Full Length Research Paper

Optimization of Carbon Nano Tubes Synthesis using Fluidized bed Chemical Vapor Deposition: A Statistical Approach

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Fluidized Bed Chemical Vapor Deposition (FBCVD) has been introduced as a promising method for carbon nanotubes (CNTs) synthesis because of its large scale, low cost and high yield production. However, there is no clear relation between synthesis parameters and CNTs growth; therefore more data are required on FBCVD synthesis of CNTs. This research intended to investigate the effects of some synthesis parameters namely reaction temperature, catalyst loading and deposition time on FBCVD growth of CNTs. In present study, CNTs were synthesized through decomposition of acetone over prepared catalysts which are Iron and Molybdenum supported on Alumina. After each run the product was characterized using Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), Thermo Gravimetric Analysis (TGA) and Energy Dispersive X-ray spectroscopy (EDX). The effects of parameters on carbon deposition yield were statistically studied using analysis of variance (ANOVA). The optimum quality and yield of the CNTs were achieved at 750 °C reaction temperature, 40min of deposition time and utilizing 5 gm of catalysts loading.

1. INTRODUCTION

In 1970, Baker et al. observed carbon nanotubes (CNTs) formation during synthesizing carbon fiber in the presence of metal catalyst at high temperature (Baker and Barber, 1972, Baker and Harris, 1973). However, compared to CNTs produced some years later by Ijima, their report was not well received. Later in 1991, Sumio Ijima first detected multi-walled carbon nanotubes (MWNTs) in his studies by Transmission Electron Microscopy (TEM). Two years later, Ijima and Bethune synthesized the first single-walled nanotubes (SWNTs) (Iijima, 1991, Dai, 2001, Dresselhaus Endo, 2001). Depending on the arrangement of carbon atoms, CNTs can be used for wide range of applications. CNTs are present as a one-dimensional novel form of fullerenes. CNTs are constructed from sp2 orbital hybridization of carbon atoms, with a few nanometers in diagonal diameter and many microns in lengths. In this structure, carbon atoms are covalently bonded to each other. Figure.1 shows a macrograph of CNTs in the laboratory.

Figure 1. Macrograph of CNTs

CNTs have unique electrical, mechanical, optical, and thermal properties. These have made them attractive substance for may advance applications. Some potential application of CNTs are in electronic devices (transistors, wires, interconnects) (Collins and Zettl, 1997, Collins Avouris, 2000), sensors and probes (Varghese and Kichambre, 2001), field emission materials (Tans and Verschueren, 1998) , batteries/fuel cells, fibers, reinforced composites, medical and biological applications, and hydrogen storage (Journet and Maser, 1997). Recently, various techniques have been developed to produce CNTs. The chemical vapor deposition in fluidized bed appears as a promising method for CNTs synthesis due to its large scale, low cost and high yield production (See Harris, 2007, Shanov and Yun, 2006, Hu and Dong, 2013).

Fluidized bed chemical vapor deposition (FBCVD) method is capable of controlling growth condition and synthesizing a large quantity of CNTs. In a fluidized bed reactor, uniform temperatures within the bed and rapid gas-solid interactions are allowed because of suitable heat and mass transfer. In addition, continuous operation is possible (Venegoni and Serp, 2002, Perez-Cabero and Rodriguez-Ramos, 2003, Son and Lee, 2007, Wang and Wei, 2002, Corrias and Caussat, 2003, Lee and Chang, 2011). However, there are still many problems such as undifined synthesis parameters that calls for more research. See et al reported that there is no clear relation between CNTs growth and synthesis parameters such as reaction temperature, deposition time and catalyst loading (See and Dunens, 2008). Therefore, more research is needed to gain better control over synthesis of CNTs by FBCVD. Further investigation may results in large scale and low cost production of CNTs.

