

R. Namitha¹, D. Radhika¹, Karthik Kannan², G. Krishnamurthy³

Manufacturing and Processing of Carbon Nanotubes for H₂ Storage

¹Jain-Deemed to be University, Jakkasandra, Ramnagara, Karnataka, India,
namitha19kkp@gmail.com, radhikadv8@gmail.com

²Kumoh National Institute of Technology, Gyeongbuk, Republic of Korea, karthikkannanphotochem@gmail.com

³Bangalore University, Jnanabharathi Campus, Bangalore, India, krisnamurthyg0@gmail.com

In pursuit of manufacturing of carbon nanotubes (CNTs) in good yield at lower temperatures, a mixture of d-block elements such as Iron, Nickel, and Cobalt was expected to be advantageous because of the high yield and low temperature (at 220 - 250 °C) synthesis. The physical state and aggregation of these catalyst particles in the reaction medium have been found to play an interesting role in obtaining CNTs at better yield. Carbon nanotubes have been successfully produced by an uncomplicated two-step solvothermal method between sodium and dichlorobenzene via Ni/Fe/Co as catalyst precursor. The dependence of yield of CNTs on the catalyst system was determined via different ratios of catalysts and at various other experimental conditions such as different heating temperatures, different durations of heating. The X-ray powder diffraction study has indicated the graphite kind of the products. Microscopic characterizations (SEM and TEM) implied us the diameters of carbon nanotubes are 10 - 14 nm. Raman spectroscopy shows the presence of graphitized carbon in carbon nanotubes. Significant influence by the heating temperature and heating duration has been observed on the product yield.

Keywords: CNT, solvothermal, Raman analysis, hydrogen storage.

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Introduction

A rigorous investigation has been aroused worldwide in the production of Carbon nanotube after their discovery by S. Iijima in 1991, due to their important applications. The distinctive size-dependent properties and small sizes of nanomaterials have led to the enormous potential for commercial applications which can get better human life and life-style through, for example, biomedical drugs and devices [1], food packaging industries and preservation, catalysis, energy conversion, electronics, information technologies, personal care products as well as environmental technologies [2-9]. Carbon nanotubes can also be used for storage of hydrogen [10], electrochemical and field emission devices, charge storage in supercapacitors, and nanotweezers [11], etc. Although several approaches of manufacturing CNTs [12, 13, and 14] have been developed, until now synthesizing CNTs under ambient conditions remained a great challenge.

Iijima synthesized CNTs in 1991 by arc discharge between graphite electrodes [15]. Yoshimura research group manufactured CNTs from polymers and many resources using hydrothermal circumstances at a temperature of 700 - 800 °C and 60 - 100 Mpa [16, 17]. Radhey Shyam Tiwari synthesized CNTs by CVD method by using Neem oil [18]. The Fan and co-workers used a nanopore alumina model of production methodology for manufacturing CNTs by using ethylene as carbon source [19]. Dingsheng Yuan and group synthesized capsule-like carbon tube via a facile solvothermal route [20]. CAO Chuanbao synthesized carbon nitride nanotubes by the Catalytic solvothermal route using NiCl₂ as a catalyst precursor at 230 °C [21]. Ezekiel D. Dikio, synthesized CNTs by using Ethyne with Cobalt-Zinc-Aluminium as Catalyst^[22]. Qian group explained a thermal reduction of the ethanol for the manufacturing of CNTs using Mg as catalyst at 600 °C [23]. However, the carbon nanotubes in the present effort were obtained with highly aligned and maximum amounts. Many processes generate through the

impression of getting sufficient carbon sources and gathering them into CNTs. Though several hard works have been prepared in the progress of manufacturing processes that includes different stages and high-priced resources, are inevitable.

In 2001, Lee and co-workers explained the manufacturing of non-porous carbon materials by using hexachlorobenzene and sodium [24]. In 2003, Xinhua Bao and researchers manufactured mesoporous carbon nanotubes through hexachlorobenzene and sodium at 230 °C [25]. Jiang [26] used solvothermal method to manufacture MWCNTs via the reduction of C_6Cl_6 or C_2Cl_4 in existence of K and Ni/Co catalysts. Z.F. Ren and researchers manufactured carbon nanotubes using the source polyoxyethylene (4) ether without any catalyst at 310 °C but a very less yield of about 20 % is reported [27]. In the current work, we have reported an enhanced method to manufacture carbon nanotubes at minimum of 220 °C and a maximum of 250 °C with a high yield.

