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Subdued Intensity Variation of Graphite Peaks and Variation of Raman Shift in the Synthesis and Characterization of Carbon Nanotubes

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Abstract: ***Carbon Nanotube (CNT) is a new form of carbon, have aroused huge interest in the research community because of their interesting electronic structure. The CNTs are possibly handy in an extensive variety of solicitations. They unveil phenomenal asset, strange electrical properties and efficient heat conductors. Currently Chemical Vapour Deposition (CVD) has been widely used for the preparing carbon nanotubes because of its simplicity, high throughput and low running cost. It is also easy to control the growth parameters compared to other methods. In the present work methane is used as the source of carbon and ferric nitrate is used as a catalyst. Also, zeolites are used as absorbents and support for catalysts. The growth is carried out in a tubular furnace. The surface morphology, vibrational and structural properties were studied by using FESEM, laser Raman. FESEM micrograph shows the growth of carbon nanotube with different shapes. The length of the tube is ~1.08µm while its diameter is ~70nm. Raman spectra shows the absence of radial breathing mode (RBM) at lower Raman shift and the D and G-band evidence the presence of multi walled carbon nanotubes. XRD pattern reveals that, the intensity of graphite peaks are very subdued because the presence of higher content of zeolite.TGA curve shows, the as grown CNT has a weight gain due to oxidation or oxygen pickup by zeolite. Carbon nanotubes is an advance developing field in research. Carbon nanotubes are the strongest and stiffest materials in terms of tensile strength and elastic modulus. The present work focuses on the growth of Carbon nanotubes using chemical vapour deposition. Although there are many methods, the chemical vapour deposition is chosen because of its simplicity, high throughput and low running cost. Also it is very easy to control the growth parameters compared to other methods. . Carbon nanotubes has a wide application range in the field of Gas sensors, medical science for drug delivery system, field emission, energy storage and solar cells.***

Keywords: CNT, Zeolite, FESEM, Laser Raman

I. INTRODUCTION

Since Iijima described the certainty of carbon nanotubes (CNT) in 1991, [1] they have concerned great courtesy because of their great physical and chemical properties. [2,3] The exclusive properties of CNTs brand them tremendously sign for electrochemical sensors and biosensors.

[4–6] Modern analyses validated that CNTs illustrate hard-hitting electro catalytic activity and minimization of surface snarling if hired to develop the electrochemical reaction of some significant bioactive materials.

[7–10] A variety of carbon-nanotube electrodes have been fabricated for identifying different redox composites[11-16], CNTs were frequently isolated in solvents [17, 18] or polymer resolutions [19, 20] that were cast on the electrodes, because CNTs were insoluble in most solvents. Modern, techniques centered on electrochemical deposition [21] and electrochemical polymerization [22, 23] has been hired for stacking CNTs onto electrodes. CNT-based electrodes have been verified to diminish the over potential significantly [4, 21].

Progression of Multi-walled CNT by CVD method has massive bids in the field of Dye engineering [24], Electronic wrapping (To guard and cool the micro-electronic chip) [25], Superconductors [26], Acrylic bone cement [27], Water treatment [28], Polymer fibers [29], Heavy metal absorption from water [30], Energy storage applications [31], DNA biosensor [32], and Tissue engineering, Cancer cell identification, Drug and gene delivery [33] . Due to these vast applications CNTs were synthesized by CVD method.

II. SYNTHESIS OF CARBON NANOTUBES - CHEMICAL VAPOUR DEPOSITION

CVD transmutes gaseous molecules (termed precursor) into a solid material, in the creation of thin film or residue, on the outward of a substrate.

CVD process was preferred because it is one of the usual approaches that compromises a auspicious track to bulk creation of great limpidness associated CNTs at low cost. It is the furthermost auspicious creation track for frugally creating huge amounts of CNTs [29] at low temperature and ambient pressure. Compared to other procedures the emergent parameters can be certainly organized.

A. Material Used

In this work, methane is used as the source of carbon. Ferric Nitrate is used as the catalyst precursors. The use of Ferric Nitrate catalyst is to give a higher carbon deposit. Zeolites are used as absorbents for catalysts. Since zeolites are leaky resources with smooth crystal structures and aperture measures, they are anticipating contrarily from further solid resources.

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Methane is elaborately obtainable, inexpensive and unwavering at upraised temperature. These are the causes that fascinated our attention in making CNTs via catalytic decomposition of methane.