With respect to the above disscusion, this work describes the synthesis of carbon nanotubes through decomposition of acetone over the prepared catalyst (i-e Fe-Mo/AL$_2$O$_3$) in a fluidized bed reactor. Alumina or α-Al$_2$O$_3$ which is known as corundum has been widely investigated because of its important applications in advanced engineering as catalyst support (Kakooei and Rouhi, 2012). To do this, a parametric study have been done to investige the effects of reaction temperature, catalyst loading, and deposition time on CNTs growth.

Analytical equipments including scanning electron microscope (SEM), transmission electron microscopy (TEM), and thermo gravimetric analysis (TGA) were utilized to analyze and characterize the fabricated CNTs. Previously, the effect of deposition time on carbon and CNTs yield was investigated (Hanaei and Fakhru'l-razi, 2012). It was shown that deposition time plays an important role in CNTs growth (Kouravelou Sotirchos, 2005, Danafar and Fakhru'l-Razi, 2009, Cadek and Murphy, 2002). Temperature is well known as most important parameter in synthesis of CNTs. It was found that the starting reaction temperature for FBCVD is 550°C and no CNTs were observed below 550°C (See Harris, 2007) There is not a confirmed maximum temperature for CNT production (Venegoni and Serp, 2002, See Harris, 2007, Muataz and Ahmadun, 2006, Morançais and Caussat, 2007). This paper is organised as below.

2. MATERIALS AND METHODS

There are two steps in FBCVD technique for synthesis of CNT which are catalyst preparation and actual reaction. These steps are discussed in the following sections.

2.1. Catalyst preparation

Generally, catalysts should be prepared on a substrate. Therefore, in this study Al$_2$O$_3$ was employed as the substrate (Kakooei and Rouhi, 2012) and Fe-Mo was selected as the catalyst. The right amount of alumina powder was added to a solution of iron nitrate (Fe (NO$_3$)$_3$•9H$_2$O) and ammonium molybdate (NH$_4$)$_6$Mo$_7$O$_{24}$•4H$_2$O. The weight ratio of Iron to molybdenum to alumina (Fe: MO: Al$_2$O$_3$) was 9: 1: 5 (Qian and Yu, 2002)

2.2. Carbon nanotubes synthesis

The experimental apparatus for nanotubes growth is presented in Figure2. The main body of the reactor was a vertical 316 stainless steel cylinder. Its dimensions were 53mm internal diameter and 1000mm in height enclosed by an electrical furnace. The gas distributor was a stainless mesh, which supports the weight of the solids before they suspended in a fluid flow.
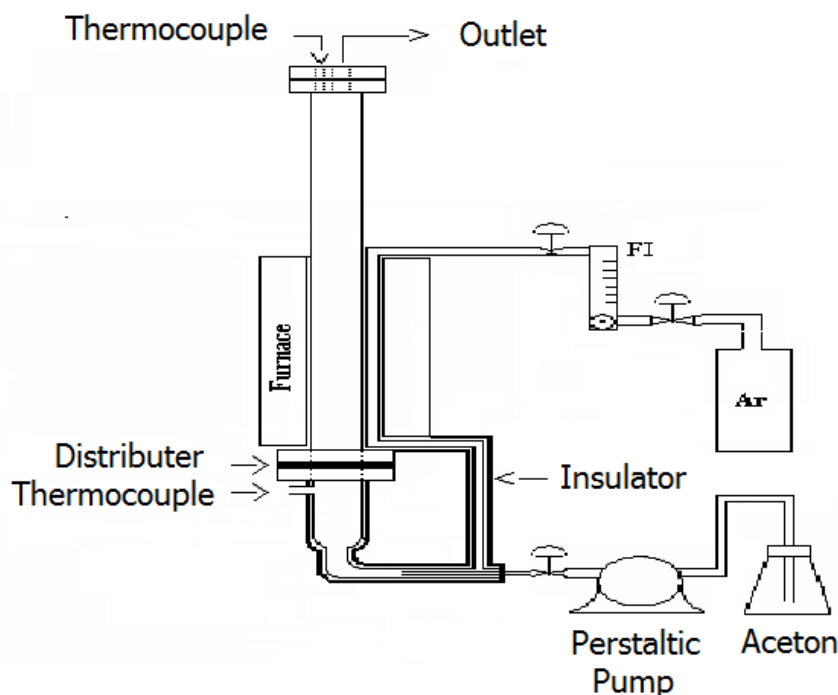
Figure 2. Schematic diagram of FBCVD setup (Fakhru l-Razi and Danafar, 2009)

