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Synthesize of $MWCNT_S$ by CCVD on Fe Catalytic Substrate and Investigation of Acidic and Thermal Treatment Effects as Purification Procedures on the Structure of the Synthesized CNT_s

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ABSTRACT

In this work, Multiwall Carbon Nanotubes (MWCNTs) were grown by CCVD method on Fe nanoparticles as catalyst. In order to purify the produced CNTs from contaminations which could be as a results of existing some impurities in the reaction chamber during synthesizing procedure, such as amorphous carbons, catalyst nanoparticles, as well as different gases from the atmosphere of reaction chamber, which could be effect the quality of synthesized CNTs and its physical characteristics some purification procedure has been employed on samples including thermal treatments, washing in acidic environments by using different acids (HCI, HNO_3 and H_2SO_4) with different molarities under reflux in ultrasonic waves in an ultrasonic bath. It is well known that the existence of impurity atoms in the structure of the produced CNTs could seriously affect its quality and physical characteristics, especially when it is used for microelectronic porpoises. In this work we have tried to recognize the most suitable method of purification applicable to MWCNTs, without destructive effects on the samples structure. To do this the X-ray diffraction pattern, Raman spectroscopy and electron spectroscopic images (Scanning, SEM and Transmission TEM) from the samples have been produced before and after purification procedure to figure out the effects of each purification procedure on the samples.
KEYWORDS: "MWCNTs; Chemical Vapor Deposition; Purification; SEM; TEM"

INTRODUCTION

Carbon Nanotubes are one of the carbon structures, which were first accidentally explored by a Japanese researcher called Sumio Iijima microscope specialist in NEC laboratory in 1991 [1-5]. Due to their unique electrical and mechanical features, these nano materials are highly focused by scientists. Special features of Carbon Nanotubes like accept ratio (ratio of length to diameter) have resulted in their application in the improvement and development of composites, Micro Electro – Mechanical Systems (MEMS), as well as Nano Electro Mechanical Systems (NEMS) [6-9]. Carbon Nanotubes can be used as emission sources for electrical fields, nano electronic, and storage systems of same gasses such as Argon and Hydrogen. Carbon Nanotubes are made of twisted crystal plates and mono atoms of graphite [10, 11]. In addition to their extraordinary elastic stability, they have displayed various electrical features dependant on their types of the twisted graphite plates and their diameters and can be considered conductor, semiconductor or insulator [12]. Purification of Carbon Nanotubes is an important matter for their applicability, so that it may be possible to use highly purified carbon Nanotubes in various applications as well as in different industries such as nano electronic industry [5]. Carbon Nanotubes are produced through several methods such as Arc – discharge, laser ablation and chemical vapor deposition (CVD) method [13-15]. In this work, the effect of thermal and acidic treatment involving an agent such as mixed acid HNO_3/HCl and H_2SO_4/HNO_3 were investigated on $MWCNT_S$.

Experimental procedure

$MWCNT_S$ were grown on boat of quartz substrate. The employed substrate were washed with acetone and distillated water, and then the Fe catalysts were distributed on low wettish substrate at atmosphere pressure. Finally, the substrate was dried in air and placed in center of quartz furnace. Synchronized with the preparation of substrate, we gradually increase the temperature of the electric furnace of CVD device to 925°C in order to synthesize the Carbon Nanotubes. During this process, to avoid the penetration of any kind of contamination to the quartz furnace, we stream the inert Argon gas with the flow rate of 800Sccm. Finally, after preparation of the substrate and reaching the temperature of quartz furnace to 925°C, we decrease the flow of Ar gas to 100Sccm and place the substrate, impregnated to catalytic Fe nanoparticles in the center of the quartz furnace and then we flow the C_2H_4 gas as the carbon source, with the flow rate of 25Sccm. After 30 minutes, we complete the synthesis conditions, turn off the electric furnace, and cut off the flow of C_2H_4 gas so that the synthesized sample will be cooled down to the atmospheric temperature by Ar gas flow.

The purification process includes three methods; thermal oxidation, acid washing and reflux with ultrasonic waves. We examined the effect of thermal oxidation operation at 450°C temperature for one hour in the purification

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process of the samples in order to remove surface contaminations including Carbon soot, Amorphous Carbon structures, etc on Sample 1.