B. Catalyst preparation

The materials used to prepare catalyst are Zeolite and Ferric nitrate. The catalyst is prepared with 2.5 wt% of Ferric nitrate in one gram of Zeolite. The Zeolite was finely ground and Ferric nitrate was added to it. The powdered mixture was immersed in 20 ml of ethanol and agitated for half an hour, gauged by smearing high frequency sound drive to agitate particles in a immersion for twenty minutes, up to homogeneous stain is formed. The slurry-strain was dehydrated at 353K immediately and crushed over into an acceptable residue.

C. Growth of Carbon nanotube

Generally, the procedure encompasses authorizing a organic vapor (typically for 15 minute to hour) via hose incinerator in which a catalyst solid is existing at satisfactorily extraordinary temperature (873 K – 1473 K) to decay the hydro-carbon. CNTs cultivate on the catalyst in the reactor, which are composed upon refrigerating the arrangement to room temperature.

Powdered catalyst (400mg) and (600mg) was dispersed into two alumina boats for the growth of CNT. The boat was introduced into the center of the furnace. Gas - fixed terminals were affixed which is coupled to the gas managing arrangement. The oven was excited up to (923K) at a rate of 288 $Kmin^{-1}$ in a rivulet of Argon gas at a rate of 95000 mm^3 min^{-1} and NH_3 gas at a rate of 5000 mm^3 min^{-1} for 1 hr. Again the oven temperature is boosted up to (1173K) at rate of 288 K with Argon gas streaming at a rate of 95000 mm^3 min^{-1} and NH_3 gas at a rate of 5000 mm^3min^{-1}, once the oven attained the response temperature, the gas stream was tainted to 5000 mm^3min^{-1} of methane. After forty minutes the gas course was distorted back to 95000 mm^3min^{-1} of Argon, and the setup is endurable to compose colder at room temperature. The sample were eliminated once the oven is refrigerated.

D. Characterization

The synthesized carbon nanotube powder samples were characterized by FESEM, Laser Raman spectroscopy, X- ray diffraction, and Thermo gravimetric analysis. The Field Emission Scanning Electron Microscopy (FESEM) is used to inspect the morphological structure of the sample. The Laser Raman Spectroscopy is used to examine the molecular vibrations of the sample. Thermo gravimetric curves represent the characteristic of a given material due to the sequence of physical transition and chemical reactions in a particular temperature range.

III. RESULTS AND DISCUSSION

3.1 Structure and Morphological Analysis

3.1.1 FESEM Micrographs Analysis

The morphology of the prepared carbon nanotube is examined by Field Emission Scanning Eletron Microscope (FESEM). Powder samples were dissolved in C2H5OH, a few dewdrops of the fusion is positioned on a Au-layered Si wafer and endorsed to dehydrate. The wafer was then glazed on a alloy sample frame using Ag as conductive coat [34] and tinny Au film glaze is conveyed to brand them conducting. CNTs are grownup using Zeolite vile at 2 dissimilar temperatures (1173K and 1123K) and with unlike drift rates (5, 15, and 20 SCCMs) and characterized using FESEM. The FESEM micrographs have revealed the evolution of CNTs with rarer produces.

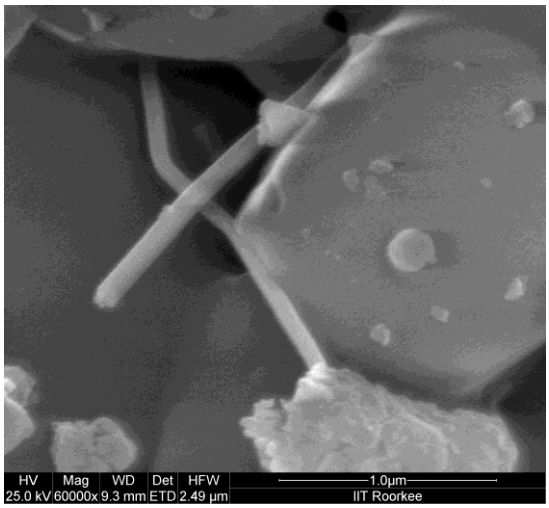

Fig- 3.1.1(a) Higher enlargement FESEM micro-graphs of MWNT acquired by CVD technique with Zeolite and Fe(No3)3 for 5 SCCM.

Fig- 3.1.1(a) shows the micro-assembly of CNT grown-up by consuming Zeolite infused Fe catalyst at 1123K and 5 SCCM of gas stream. The evolution of traditional CNT is grasped in the micro-graphs revealed in Fig- 3.1.1(a). The diameter of this CNT is witnessed to be 70 nm, and the length is 1.08µm. Long CNTs usually remain straight when they are in oriented structures.