Catalyst was placed into the reactor and argon gas with 1.4 l/min ratio was introduced into the bottom vessel of the reactor. Then it passed through the gas distributor, and finally flowed out into the atmosphere. When the fluidized-bed reached the desired temperature, acetone vapor which was preheated with the argon, flowed in to the reactor. Reaction occurs within the catalyst particle that acts as the sites to promote CNT growth. Both the catalyst and CNT were smoothly fluidized according to specific intervals of 30 min, 40 min and 50 min. As a result, the acetone was decomposed over the catalyst to form CNTs. At the end of each run, the electric furnace was turned off and argon flow was used to cool the reactor to room temperature. After reaction, the morphology and microstructure of the CNTs were observed using SEM. The exact amount of carbon deposit was determined by weighing the catalyst before and after reaction.

Based on the amount of deposited carbon, carbon yield during the reaction was obtained from the following formula:

$$\text{Carbon Yield (\%)} = [(m_{tot} - m_{cat})/m_{cat}] * 100$$

where m_{cat} and m_{tot} are the initial amount of the catalyst before reaction and the total weight of the product after reaction, respectively (Hernadi and Fonseca, 1996)

The as-synthesized products were also analyzed using TEM (Hitachi H-7100) and TGA (STDA851 METTLER). For TGA analysis, each sample was heated from room temperature to 1000°C at a 10°C/min heating rate. Sample burnt off until all carbon was oxidized and merely metal oxide remained (Shajahan and Mo, 2003). The purity and quality of the CNTs can be determined by TGA results.

2.3. Statistical Analysis

Three-way analysis of variance (ANOVA) was used to determine the difference between samples, using SPSS software. Duncan test and Hsu's test was used to find the difference of means between pairs. ANOVA was employed to determine the best level of sample with P value less than 0.05. A full factorial design showed the effect of third order interaction. As a result of employing the full factorial design, 45 runs were conducted randomly. Table 1 describes the experiments designed by full factorial in this study.

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.Table 1. Full Factorial Design Table Investigated in This Study

A	-2	-2	-2	-2	-2	-2	-2	-2	-2
B	-1	-1	-1	0	0	0	+1	+1	+1
C	-1	0	+1	-1	0	+1	-1	0	+1
A	-1	-1	-1	-1	-1	-1	-1	-1	-1
B	-1	-1	-1	0	0	0	+1	+1	+1
C	-1	0	+1	-1	0	+1	-1	0	+1
A	0	0	0	0	0	0	0	0	0
B	-1	-1	-1	0	0	0	+1	+1	+1
C	-1	0	+1	-1	0	+1	-1	0	+1
A	+1	+1	+1	+1	+1	+1	+1	+1	+1
B	-1	-1	-1	0	0	0	+1	+1	+1
C	-1	0	+1	-1	0	+1	-1	0	+1
A	+2	+2	+2	+2	+2	+2	+2	+2	+2
B	-1	-1	-1	0	0	0	+1	+1	+1
C	-1	0	+1	-1	0	+1	-1	0	+1

| Variables	ID	-2	-1	0	+1	+2
Reaction Temperature (°C)	A	550	650	750	850	950
Deposition time (min)	B		30	40	50	
Catalyst loading (gr)	C		5	10	15	

3. RESULTS AND DISCUSSION

In this work, we investigated the effects of three influential parameters which are synthesis temperature, catalyst loading and deposition time. Moreover, their interactions on the resulting carbon yield were also discussed. To differentiate carbon yield from CNT yield, TGA method, described by See and Harris, was used (See and Dunens, 2008) This method involves deconvoluting the TGA weight loss profile into three general groups: (i) amorphous carbon, (ii) CNT and (iii) carbon microtube. The SEM and TEM images of synthesized CNTs for selected runs are displayed in Figure 3.