The purification process of Sample 2, 3, and 4 is as follows:

1. Thermal oxidation at 450°C temperature for one hour
2. Reflux of the sample by ultrasonic waves in distilled water solution for 3 minutes and sample resting for 3 hours
3. Passing the sample through filter and collecting it
4. Reflux by ultrasonic waves in (HNO_3/HCl (1:1) 3M for sample 2, H_2SO_4/HNO_3 (3:1) 3M and 5M for samples 3 and 4 respectively acid solution for 5 minutes and sample resting for 3 hours
5. Passing the sample through filter and washing the sample with distilled water
6. Collecting the sample and drying at the moderate temperature of 50°C
7. Thermal oxidation at 450°C for one hour

RESULTS AND DISCUSSION

Figure 1 show the SEM image of Fe Nanoparticle was obtained from Fluka, Aldrich Chemicals, USA. The SEM image and the results of XRD pattern and Raman Spectroscopy of the synthesized sample are shown in Figures 2, 3 and 4, respectively.

Figures 5, 6, 7 and 8 show the SEM images of $MWCNT_S$ (1^{st}, 2^{nd}, 3^{rd} and 4^{th} samples). Besides, the XRD and Raman spectra of sample 5 are also shown in Figure 9. At the end, for better study and analysis of the synthesized $MWCNT_S$ samples, Transmission Electron Microscope is used. The TEM image of Sample 5 is shown in Figure 10.

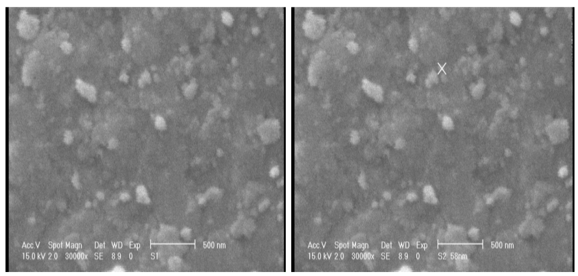

Figure1: SEM Image of Iron Nanoparticles

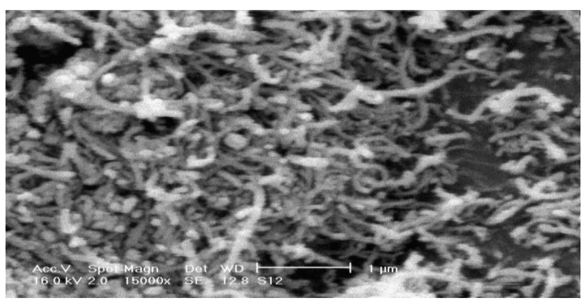

Figure 2: SEM image of the raw $MWCNT_S$

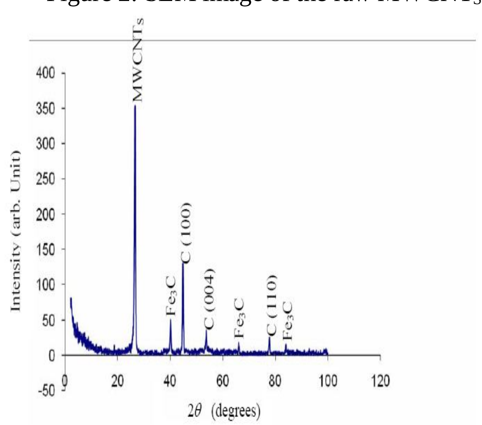

Figure 3: XRD pattern of the as-grown $MWCNT_S$

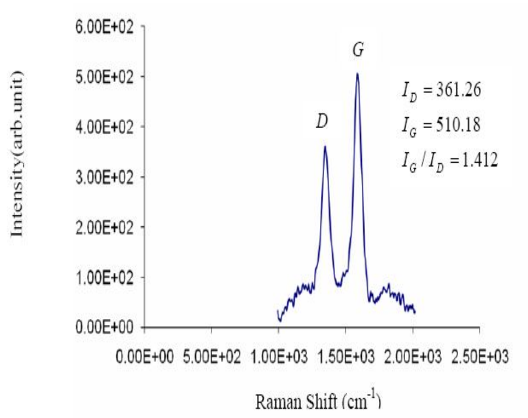

Figure 4: Raman spectra of the as-grown $MWCNT_S$

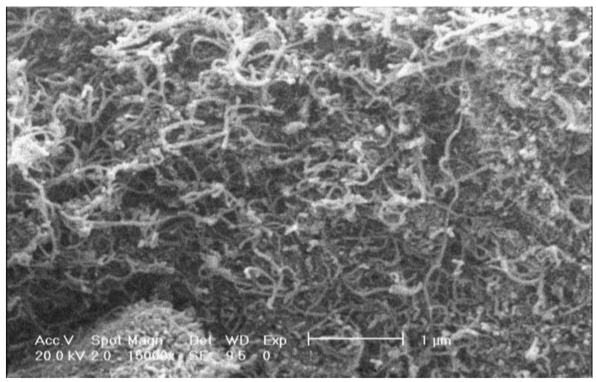

Figure 5: SEM image of the purified $MWCNT_S$ by thermal treatment (450°C for 1h)