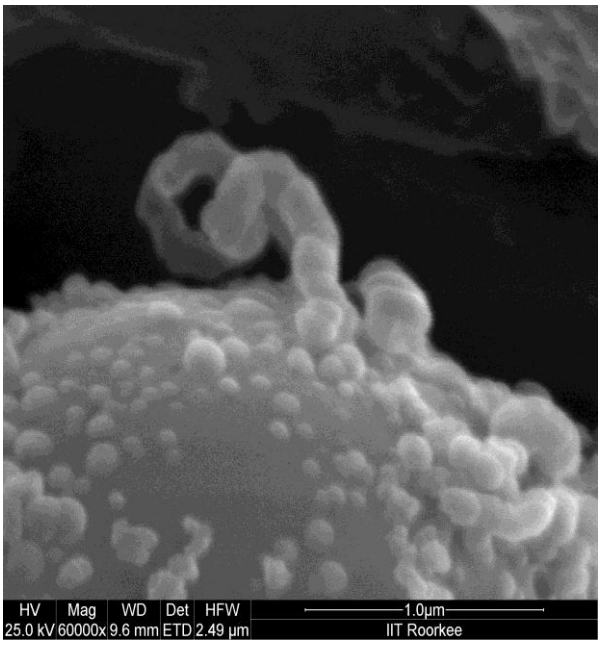

Fig-3.1.1(b) Higher magnification FESEM micrographs of MWNT obtained by CVD method with zeolite and iron nitrate for 15sccm.

The microstructure shown in Fig-3.1.1(b) represents, Zeolite impregnated Fe catalyst at 1173K temperature and 15SCCM of gas flow. Morphology of the above image is observed at 1.0µm. In the present condition also coiled carbon nanotube growth is observed. Coiled CNTs are MWCNTs with more or less incomplete crystalline structures. Since they grow out of the zeolite and maintain their self-organization during growth. Coiled CNTs having various diameters and pitches were observed.

3.1.2 Laser Raman spectroscopy

Laser Raman spectroscopy [35] is a NDT characterization method for the study of vibrational properties of CNTs. The positions, half widths, and the relative intensities of spectral bands are governed by the nature of chemical bonds of carbon. The sample is irradiated with He-Ne laser with the wavelength of 633 nm, double grating mono-chromator and cooled photo multiplier tube. Vibrational properties of the primed CNTs were explored by Raman spectroscopy.

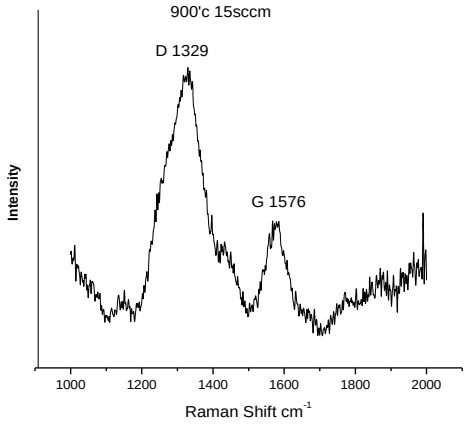

Fig- 3.1.2(a) Spectral Analysis of MWNT obtained by CVD method with zeolite and iron nitrate.

The spectrum shown in Fig- 3.1.2(a) represents, multi-walled carbon nanotubes synthesized at 900°C temperature. Disorder and Graphite band are observed in the spectrum. The D-band is observed at 1329cm-1 and the G-band is observed at 1576cm-1 and it is very well matched with the literature reported range [36]. Intensity and width of the Disorder band is greater compared to the graphite band. The integral ratio between the band intensity (ID/IG) is 2.10.

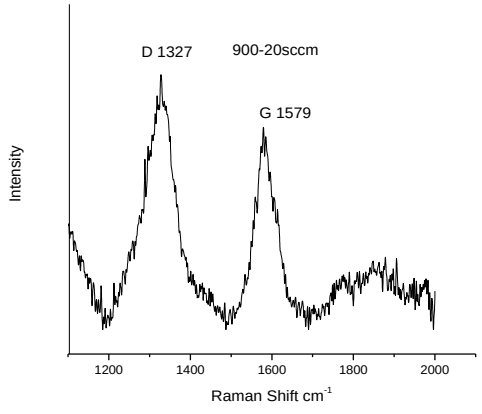

Fig- 3.1.2(b) Spectra report of MWNT obtained by CVD method with zeolite and iron nitrate for 900°C.

Fig-3.1.2(b) shows the Raman spectra of multi-walled carbon nanotubes synthesized at 900°C temperature. The D-band is observed at 1327cm-1 and the G-band is perceived at 1579 cm-1. The integral ratio between the intensities (ID/IG) is 1.25.