By considering the above synthetic methods, general supervision for environmentally friendly methods has been provided, which results in controlling pollution, protecting the health and the planet, and moving towards sustainability. In this work, we convey a simple solvothermal method for the manufacturing of MWCNTs at 220–250 °C using Fe/Co/Ni as catalyst and dichlorobenzene is used as a carbon source.

It is to be renowned that numerous researchers have reported the synthesis of MWCNTs. In general, growing the multi-walled carbon nanotubes by using conventional synthesis methods at low temperature is a difficult task. However, interestingly, the carbon nanotubes were obtained by a low-temperature solvothermal method. This methodology for the manufacturing of MWCNTs would appear to be uncomplicated and cost-effective.

Growth of CNTs explained in the vapor-liquid-solid (VLS) theory was industrialized by Wagner and Ellis to explain Si whisker formation. As the replica proposes, the hydrocarbons are going to adsorb on the transition metal catalysts and undergo catalytic decomposition. This grade in the dissolution of carbon into the transition metal particles becoming a liquid eutectic. Upon supersaturation, carbon precipitates as tubular in the form of crystalline. However, the appropriate portrayal of growth likely depends on the synthesis methodology, catalysts, and circumstances used. We are reporting herein that, the catalytic performance of various transition metal mixtures in the production of CNTs by the solvothermal method. It is prompted us to determine the catalytic performance of 3d- transition metal ions for the production of CNTs.

I. Materials and methods

The Catalyst mixtures were prepared by dry impregnation method. To prepare a typical sample (0.1 g) metal ion mixture containing Fe, Co, and Ni in the ratio of 1: 1: 1. About 0.16g of iron chloride, 0.23 g of Nickel chloride, and 0.23 g of Cobalt chloride were mixed and ground well to get a homogeneous mixture of the metal ions. The material thus obtained was heated in an air oven at 120 °C for 20 h. The dried sample was

further ground well in a pestle and mortar to obtain a finepowder of metal ions mixture. The other metal powders prepared were (i) binary mixtures of metal ions such as Fe/Ni, Co/Ni, Fe/Co with 1:1 wt % ratio of each metal ions. (ii) A ternary mixture of ions such as Fe/Co/Ni with 1:1:1 wt % of the respective ions.

The solvothermal method was engaged in the manufacture of CNTs as described below. The typical method involves 2 stages. In the 1st stage, 1.0 g of Sodium and dehydrated catalyst were taken in 30 ml cyclohexane solvent were taken in a Parr reactor. The reaction mixture was kept in an autoclave at 220 °C for 6 h and cooled. In the 2nd stage, 2 ml of dichlorobenzene was combined as a carbon source into the reactor and it was occupied with cyclohexane to two-third of its capability. The reactor was dried again at 220 °C for 10 h and cooled. The outcome was collected after rinsed sequentially with cyclohexane, ethanol, dil. H_2SO_4 , and distilled water. As-obtained product was heated in an oven at 80 °C.

The manufactured and purified CNTs were analysed by XRD, which was executed on Powder X-ray diffractometer (Bruker D8). Raman characterizations were carried out using FT-Raman Spectroscopy to acquire details on graphite formation. The shape and structure observation of the products was investigated with SEM and TEM analysis. The electrochemical hydrogen storage studies were performed via an EG and G Potentiostat-Galvanostat mAAutolab with PGSTAT30 instrument with GPES software.

II. Results and discussion

The powder XRD study was employed since it is an important tool for the structural studies of CNTs. The given XRD pattern (Figure 1) of as-produced carbon nanotubes shows the purity of the product after the removal of the catalysts by post-treatment.