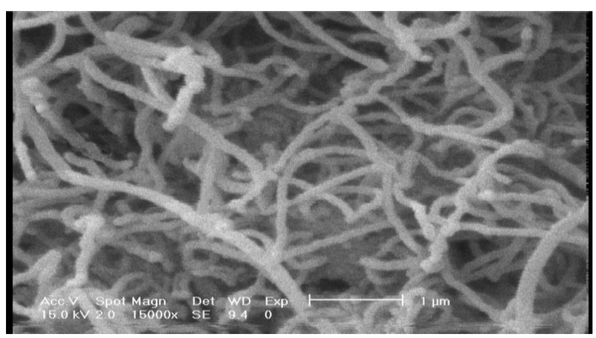

Figure 6: SEM image of the purified $MWCNT_S$ by HNO_3/HCl (1:1) 3M

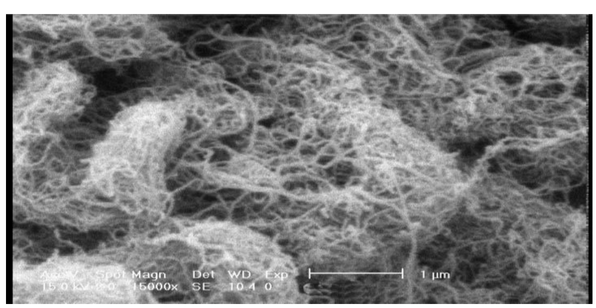

Figure 7: SEM images of the purified $MWCNT_S$ by H_2SO_4/HNO_3 (3:1) 3M

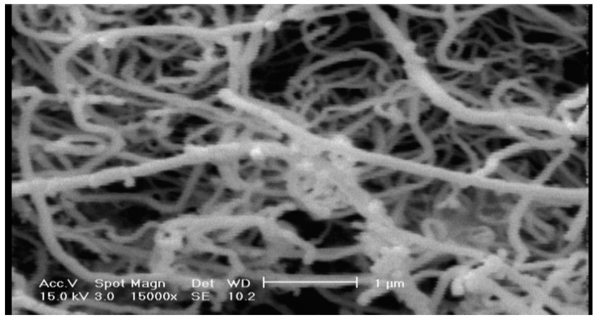

Figure 8: SEM images of the purified $MWCNT_S$ by H_2SO_4/HNO_3 (3:1) 5M

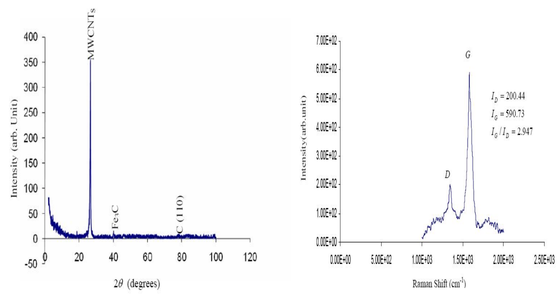

Figure 9: XRD pattern and Raman spectra of the purified $MWCNT_S$ by H_2SO_4/HNO_3 (3:1) 5M

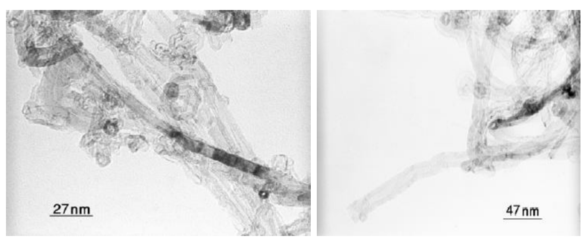

Figure 10: TEM images of the purified $MWCNT_S$

Conclusions

Washing the samples with acid results in more purification of the product from surface and catalyst contaminations and the purification of samples leads to more desirable SEM images. By comparing the image of prototype with those of the other samples, it can be concluded that the purification processes have been effective in removing contaminations from the prototype. The results obtained from XRD pattern for the purified Sample 5 compared with the prototype (before purification) indicate that most of the peaks related to the impurities are removed properly from the prototype and also the increase in the amount of I_G/I_D to 2.947 in Raman spectrum shows good quality of Carbon Nanotubes in the sample. In addition, according to the obtained TEM images, one can confirm that the synthesized samples are Carbon Nanotubes. The diameters of Carbon Nanotubes formed in the samples are within the range of 20 to 100 nm.

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