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Research Article

Oxidizer gases effects on the diameter-controlled synthesis of carbon nanotubes

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Abstract

In this study, the influence of the oxidizers on the synthesis of carbon nanotubes by C_2H_4 decomposition over Fe catalyst has been investigated. CO_2, O_2, and H_2O have been used as oxidizers, and to control catalyst particle formation and their sizes in the pretreatment stage. The same oxidizers have also been used in the growth stage to maintain the catalyst particle size, remove amorphous carbon formation to keep catalyst particle active. The results of scanning electron microscopy indicated that the average diameters of nanotubes decreased from 13.4±1.2 nm to 6.2±0.5 nm and extremely dense nanotubes were obtained when we added a small amount of CO_2. Adding O_2 extremely decreased the areal carbon nanotube density while widens the diameter distribution. H_2O addition resulted in larger average diameters and made the growth strongly pretreatment dependent. Within the parameters tried for catalyst pretreatment and CNT growth processes, CO_2 seemed the best choice for a weak oxidizing assistant. The strong dependency of the average diameter on pretreatment conditions indicated that pretreatment is a very important step in deciding the final diameters and their distribution.

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1. Introduction

Carbon nanotubes (CNTs), due to their superior mechanical and physical properties [1], are undoubtedly one of the most promising nanomaterials for the growing number of applications in the nanotechnology era [2]. The distinctive characteristic properties of carbon nanotubes stem from the size of these materials and their atomic structure. Nanotubes can be either metals or semiconductors depending on their various helicity structure and diameters [3]. Also, the diameters of CNTs significantly affect their mechanical properties and thus influence their applications. Some of the applications to highly these growing areas include field emission device [4, 5], scanning probe [6-8], sensors [9, 10], high strength composite [11], and supercapacitors [12-14].

On the other hand, the growth process and controlling mechanisms of the CNTs are still not completely understood. Chemical Vapour Deposition (CVD) is the growth technique that allows control of the growth to a certain extent and it is scalable to industrial needs. All of the CVD techniques require catalyst nanoparticles for the growth of CNTs. The size of the catalyst particles which are usually chosen from transition metals plays an important role in determining the diameters of CNTs. These catalyst particles can be obtained using various techniques. Among these, the ones grown on a substrate [15] are crucially important since they will be building the bridge between microelectronics and nanotechnology. These very thin films (1-2 nm) will oxidize immediately once they are

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exposed to air till CNT growth. Most assume that the effective CNT growth proceeds after metal oxide to metal reduction. It is also known that metal oxide does not coalesce completely without the reduction.

For the reduction, various oxidizers [16-18] besides hydrogen [19-21] have recently been used to adjust the reaction rate and etch away the amorphous carbon deposition during the CNT growth [22, 23]. The reduction pretreatment step and its duration are significant in defining CNT characteristics. If the pretreatment time was kept too long, the catalyst particles would have wider size distribution and larger sizes [24] due to the high temperatures needed for growth. It would lead to nanofiber formation having larger diameters and inferior characteristics compare to CNTs. All these indicators point that the reduction pretreatment and the growth start timing are the deciding factors in CNT formation. Based on this concept, there are significant efforts to optimize CNT growth conditions.

Recent efforts have focused on the oxidizers as the growth enhancers and optimization of forest growth related to CNT yields and heights in the presence of weak oxidizers [25]. Even though those are very important to improve for industrial applications, their diameters and keeping diameter distribution tightly narrow usually have not been paid enough attention. The diameter is the most important parameter in CNT production since it will be deciding the CNT characteristics [26]. For the diameter control, the thickness of the catalyst film is a good starting point however with the process parameters the diameter distribution can readily go off track.

Effective CNT growth was studied in different studies [27,28] and these studies indicated that the presence of CO_2 caused a domed CNT growth, and the height of CNT distributes was more uniformly and converted to radial block figure. CNT structures such as dishes were obtained by increasing line heights with the increased amount of CO_2. Different CNT morphologies occurred with the increase of CO_2 amount because of the change of CNT growth velocity and change of bonding force between CNT and support material. Futuba et al. studied H_2O vapor, acetone, ethanol, metilbenzoat, and CO_2 was used to improve CNT growth by etching amorphous carbon in catalysts particle poisoned. Their results indicated that from these enhancers H_2O provided the best CNT growth atmosphere among these gases and the highest CNTs were obtained with H_2O. Carpet height CNT arrays at some samples terminated after one minute. Li et all. indicated that the heights of CNT grown with the addition of air continuous to not decrease till 500 min. Average CNT diameters were 35 nm close areas of buffer layer and 100 nm up of buffer layer were obtained. These diameter values showed that the structure of CNTs was closer to fiber than a cylindrical structure [29]. CO_2 oxygenizes the amorphous carbon in CNT synthesis process, and hence the catalyst stays active longer time, and therefore length and yield of CNTs increase. Besides, CO_2 can be decomposed on the catalyst clusters surface and reacts with other molecules and radicals, resulting in intermediates that can be more effective for the CNT growth. However, in this present study, oxidizers were utilized to control catalyst size and hence CNT diameters. Besides, diameter distributions were 2-3 nm in terms of standard deviation. This value was similar to the diameters distribution of SWNT obtained in the literature [30].