The crystalline nature of the established sample as deliberated by X-ray diffraction studies could be revealed by the patterns shown in Figure 1, which was obtained for carbon nanotubes before and after washing with acid. The peak at $2\theta = 20^\circ$ that relates to the (002) graphite

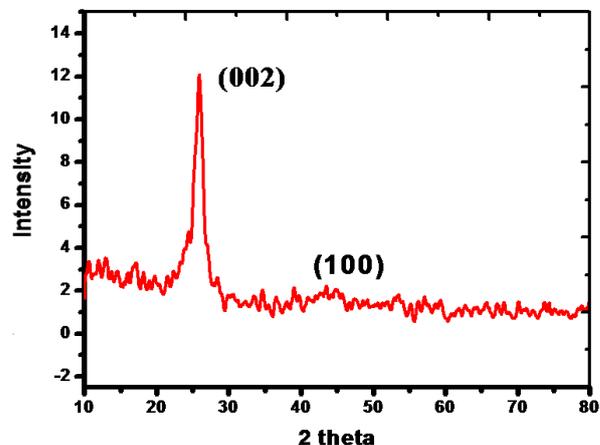


Fig. 1. X-Ray diffraction analysis of synthesized CNTs.

plane. It occurs owed to the interlayer assembling of graphene sheets. It specifies the concentric tubular structure of graphene layered collectively to form multiwall- CNTs. The X-ray diffraction analysis of all the manufactured samples at different conditions is of the same pattern as shown in Fig 1.

Raman spectroscopy (RS) is an immense method that gives us evidence on the occurrence of the disorder in the CNTs. Raman characterization also shows the eviction of the crystalline defects from carbon nanotubes.

In Figure 2, the Raman spectra give the peak at 1321 cm⁻¹ corresponds to D-Band and the peak at 1588 cm⁻¹ corresponds to G-Band [28, 29]. Figure 3, shows the RBM bands of produced carbon nanotubes which give us, the information about the diameter of the tubes obtained with peaks at 154.2 cm⁻¹, 196.63 cm⁻¹, 239.06 cm⁻¹, 293.06 cm⁻¹ and 325.8 cm⁻¹ shows the diameter of obtained carbon nanotubes of 0.88 nm, 1.2 nm, 1.37 nm, 1.69nm, 1.87 nm respectively.

Figure 4 shows the SEM images of the manufactured

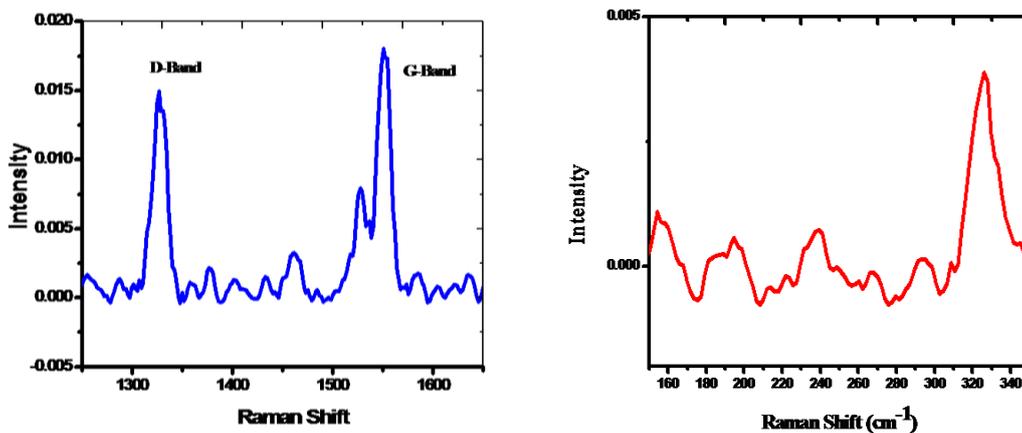


Fig. 2 & 3. Raman analyses of the manufactured CNTs and RBM bands.

