the tubular structure of graphene or graphite consist of carbon atoms bonded by sp<sup>2</sup> hybridization in planar sheets which adhesive to one /or more than one sheet of graphene by Wan-der Walls forces [2]. Chemical vapor deposition CVDs, arc - discharge ADCs, and laser ablations LAs represent the primary methods for synthesized different types and quality



with quantities of CNTs [3]. Mostly graphite after evaporation under electrical discharge or decomposition of hydrocarbons with or without metal nanoparticles in a reducing or inert atmosphere represent a source of carbon for preparing CNTs [4]. The coal is black or brownish-black carbonaceous material with different hybridization and trace elements bonded or adsorbed with carbon [5] which decided combustibility. Coal as a source of carbon rarely used for synthesized CNTs for many causes such as purities and difficult to convert to make it suitable for CVDs, ADSs, and Las [6]. The solid state for coal prevents or at least limits the abilities to use it only for specific ADSs [6,7]. The best chemical treatment with coal which could produce CNTs is chemical flame deposition method CFDMs [2]. The process needed for a specific chamber to achieve the principle of decomposition for coal over support with less value of oxygen. In this work, MWCNTs were synthesized from a mixture of coal/kerosene/sulfur under limit oxygen at 140°C with and without Mg as a catalyst for growth tubular structure.

## **2. Experimental**

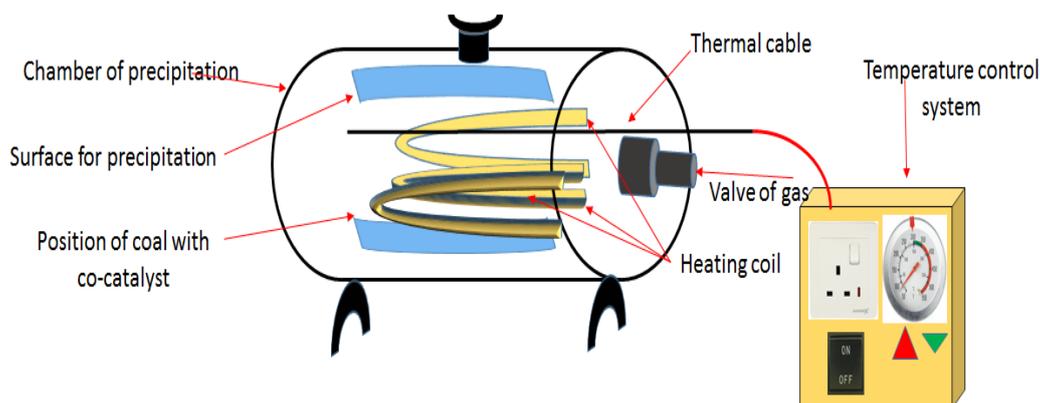
### **2.1. Materials:**

The coal which used as a source of carbon and kerosene were purchased from the local market. Magnesium nitrate  $Mg(NO_3)_2$  was purchased from Sigma-Aldrich with purities 98% and Hydrogen peroxide  $H_2O_2$  was purchased from Barcelona, Spain with 60% percent weight.

### **2.2 Chamber of synthesis V-MWCNT by CFDMs:**

Carbon nanotubes were synthesized by using coal as sources of carbon with a used mixture of kerosene and sulfur at 140°C under limit atmospheric oxygen without/or with Mg as a catalyst for growth CNTs. The process of CFDMs was done by homemade reactor as shown in Figure 1. The iron cylindrical reactor with 30 cm<sup>3</sup> in volume and 50 cm in

length supplied with three heating coil U tubes Heater, 1.5 KW, which equipped with a thermal cable to controlling the heaters inaccuracy  $\pm 1$  °C.



**Figure 1: Skim diagram for the reactor of CFDM**

At first, we should refer to important things which are coal alone or with kerosene or with sulfur did not show any activity towards produce CNTs in the conditions of preparations. The mixture was placed in the center of the chamber which includes coal/kerosene/sulfur with the ratio (4 / 1.3 / 1), respectively. The rolls of kerosene and sulfur represent by increase the ignition capacity of the coal in order to obtain a minimum degree of deposition with lower temperature. Switched on the apparatuses until reach for 140 °C which setting for 15 minutes then switched off with kept close the doors of the chamber until reach room temperature. The synthesized CNTs were purified by dispersion with 25 mL hydrogen peroxide <sup>[8]</sup> at 20 °C with stirring for half an hour then allowed to reach room temperature. Shake the product by separating funnel for 15 minutes, then separate the precipitation with washing by distal water before thermal treatment at 100 °C for 3 hours.