This study aims to analyze and determine the role of oxidizer gases during both catalyst pretreatment and CNT growth processes. The effects of the ratios of oxidizer gases in pretreatment to those in growth, time of oxidizer gas introduction in pretreatment process, and CNT growth temperature on CNT morphologies. CO_2, O_2, and H_2O were used in small amounts as oxidizers. The main aim was to utilize oxidizers to control catalyst particle size during the reducing pretreatment process and the growth process at the same time

preventing amorphous carbon deposition over the outer walls of CNTs and catalyst particles.

All as-grown samples were characterized by scanning electron microscopy (SEM), CNT diameters were measured and diameters distributions were obtained. CNT diameters within very narrow ranges were controlled. The growth with CO_2 yielded samples with better characteristics. Lower CNT density was observed by using O_2 compared to the other oxidizers. Using oxidizers both in the pretreatment and in the growth, stages helped to keep wider metal catalyst sizes and hence CNT diameters in narrow ranges which lead to effective CNT growth.

2. Experimental Section

In this study, Fe thin films were utilized as the catalyst material for CNT growth. $Fe/Al_2O_3/SiO_2$ Commercially purchased Si substrates were utilized as a support layer and cleaned in methanol by rinsing for 15 min in an ultrasonic bath and then in ultra-pure water for 15 min. Cleaned silicon substrates were oxidized by a dry thermal oxidation system. 10-15 nm thick Al2O3 and then 1.0-1.5 nm thick Fe layers were deposited on SiO2 substrate by DC magnetron sputtering technique. In the sputtering process, base pressure was 1×10^{-6} Torr prior to catalyst film growth while film growth was carried out at a deposition rate of 0.1 Ao/s, 20 W, and a growth pressure of 0.5 mTorr.

The thermal chemical vapor deposition (CVD) method was used for the CNT growth process. In a typical experiment, 4x4 mm2 $Fe/Al_2O_3/SiO_2$ thin film samples were put into a quartz boat and placed in the middle of the quartz tube and the CVD furnace was heated to 760° C. Experiments were carried out at atmospheric pressure. C_2H_4 was utilized as a carbon source and Ar was used as the carrier gas to carry the oxide gases to the reaction chamber. At a growth temperature of 760° C, gas flows were then switched to C_2H_4, oxide gases (CO_2, O_2, H_2O), and H2 for the desired growth time. The total gas flow was kept at 250 sccm at growth. Different gas flow rates and pretreatment times for oxidizers were investigated. The flow rates of other gases were constant in all experiments and 150 sccm for Ar gas, 140 sccm for H_2 gas, and 180 sccm for C_2H_4 gases. After growth, C_2H_4, H_2O, and H2 flows were turned off and CNTs were kept to cool down to ambient temperature under Ar flow with a flow rate of 150 sccm.

In order to analyze the morphology of CNTs scanning electron microscopy (SEM) was utilized. The quality and type of CNTs were examined with Raman Spectroscopy with He-Ne laser with the excitation wavelength of 632.8 nm.

3. Results and Discussion

One of the primary objectives of this study was to examine various oxidizers effects in the catalyst pretreatment and CNT growth stages. It has been suggested that “weak” oxidizers play a sanitary role by etching away amorphous carbon accumulation over catalyst particles which poison them and, thus, extending their life so that they can remain active longer for CNT growth [31]. This way highly efficient growth could be achieved. To do this, we studied four different pretreatment times and various amounts of CO_2, O_2, H_2O. We continued to send the smaller amount of oxidizers during growth time. Consequently, we reached narrow diameter distributions in the presence of various oxidizers. Standard deviations and standard errors of the average CNT diameters were analyzed and standard deviations of all samples were obtained in the range of 1-2 nm. The standard error of the average CNT diameter was found moderately small.

3.1. The role of CO_2 for effective CNT growth

In this study, influences of CO_2 were analyzed on a variety of growth conditions to control CNT diameter at a constant CNT growth temperature. SEM images were utilized to analyze CNT diameter and more than 70 tubes diameter were counted for each sample.