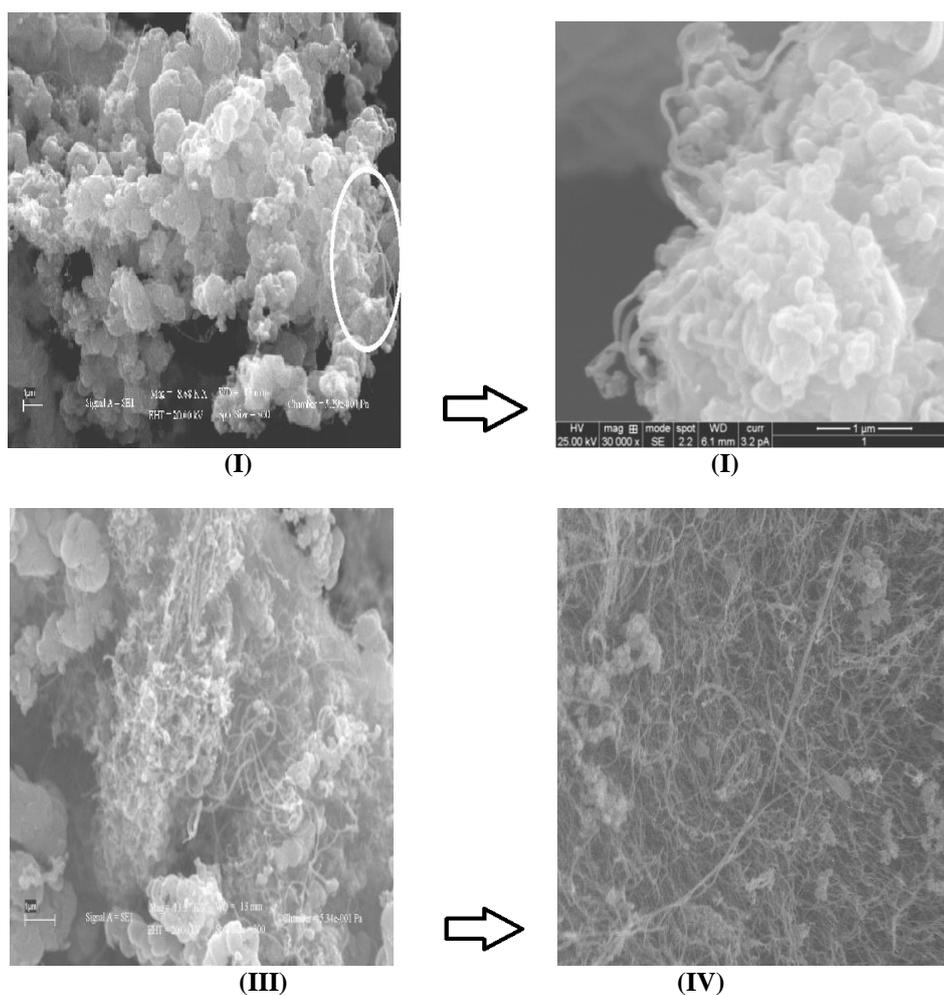


Fig. 4. (I), (II), (III) and (IV). SEM images of the manufactured carbon nanotubes at low and high temperatures at a different resolution.