### 3. Characterization

X-ray diffractometer (Riga Rotaflex) (RU-200B) was used to analysis the crystallography of MWCNTs with 0.15405 nm radiation from Cu  $K\alpha$  between  $10^\circ$  - $90^\circ$  with  $5^\circ/\text{min}$  in a scan rate and  $0.02^\circ$  for resolution. Raman spectroscopy was done by Sentara infinity 1 Broker, intensity 2m W for 5 lops per 2s at 530 nm using light leaser and resolution 3-5 cm. Scanning electron microscopy SEM measurements were carried out by a JEOL JSM--6700F.

#### 4. Results and discussion:

Figures 2 and 3 refer to two types of synthesized CNTs with and without catalyst were shown two peaks at  $24^\circ$  and  $43^\circ$  which represent typical tubular structure <sup>[9]</sup>. The CNTs which prepared without catalyst shows less intensity and width as compare with CNTs when prepared by using catalyst. The XRD in figure 2 based on sample without catalyst , many peaks at  $2\theta = 28^\circ$  ,30,32,33,35 and  $48^\circ$  which represent to amorphous carbon with tubular structure, while the sample which supported by Mg catalyst have more amorphous carbon with tubular structure ,so we believe that it can be related to abilities for Mg towards carbon growth to CNTs, figure 3. The Mg particles act as an active site for interact with carbon free radical with  $sp^2$  hybridization as unite cell for the hexagonal of the tubular structure.

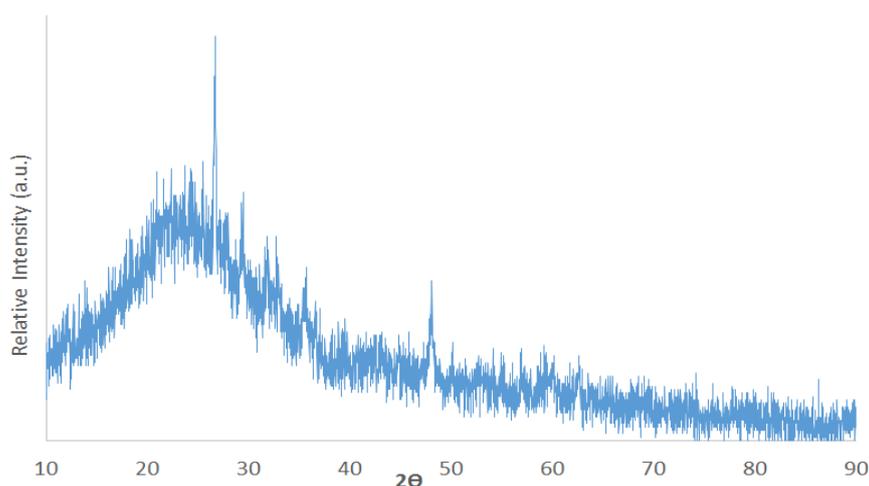


Figure 2: XRD patterns for synthesized CNTs without a catalyst.

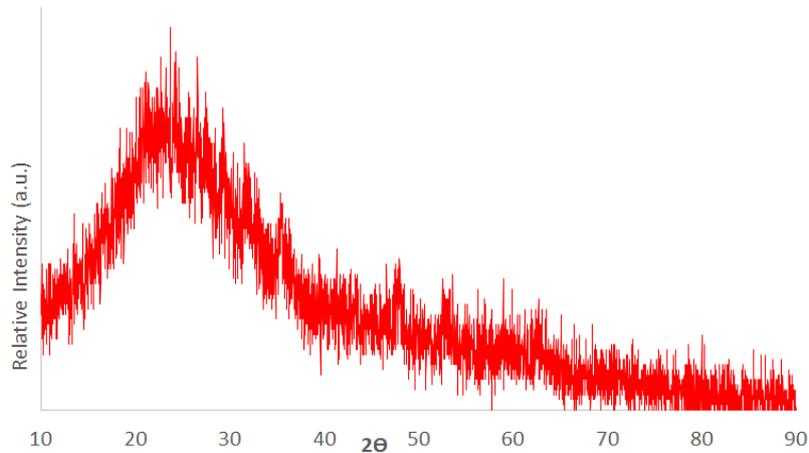


Figure 3: XRD patterns for synthesized CNTs on the Mg as catalyst.