3.1.1 Catalyst pretreatment time effect

The effect of CO_2 in the CNT growth using C_2H_4 as a carbon source was studied by changing both pretreatment time and CO_2 level using the $Fe/Al_2O_3/SiO_2$ thin film as catalyst material and a smaller amount of CO_2 proceeded to be sent during CNT growth. Four different pretreatment times were tested in this part of the study and these were 2, 5, 10, 15 minutes. Besides, CO_2 flow rates were sent into the CVD system during both catalyst pretreatment and CNT growth as 10:1, 10:2, 5:1, 5:2, 2:1, and 2:2 sccm, respectively. The effects of CO_2 under growth conditions were investigated by SEM images of CNTs. The catalyst particle sizes were increasing with increased pretreatment time at the ratios of 10:1 and 5:1 gas flow.

This growth carried possibly out converting elemental form after the reduction of metal-oxide to metal film with H_2 and CO_2 controlled the reduction velocity of the metal oxide film.

Catalyst immediately clustered with the effect of high temperature on the elemental metal form and especially the SiO_2 layer. Also, these clusterings were active on the surface and occurred larger nanoparticles the results of collisions with others during these motions. Large nanoparticle influence negatively CNT growth, they decreased the density of CNT at the unit area. Here are significant amounts of H_2 and CO_2. The results showed that ideal particle sizes were obtained by adjusting both pretreatment time and CO_2 level (Table 1).

Once we carefully examined that the samples of CNTs exposed to shorter pretreatment time have more uniformity, smaller average diameters, and more density of growth. This supported our events that longer pretreatment time caused more growth of catalyst particles. Once we looked from the sides and anglely (Fig. 1), we observed that 15 and 10 min. samples include much more space than samples of 5 and 2 min. This state could be explained by decreased density of CNTs by forming bigger particles of the catalyst particle. When we calculated diameter, CNT diameters change among 5nm-8nm at short pretreatment time.

The dependence of CNT diameters on pretreatment time was analyzed in detail and the results were given in Fig. 2. A high concentration of CO_2 (10 sccm) during the pretreatment process and short pretreatment time resulted in larger CNT diameters without depending on CO_2 amount during growth time. When pretreatment time was increased at high amounts of CO_2, diameters of the CNTs grown under 2 sccm CO_2 were directly decreased. However, at growth time, diameters of CNTs grown under 1 sccm CO_2 were increasing because in the presence of high CO_2, reduction of metal oxide to metal was not completely achieved, therefore large nanoparticle clustered and CNT diameter got larger. CNT diameter decreased with pretreatment time because samples were grown at high CO_2 during pretreatment time and 2 sccm CO_2 during growth prevented sufficient oxygen and smaller catalyst particle growth occurred.

The studies with low CO_2 during pretreatment time supported that 2 sccm CO_2 flow rate provided smaller diameters than 1 sccm CO_2 flows during growth time.

Table 1 Catalyst pretreatment and CNT growth parameters and average CNT diameters in the presence of CO_2/H_2 for both different pretreatment times and growth times (CNT Growth temperature: 740 °C).

Sample	Pretreatment time (min.)	$CO_2(p)/CO_2(g)$ (sccm)	Ave. diameters (nm)
CNT1	15	10/1	12. 5
CNT2	10	10/1	10. 0
CNT3	5	10/1	9. 5
CNT4	2	10/1	8. 8
CNT5	15	10/2	6. 1
CNT6	10	10/2	6. 7
CNT7	5	10/2	6. 8
CNT8	2	10/2	11. 2
CNT9	15	5/1	9. 8
CNT10	10	5/1	9. 3
CNT11	5	5/1	8. 9
CNT12	2	5/1	6. 1
CNT13	15	5/2	5. 7
CNT14	10	5/2	6. 3
CNT15	5	5/2	7. 3
CNT16	2	5/2	7. 3

3.1.2 Temperature effect

The temperature effect in CO_2-assisted CVD was studied for four different pretreatment times at two different growth temperatures. CO_2 effects with a flow rate of 2 sccm at different growth temperatures were investigated. The results showed that average diameters were ranging between 15-29 nm at 740 °C and 5-8 nm at 760 °C (Table 2). High CNT growth temperature provided thinner nanotubes.

Carbon-oxygen reaction resulted in CO owing to catalyst particle metal oxide form [32]. If catalyst particle did not keep suitable pretreatment time and growth parameters, then catalyst surface is coated by amorphous carbon and CNT formation did not occur. The coming gas amount should be optimized and mixed with an appropriate amount of H_2.

It was expected that film coalesces by removing from the surface with the effect of temperature and hydrogen and catalyst nanoparticles properly spread the whole surface. Uniform and small particle size was necessary for efficient CNT growth.