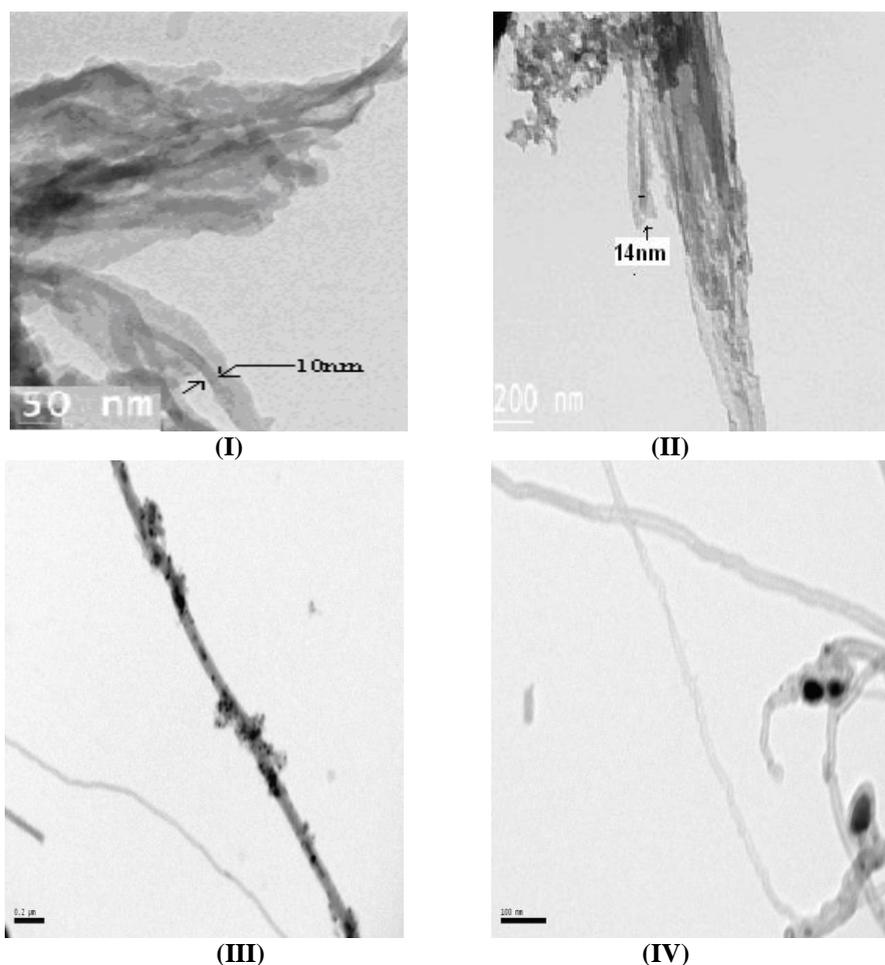


Fig. 5. TEM images of the manufactured carbon nanotube.

product. Image 4(i) contains the amorphous carbon like unstretched, short, and wounded in the forms of balls or lumps. This for the product synthesized at lower temperatures (around 200 °C) in which we can see the lesser formation of less percentage of carbon nanotubes. Image 4 (ii) shows the increased formation of carbon nanotubes at further elevated temperature (around 220 °C). Figure 4 (iii) shows the formation of CNTs at a higher temperature around 240 °C. They will have extended growth and stretched like tubular structures. Figure 4(iv) is the image taken after purification by treating with 5M HCl which contains maximum CNTs (98 %). The tube dimensions are confirmed by TEM images.

Figure 5(i), (ii), (iii) and (iv) comprises the TEM images for the product. The images in them show the presence of CNTs with some catalyst impurities deposited in them and the diameter of CNTs is found to be in the range of 10 - 14 nm.

Metal in the catalyst system shows a considerable role in processing the type of carbon nanotubes formed. In 2000, Resasco and co-workers showed that the ratio of Co and Mo was critical in manufacturing Single-walled nanotubes. In 2003, Liu et al and researchers had successfully produced 70 % Single-walled nanotubes and 30 % double-walled nanotubes over Fe-Mo/Al₂O₃ without the production of amorphous carbon. The addition of the particular catalyst system improves the

carbon capacity, persuades the type and morphology of carbon nanotubes formation, and improves the catalytic lifetime as well when compared with those of individual metals [30, 31].

Carbon nanotubes can be obtained on the particular diameter of metallic particles in the catalyst environment. The diameter and structure of CNTs and physical properties are dependent on the size and format of catalytic particles, curious growth of CNTs expect an adequate and portrayed method for catalytic preparation. Hence it is important to obtain an effective catalyst with the particular size of active Fe, Co, or Ni metal particles. These particles are catalytically very vigorous, because of the presence of d-block elements Iron, Cobalt, or Nickel and are free from being oxidized. Physical interaction between the substrate and the catalyst's material reduces the sintering which results in counteraction of the size of catalyst particle during the synthesis. Hence it is important to obtain catalysts with high metallic diffusion showing high particle solidity against the controlled growth of CNTs [32, 33].

Transition metals, importantly like Fe, Co, and Ni in the periodic table, are preferably used in catalytic decomposition of carbon source. It is known that compare to other metals, nickel is the most effective catalyst where it shows high catalytic activity at a lower temperature. In terms of carbon capacity, nickel is more active than iron at similar reaction conditions [34-39].

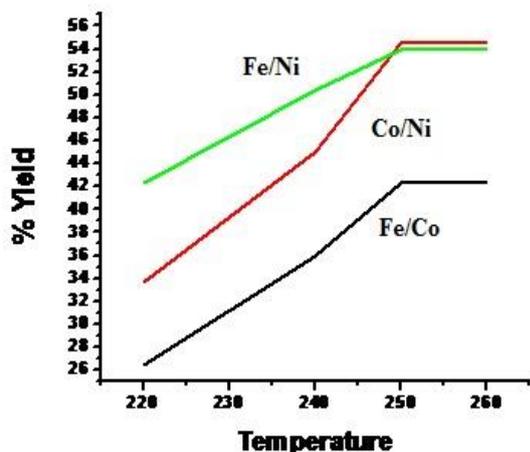


Fig. 6. Graph of the % yield of CNTs with different catalysts versus temperature at a different duration of time 10h and 20h.