The two synthesized materials were analyzed with Raman spectra to confirm the presence of CNT as shown in figure 4 and 5. Characteristic D-band and G-bands, were obtained from the MWCNT at  $\approx 1343 \text{ cm}^{-1}$ ,  $1593 \text{ cm}^{-1}$  and  $1365 \text{ cm}^{-1}$ ,  $1569 \text{ cm}^{-1}$  which related to sample without and with Mg as catalyst respectively[10]. Raman spectroscopy witness more strong peak for MWNTs which produce in exist of the catalyst at  $2958 \text{ cm}^{-1}$  for D+G band while for MWCNTs without catalyst was unclear. The reason for reducing D+G for MWCNTs when growth without catalyst can be related to reducing the growth ratios of MWCNTs[11] which prevent make the peak clear. The ratios of  $I_D/I_G$  for the two synthesized materials 1.43 and 1.46, respectively shows that use Mg as catalyst enhance the disorder and the defects on the sidewall of tubes [12].

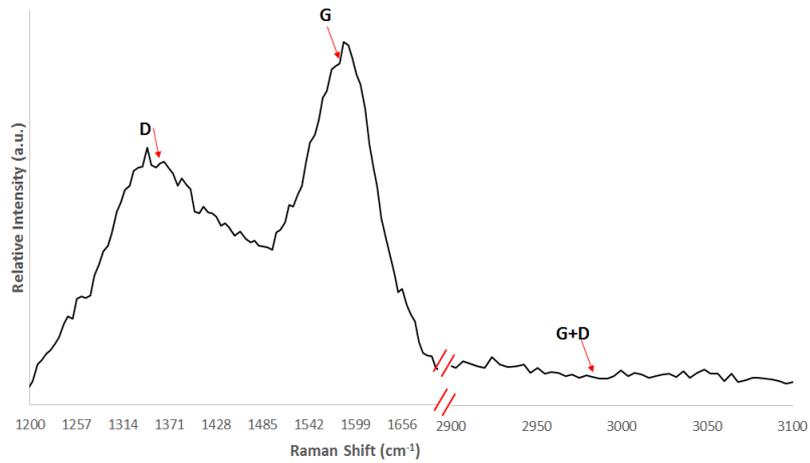


Figure 4: Raman spectroscopy for synthesized CNTs without a catalyst.

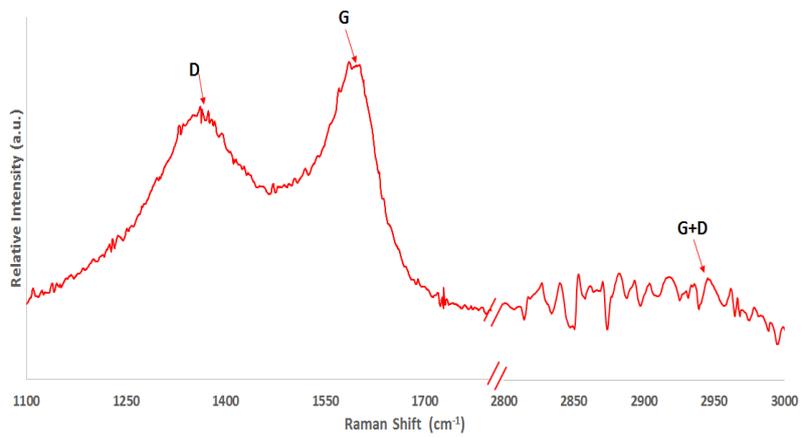


Figure 5: Raman spectroscopy for synthesized CNTs using Mg as catalyst.

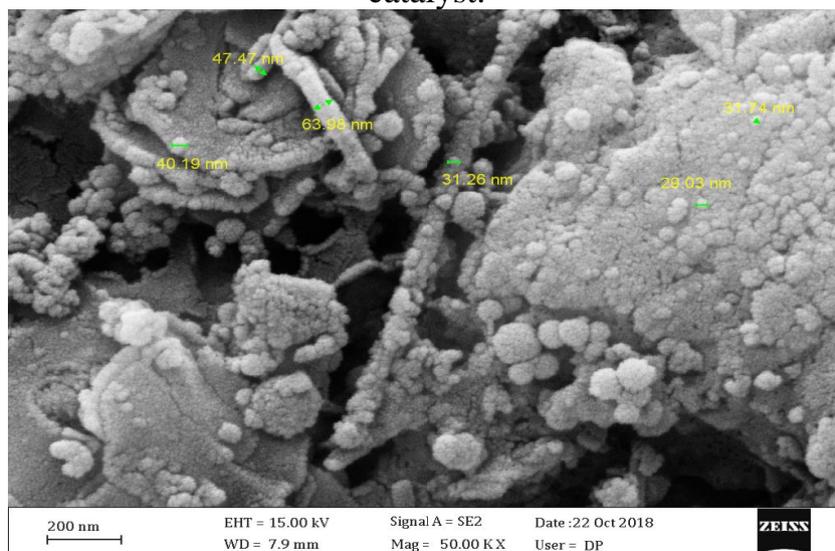


Figure 6: SEM image for synthesized CNTs using Mg as a catalyst.