In Raman Spectra the absence of radial breathing mode at lower Raman shifts and only the D and G-bands indicate the presences of multi-walled carbon nanotube.

3.1.3 XRD Analysis

The as-synthesized CNTs were pigeonholed by consuming a Powder X-R-D system with CuKα radiation (1.541A°) used with a Ni filter to study its structural properties.

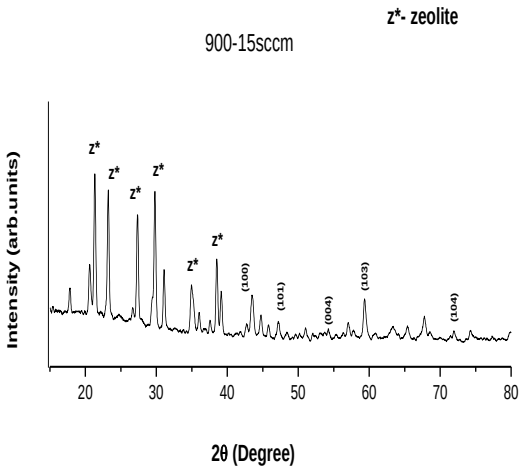

Fig- 3.1.3(a) XRD pattern for CNT obtained by CVD method with zeolite and iron nitrate with the gas flow of 15 sccm.

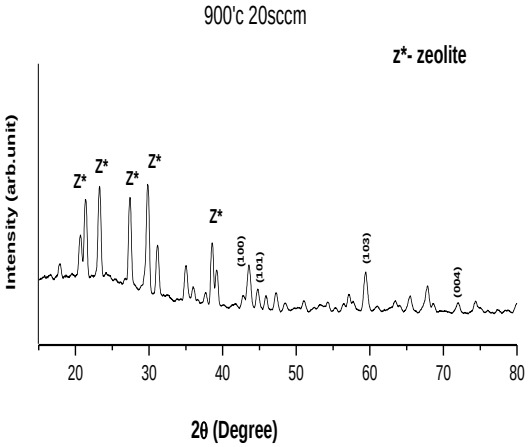

Fig- 3.1.3(b) XRD pattern for CNT obtained by CVD method with zeolite and iron nitrate with the gas flow of 20 sccm.

3.1.4 Thermo gravimetric Analysis investigations

Thermo gravimetric analysis is the most important technique to find the purity of the CNT. This involves heating the powder samples.

The different forms of carbon undergo oxidation and converted to CO or CO_2 at distinct temperatures.

The weight loss in the TGA curve gives information on the amount of different form of carbon and final residual weight correspond to the catalytic content in the CNT. The CNT sample synthesized at 1123K and 5 SCCM are subjected to TGA is shown in Fig- 3.1.4 (a). Around 20 mg of powder samples are taken in alumina crucible and this sample is heated up to 1073K with a warming proportion of 10 K/ min. However, due to the presence of large content of zeolite base the characteristic weight losses due to different forms of graphite were not observed. Rather a weight gain due to oxidation or oxygen pickup by zeolite was observed. There is no weight change is observed at 1173K temperature.

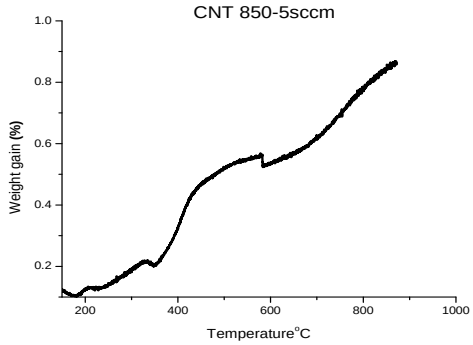

Fig- 3.1.4(a) TGA curve of CNT obtained by CVD method with zeolite and iron nitrate prepared at 850°C with gas flow of 5sccm.

IV. CONCLUSIONS

The surface morphology, vibrational and structural properties were studied by using FESEM, laser Raman, and XRD. FESEM micrograph shows the growth of carbon nanotube with different morphology. Length of the tube is ~1.08µm while its diameter is ~70nm. Raman spectra shows the absence of radial breathing mode (RBM) at lower Raman shift and only D and G- band evidence the presence of multi walled carbon nanotubes. XRD pattern reveals that, the intensity of graphite peaks are very subdued because the presence of higher content of zeolite in the specimen. It is observed that there is an increase in temperature and gas flow rate decrease the intensity of the graphite peaks. TGA curve shows, the as grown CNT has a weight gain due to oxidation or oxygen pickup by zeolite. The weight loss in the TGA curve gives information on the amount of different form of carbon and final residual weight correspond to the catalytic content in the CNT.

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