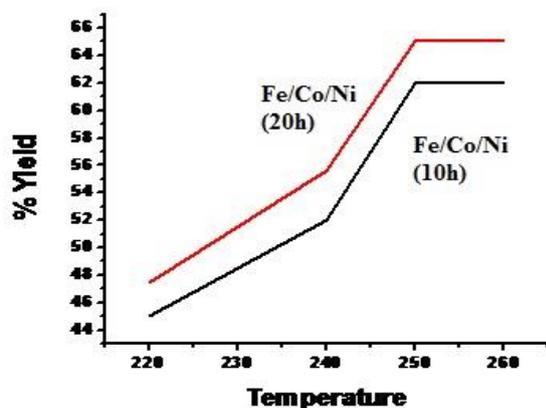


Fig. 7. The trend graph of % yield of CNTs with a mixture of Fe/Co/Ni catalyst as a function temperature and time.

Our results show from Figure 6, high yield of 54 % Carbon nanotubes are obtained with the mixture of Fe/Ni and Co/Ni as catalysts compared to Fe/Co which gives a yield of 42.3 %. In the case of a mixture of three catalysts, Fe/Co/Ni we got a yield of 65 % (Table 1), due to the presence of nickel which is highly active in presence of iron and cobalt mixture.

Reaction temperature has very much induced on catalyst lifetime, catalyst activity, and morphology of processing of carbon nanotubes obtained. As we know that various types of catalysts are active at various reaction temperatures. At high temperatures, it is considered that catalysts will appear in a quasi-liquid state where the catalyst particles are effortlessly cut into smaller particles. The smaller particles which are obtained easily encapsulated by the carbon layer produced during decomposition, causative to faster catalyst deactivation. On the other hand, at low temperatures, the catalyst vestige in a solid-state rather than in a quasi-liquid state and it maintains the activity of the catalysis process. Figure 7 indicates the increase in yield as we increase the temperature and at the different

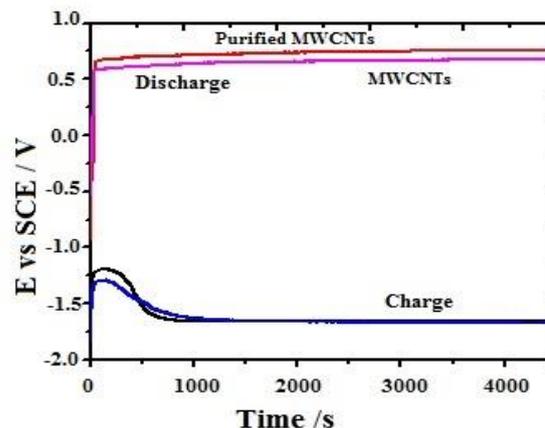


Fig. 8. The Charge-discharge curves for MWCNTs at 100 mA current.

catalytical conditions. It also represents the higher yield with Co/Ni catalyst at 250 °C and 20h duration.

It can be observed from the graph (Fig. 7), which shows the yield of samples at 250 °C is about 65 % synthesized at 20-hour duration. We can also observe the upsurge in yield with an upsurge in temperature. The yield of the MWCNTs manufactured has been achieved through the equation, $\text{yield } m (\%) = (m \text{ after}/m \text{ before}) \times 100$, where m before and m after are the product before and after purification [40-43]. The yield of products is in the range of 30 - 70 % for different catalysts. This indicates the effect of catalysts and temperature is a good yield as compare to other methods which are performed at low temperature.

In general, we have found that temperature influences the processing of the carbon nanotubes produced. The favorable condition for the manufacturing of carbon nanotubes involves the temperature range of 250°C and heating time of 20 hours in presence of mixture of Ni/ Fe/ Co catalyst in 1:1:1 ratio (Fig. 7). Besides, instead of using a mixture of three catalysts, we also used a mixture of two catalysts (Fig. 6) and could able to obtain CNTs in high yield. We found maximum yield with Fe/Ni and Co/Ni compare to Fe/Co catalysts.

During the process, the Cobalt chlorides, Nickel chlorides and Dichlorobenzene is also reduced by sodium metal via eliminating chlorine forming carbon bundles and NaCl. Due to the constant surface diffusion, when the nucleation of carbon nanotubes has completed, the carbon bundles disperse to the evolution spots and then the formation of nanotubes takes place. For MWCNTs, it is favorable that the existence of the outer wall alleviates the inner wall, maintaining it susceptible for incessant development.