The Fe particles were expected to diffuse uniformly on the whole surface and coalesce removing the surface with the effect of temperature and hydrogen. The temperature was more important in this situation. The coalescence was carried out after the metal oxide film

was converted to the metal film. During pretreatment, CO_2 provided controlled coalescences by controlling the speed of this translation. Also, CO_2 assisted to remove amorphous carbon that forms throughout CNT growth. Our results showed that particle size decreased with increasing temperature because more effectively reduced metal oxide form to metal form with increased temperature. At this state converter particle state from the film was easier.

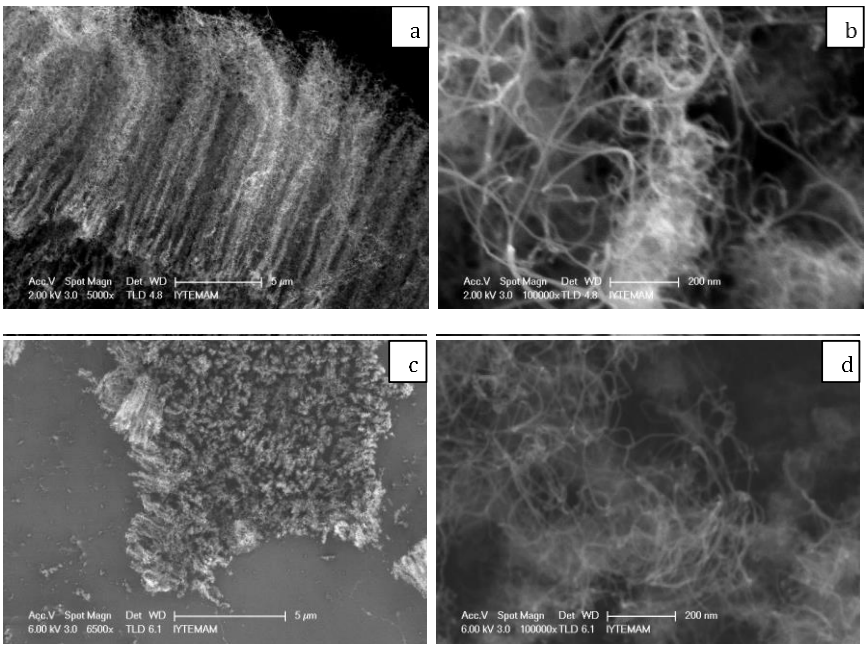
By using H_2 and CO_2, smaller CNT diameters and more density CNTs were obtained at high temperatures when compared to 740 and 760 °C (Figure3). CNT growth temperatures of 740 and 760 °C displayed similar behavior depending on pretreatment time. Temperature is also a significant parameter to determine CNT diameters, as well as CO_2.

According to SEM analysis results (Fig. 4) CNT density on the surface was rare and diameters change between 15-20 nm at 740 °C. However, at 760 °C, growth on the surface enhanced and the average diameter of CNTs decreased to the range of 5-6 nm at 760 °C.

Fig. 5 showed the relationship between average CNT diameter and pretreatment time for the growths carried out at 740 and 760°C. The results indicated that firstly, CNT diameters were increasing with increasing pretreatment time and then slowly decreasing for pretreatment time of 10:1 sccm. Average diameters of both short pretreatment time and CO_2 level of 1 sccm were smallest. At large parts of 1 sccm oxidizer, this identified that catalyst nanoparticle clustering through high temperature could not prevent more large catalyst particles to occur.

3.1.3 Effect of discontinuous catalyst pretreatment

CO2 effect on CNT growth with thermal CVD has investigated the states that the pretreatment duration of CO_2 was shorter than H_2. CO_2 flow rates were 10:1, 10:2, 5:2, 5:1, 2:2 ve 2/1 sccm and pretreatment time for H2 was studied 15 and 10 min., pretreatment time for CO_2 was studied for 5 and 2 min. (Table 3). Diameters chanced between 5-8 nm. Shorter CO_2 pretreatment time according to H_2 caused by keeping a narrow range of the diameters (Figure 6).