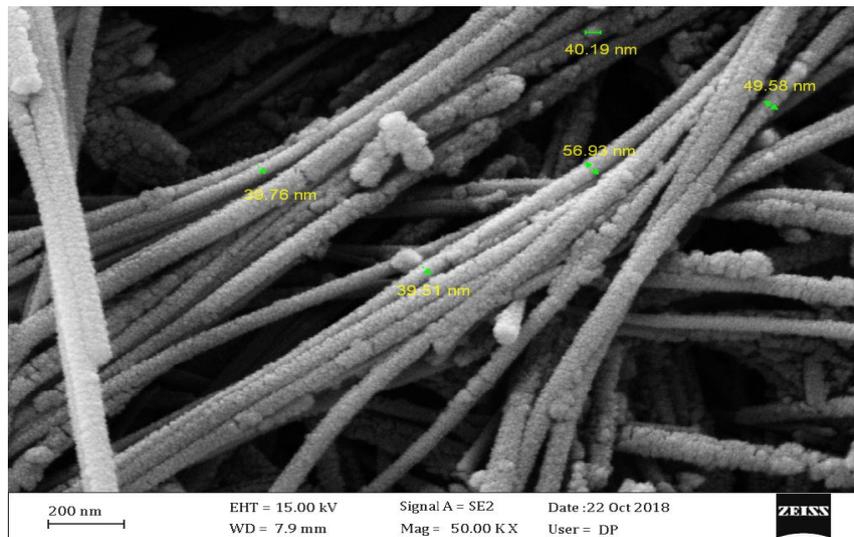


Figure 7: SEM image for synthesized CNTs by using Mg as a catalyst.

Figure 6 and 7 shows the SEM images for MWCNTs which synthesized with two different conditions for precipitation. The MWCNTs which prepared without using catalyst shows few filament of carbon with average diameter 31-64 nm and length more than 1.5  $\mu\text{m}$  with many species of unconverted carbon. The second types were prepared by deposited onto Mg surface showed many filaments of carbon at diameter 39-56 nm with length more than 2  $\mu\text{m}$  and less amount of unconverted carbon. The process of building tubular structure depend on the temperature of condensation and the effect of the catalyst with free radical. Influence of temperature was controlled when Kerosene and sulfur enhance the coal ignition to obtain a maximum degree of deposition with lower temperature when prevent converting coal to carbon dioxide [13]. The Mg particles adsorb carbon monoxide molecules then decomposed of them to produce carbon free radical, which interact with catalyst surface after that complete the growth of opening tubes to the upper direction. As reported [14] the catalyst act as a donor or acceptor electrons enhance the formation and condensation of free radicals. During decomposition by flame, different free radicals fragments will produce than the catalyst particle's role simply to provide

an interface where carbon rearrangement can occur and act as a template for growth [15]. From the analysis technique that used to characterized MWCNTs" hydrocarbon dissociation is not essential for growth of CNTs when exist the catalyst on the support" of growth without ideal conditions for precipitation [16]. The preferential reactivity of Mg towards the growth of CNTs may also be related to many active sites for able to build a tubular structure with sp<sup>2</sup> hybridization of carbon atoms which did not exist without Mg.

#### Conclusion:

Two conditions for preparing MWCNTs in the flame method without and with Mg as a catalyst for growth in this work which explained by condensation and free radical as causes for the mechanism. The results show that MWCNTs which prepared with Mg are denser and length compare with the product without a catalyst. The mixture of kerosene and sulfur were succeed to reduce the temperature of ignition of coal which prevent or at least reduce the ratios of CO<sub>2</sub> as compare with CO.

The ratio of I<sub>D</sub>/I<sub>G</sub> of the MWCNTs for the process with catalyst Mg was more than the product without a catalyst. SEM show that the averaged diameter and length of carbon nanotubes synthesized by the catalysts are more homogenous and graphitized that refer to improving the production quality and quantities of carbon nanotubes.

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