The hydrogen storage behavior of MWCNTs was studied in an electrolyte. To obtain more detailed information on electrochemical responses measured using galvanostatic methods (Fig. 8). MWCNT electrode was charged and discharged using a scan pace of 10 to 100 mV s^{-1} . The voltammogram indicates that the reaction is governed by the diffusion process. The voltammogram suggests that the reduction peaks are due to the reduction of dissolved O_2 to HO_2^- and OH^- , respectively. This indicates that charge is stored in the

Table 1

Numerical data showing the Product yield for the experiments done at different conditions

S. No	Catalyst	Duration of heating (hours)	Yield (%) at T = 220 °C	Yield (%) at T = 240 °C	Yield (%) at T = 250 °C	Yield (%) at T = 260 °C
1.	Fe/Co	10	25.0	33.0	40.0	40.1
		20	26.5	36.0	42.3	42.3
2.	Co/Ni	10	30.0	43.0	52.0	52.0
		20	33.6	45.0	54.5	54.6
3.	Fe/Ni	10	40.0	48.0	52.2	52.3
		20	42.3	50.4	54.0	54.1
4.	Fe/Ni/Co	10	45.0	52.0	62.0	62.1
		20	47.2	55.6	65.0	65.0

material via a charge transfer reaction. It was observed in Figure 8, the maximum hydrogen adsorption of 800 mAh g⁻¹ was achieved which corresponds to 2.8 wt % of hydrogen [44-46].

Conclusion

Carbon Nanotubes have been productively manufactured and processed by uncomplicated solvothermal reactions at various crucial states were proposed at different temperature and time durations. The high yield of carbon nanotubes was obtained with a mixture of two catalysts of Co/Ni and Fe/Ni catalysts compare to Fe/Co and in the case of a mixture of three catalysts Fe/Co/Ni at 20 hours duration at a reportedly

lower temperature in the rating 200 - 250 °C.

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Namitha R. – PhD in Chemistry, Assistant professor of the Chemistry Department;
Radhika D. - PhD in Chemistry, Assistant professor of the Chemistry Department;
Karthik Kannan – PhD in Physics, Senior Researcher;
Krishnamurthy G. - PhD in Chemistry, Professor of the Chemistry Department.

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Р. Наміза¹, Д. Радхіка¹, К. Каннан², Г. Крішнамурті³

Виробництво та переробка вуглецевих нанотрубок для зберігання H₂

¹Університет Джайн, Джакасандра, округ Рамнагара, Індія, namitha19kkr@gmail.com; radhikadv8@gmail.com

²Національний технологічний інститут Кумо, Кьонгбук, Корея, karthikkannanphotochem@gmail.com

³Університет Бангалору, Бангалор, Індія, krisnamurthyg0@gmail.com

Для процесу ефективного виробництва вуглецевих нанотрубок (УНТ) за відносно низьких температур припускається, що цікавою є суміш d-елементів, таких як залізо, нікель та кобальт, через високий вихід та низьку температуру синтезу (220 - 250°C). Встановлено, що фізичний стан та агрегація таких частинок каталізатора в реакційному середовищі відіграє цікаву роль у отриманні УНТ з добрим виходом. Вуглецеві нанотрубки успішно отримано за допомогою неускладненого двоступеневого сольвотермічного методу між натрієм та дихлорбензолом за допомогою Ni / Fe / Co, як прекурсорів каталізатора. Залежність виходу УНТ від каталітичної системи визначали за різних співвідношень каталізаторів та за різних експериментальних умов, таких як температури нагрівання, тривалість нагрівання, тощо. Дослідження X-променевої порошкової дифракції вказали на наявність продуктів типу графіту. Мікроскопічні характеристики (SEM та TEM) показують, що діаметр вуглецевих нанотрубок становить 10 - 14 нм. Раманівська спектроскопія вказує наявність графітизованого вуглецю у вуглецевих нанотрубках. Спостерігається значний вплив температури нагрівання та часу нагрівання на вихід продукту.

Ключові слова: вуглецеві нанотрубки, сольвотермальний метод, Раманівський аналіз, накопичення водню.