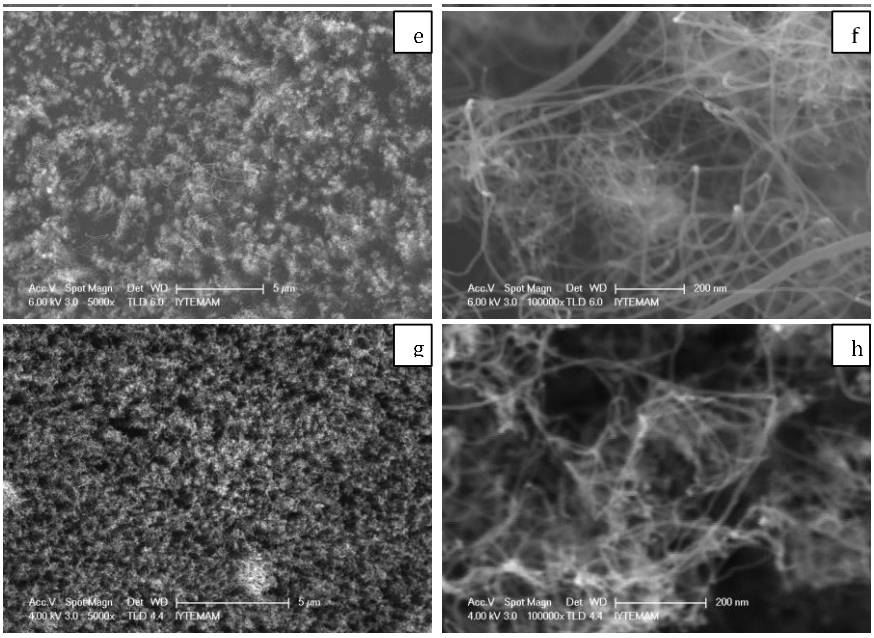


Fig. 1 SEM images of the nanotubes grown with CO2 as oxidizer at 740°C for different pretreatment times a), b) CNT1 15 min. , c), d) CNT2 10 min. , e), f) CNT3 5 min. , g), h) CNT4 2min

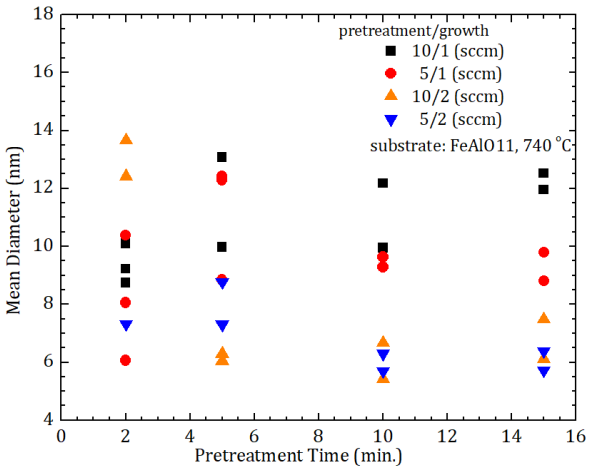


Fig. 2. Dependence of obtained average values of diameters on pretreatment time and CO_2 ratio

Table 1. Catalyst pretreatment and CNT growth parameters and average CNT diameters in the presence of CO_2/H_2 at different temperatures

Sample	Temperature (°C)	Pretreatment time (min.)	CO_2/H_2(sccm)	Ave. diameters(nm)
CNT17	740	15	2/140	20. 74
CNT18	740	10	2/140	18. 57
CNT19	740	5	2/140	17. 36
CNT20	740	2	2/140	21. 91
CNT21	760	15	2/140	6. 77
CNT22	760	10	2/140	5. 72
CNT23	760	5	2/140	5. 12
CNT24	760	2	2/140	6. 10

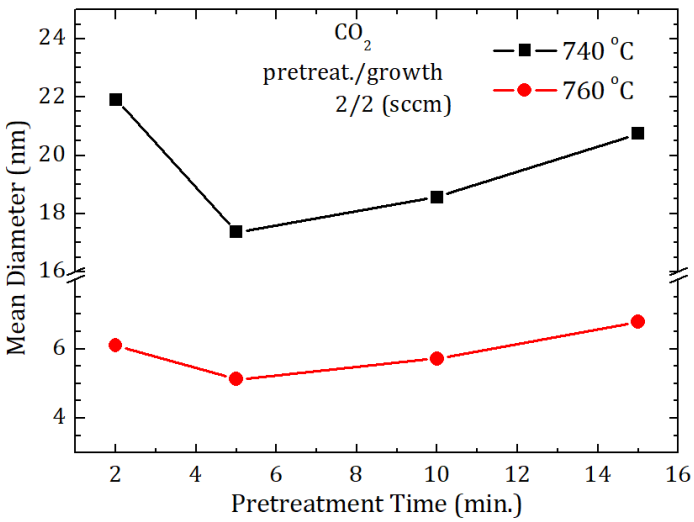


Fig. 3. Mean CNT diameters vs. catalyst pretreatment time for 740 and 760 °C, CO_2 flow rate: 2 sccm and 2 sccm during pretreatment and growth processes, respectively