SEM analysis was used to qualitatively characterize the carbon deposits formed over the metal particles. Results showed that the quality and purity of produced CNTs at 750°C, 40 min, 5 gr is better than other samples as shown in Figure 3a. The SEM images in Figure 3b and c indicates that the CNTs formation decreased in 950°C. This reduction in CNTs formation is regardless of the various deposition times. SEM images showed that the products at 950°C were mostly composed of amorphous carbon and microtubes. Despite the fact that in 950°C the carbon yield percentage increases, the quality of synthesized CNTs was not improved.
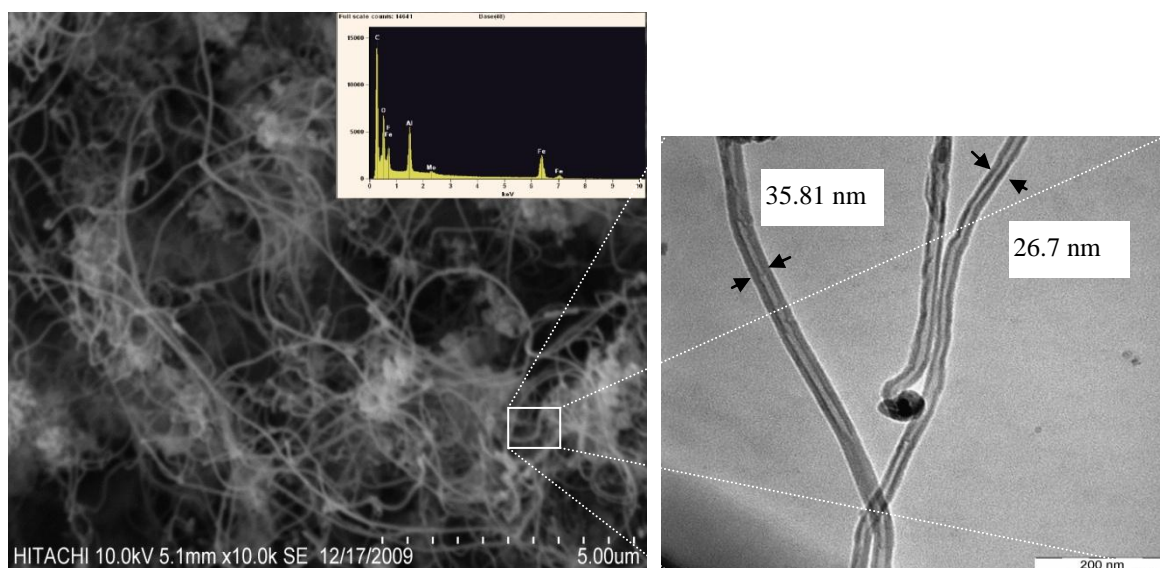
(a)
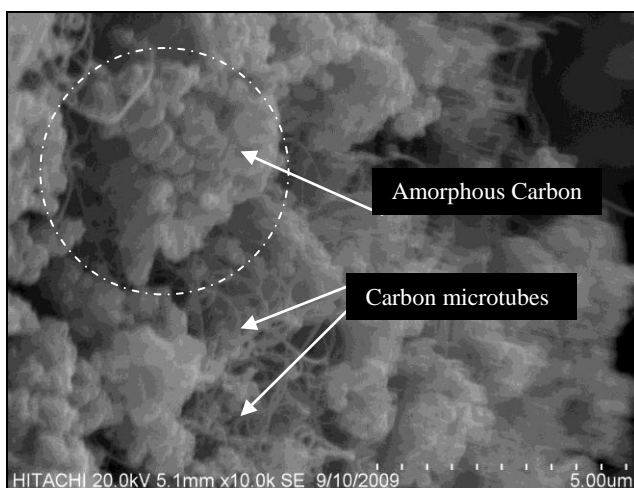
b
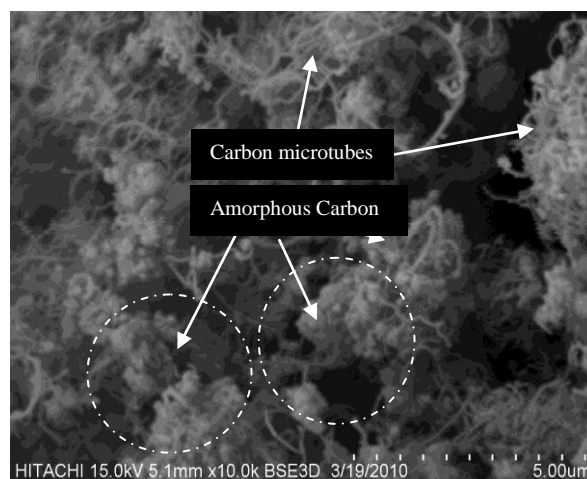
c