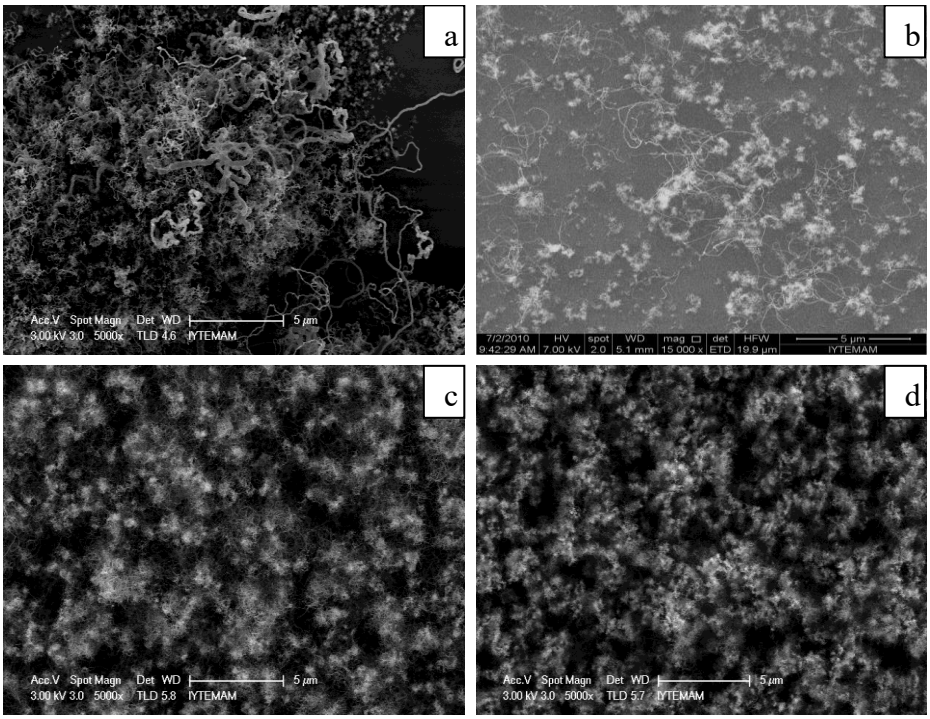


Fig. 4. SEM images of CNTs grown with CO_2, a) 740°C, CNT17, 15 min. b) 740°C, CNT18, 10 min. c) 760°C, CNT21, 15 min. d) 760°C, CNT23, 5 min

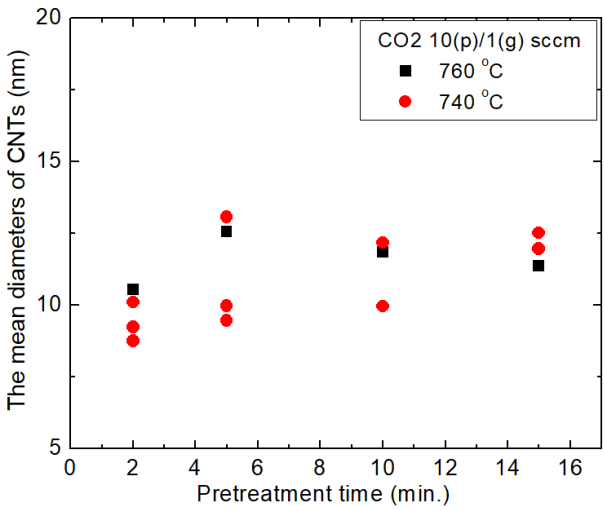


Fig. 5. Mean CNT diameter vs. catalyst pretreatment time, CO_2 flow rate: 10 sccm and 1 sccm during pretreatment and growth processes, respectively

Table 2. Catalyst pretreatment and CNT growth parameters and average CNT diameters in the presence of CO_2 +H_2 gases for different pretreatment time (CNT Growth temperature: 760 °C)

Sample	Pretreatment (min) H_2/CO_2	CO_2(p)/CO_2(g) (sccm)	Ave. diameters (nm)
CNT25	15/5	10/2	5. 96
CNT26	15/10	10/1	8. 07
CNT27	15/2	10/1	6. 60
CNT28	15/2	5/2	7. 02
CNT29	15/5	5/1	7. 50
CNT30	15/2	2/2	5. 45
CNT31	10/5	10/2	6. 82
CNT32	10/5	5/2	4. 69
CNT33	10/5	2/1	7. 29
CNT34	5/2	5/1	8. 90

3.2. The role of O_2 for effective CNT growth

The effect of O_2 for effective CNT growth with C_2H_4-CVD technique on the Fe/Al_2O_3/SiO_2 thin film was investigated for different pretreatment times and different O_2 flow rates. Examined pretreatment times were 15, 10, 5, 2 minutes. Studied O_2 flow rates during catalyst pretreatment and CNT growth were 5:1, 5:2, 2:2, 2:0.5, respectively. SEM images showed that Fe/Al_2O_3/SiO_2 thin film samples exposed to O_2 gas flow of 5:2, 5:1, 2:2, 2:0.5 sccm, resulted in larger CNT diameters while pretreatment time decreased. This growth carried out translating elemental form by removing the oxide via H_2 the catalyst being metal-oxide form. As a result, small CNT diameters ranging between 6-8 nm at the pretreatment time of 15 minutes were obtained in the presence of O_2. Average CNT diameters for pretreatment times of 10, 5, 2 min. were in the range of 9-14 nm (Table 4).

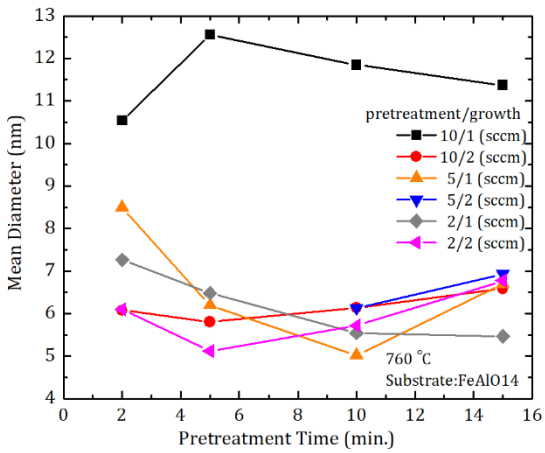


Fig. 6. Mean CNT diameter vs. catalyst pretreatment time for different pretreatment/growth CO_2 amount at growth temperature of 760 °C

Table 3. Catalyst pretreatment and CNT growth parameters in the presence of O_2 for different pretreatment times (P: 1 atm., T: 760 °C, Ar: 150 sccm H_2: 140 sccm).

Sample name	Pretreatment		CNT Growth		
	O_2 (sccm)	Time (min.)	C_2H_4 (sccm)	O_2 (sccm)	Time (min.)
CNT35	5	15	180	2	15
CNT36	5	10	180	2	15
CNT37	5	5	180	2	15
CNT38	5	2	180	2	15
CNT39	5	15	180	1	15
CNT40	5	10	180	1	15
CNT41	5	5	180	1	15
CNT42	5	2	180	1	15
CNT43	2	15	180	2	15
CNT44	2	10	180	2	15
CNT45	2	10	180	2	15
CNT46	2	15	180	0. 5	15
CNT47	2	5	180	0. 5	15
CNT48	0. 5	15	180	-	15
CNT49	-	15	180	-	15

3.3. The role of H_2O vapor for effective CNT growth

A small amount of H_2O vapor in the CNT growth process reduces amorphous carbon amounts and provided more effective production [32]. H_2O extends catalyst lifetime during pretreatment and therefore assisting efficient CNT growth.

In this part of the study, under the same parameters, the influences of H_2O vapor for effective CNT growth on the $Fe/Al_2O_3/SiO_2$ thin film were investigated. The effect of H_2O on CNT growth was examined. Pretreatment time was kept constant (15 min) and different H2O vapor levels were tested. To send H_2O vapor, a new apparatus was added to the system (Figure 7). A expressed a little amount of Ar of H_2O vapor was sent during pretreatment time. B expressed much more H_2O and Ar than A, namely B was more diluted than A. Studied H_2O vapour amounts were 50°C(A)/50°C(A), 50°C(A)/50°C(B), 50°C(B)/50°C(B), 50°C(B)/50°C(A), 60°C(A)/60°C(A), 60°C(A)/60°C(B), 60°C(B)/60°C(B), 60°C(B)/60°C(A), 70°C(A)/70°C(A), 70°C(A)/70°C(B), 70°C(B)/70°C(B), 70°C(B)/70°C(A). The results of the H2O vapor study indicated that the optimum level was 60°C(A)/60°C(A). Dense CNT growth and narrow average diameters were obtained as the samples grown with CO_2 (Table 5).

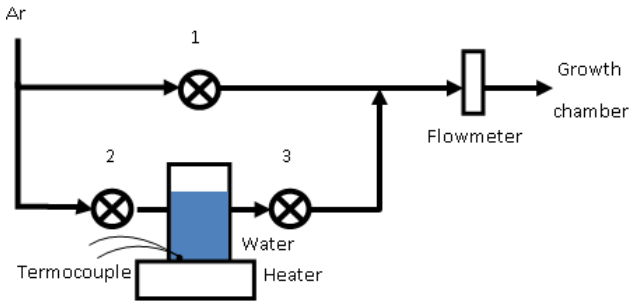


Fig. 7. The setup integrated to send H_2O vapour in the different amounts A: valve1 off, valve2 ve valve3 on, B: all valves on, C: valve1 on, valve2 ve valve3 off

Table 4. Catalyst pretreatment and CNT growth parameters and average CNT diameters in the presence of H_2O vapor (CNT growth temperature: 760 °C)

Sample Name	Pretreatment (min.)	H_2O(p)/ H_2O(g) (°C)	Ave. diameter (nm)
CNT50	15	60^0C(A)/60^0C(A)	7. 01
CNT51	15	60^0C(A)/60^0C(B)	7. 47
CNT52	15	60^0C(B)/60^0C(A)	8. 04
CNT53	15	60^0C(B)/60^0C(B)	6. 80
CNT54	15	50^0C(A)/50^0C(A)	7. 60
CNT55	15	50^0C(A)/50^0C(B)	9. 11

3.4. Raman Analysis of CNTs

Raman spectroscopy is an important characterization method to analyze the type and quality of the CNTs. In the Raman spectrum, the G-peak of the CNT sample is a characteristic feature of the graphitic layers placed at ~1580 cm^{-1}. The other characteristic CNT peak is D-peak, an indicative peak of graphitic structure defects such as amorphous carbon, soot and additional carbon structures placed at ~1335 cm^{-1}. The ratios of intensities of D/G peaks represent the quality of CNT samples. Representative Raman spectrum of a CNT sample grown under 760 °C growth temperature by utilizing CO^2 as an oxidizer with pretreatment/growth: 5/1(sccm) flow rates is given in Fig. 8. The high D peak intensity of this sample indicated defects presence in the obtained CNTs structures, I_D/I_G was calculated as 0.68 for this sample indicating a significant level of defective carbonous products. A third peak named the radial breathing mode (RBM) being very sensitive to the CNT diameter, giving information about CNT type and placed at between 100–300 cm^{-1}.

RBM peaks were not observed for the CNTs grown by utilizing CO_2 as an oxidizer and it means this sample includes multi walled CNTs. CNTs exhibit metallic or semiconducting characteristics depending on their chirality (how the graphitic sheets are rolled) [33] and the splitting of G band to G- and G+ bands (tangential modes) in Raman spectroscopy highly reveal semiconducting or metallic properties of CNTs [34]. As seen in Fig. 8, G-band did not split G- and G+ bands for the sample grown with CO_2 as oxidizer indicating the abundance of metallic multi walled CNTs.

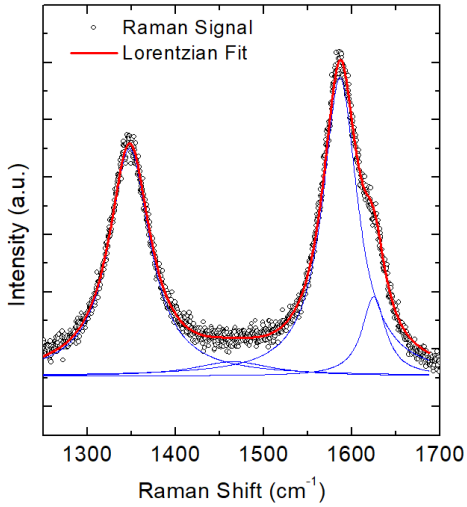


Fig. 8. Raman spectrum of CNT grown under 760 °C growth temperature by utilizing CO_2 as oxidizer with pretreatment/growth: 5/1(sccm) flow rates

4. Conclusions

In this study, the diameter-controlled synthesis of CNTs under different oxidizer gases was discussed. Systematic ways of obtaining controlled nanoparticles from catalyst film to grow the effective CNT were presented with oxidizer gases. Utilized oxidizer gases were CO_2, O_2, and H_2O vapor. Among these oxidizers, the best results were obtained by using CO_2 which provided dense CNT growth and narrow CNT diameter range.

Pretreatment time was another parameter affecting CNT quality and diameter. CNTs exposed to a shorter pretreatment time deposited uniformly and had smaller average diameters and showed more density of CNT. Longer pretreatment time caused bigger catalyst particles resulted with larger CNT diameter.

740 and 760 °C growth temperature of CNT was examined to determine the effect of temperature on CNT diameter and average diameters were significantly decreased for higher temperature.

Overall, obtained CNTs in this study were multi walled CNTs in the diameter range of 5-22 nm. By using oxidizers, a narrow range of metal catalyst sizes and thus narrow diameters of CNTs were maintained. Therefore, diameter-controlled effective production of CNTs was provided. The ability to control over diameters of CNTs makes easier to understand the fundamental properties of CNTs and develop new CNT applications.

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