Figure 3.a) SEM and TEM image of CNT at (750°C, 40min, 5gr), SEM image of CNT **b)** at (950°C, 40min, 5gr) and **c)** at

(750°C, 50min, 5gr)

The SEM and TEM images depicted that the products in lower than 750°C were composed of amorphous carbon. The EDX graphs of synthesized nanotubes revealed that large amount of carbon were surrounded by iron particles as shown in Figure 3a. TEM studies also indicated good quality products. A typical individual CNTs with corresponding diameters are shown in Figure 3a. Consequently, it could be concluded that the combination of 750°C, 40min, 5gr is the best condition for synthesis of CNTs in prospective of high yield and good quality of the products.

TGA plots were used to differentiate carbon yield from CNT yield. See et al. reported that the oxidation of CNTs occurs in temperature range of 420°C to 620°C (See and Dunens, 2008). Kitiyanan et al. and Tang et al. observations showed that amorphous carbon oxidation temperature was around 330°C (Kitiyanan and Alvarez, 2000, Tang and Zhong, 2001). In order to measure the oxidation temperature of CNTs, 10 samples with higher yield of carbon were selected by SEM analysis. TGA plots for selected samples are shown in Figure 4. In the graph, effective parameters which are temperature, time and catalyst weight are separated by comma, respectively. CNT yield was defined by TGA weight loss from 420 to 620 °C.
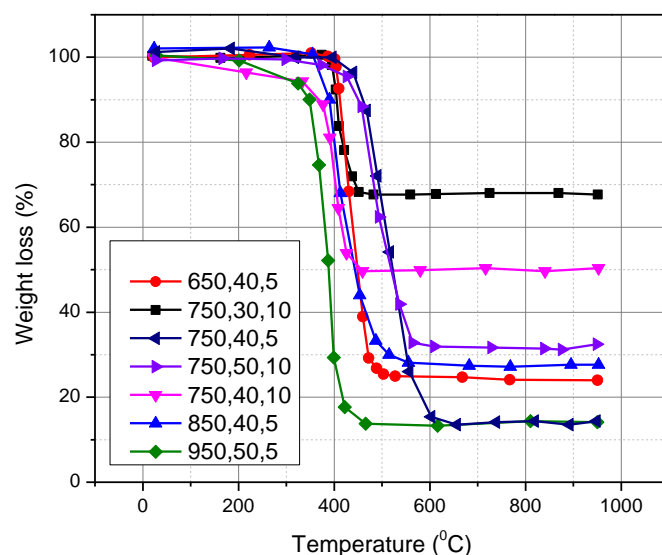
Figure 4.TGA graph of some selected samples.

It can be seen that the TGA graph at 750°C, 40min, and 5gr had more weight loss (low residual mass) than other samples. The oxidization temperature was between 500°C to 600°C which was in good agreement with See et al. reports. This result indicates that the CNTs formation under above condition (750°C, 40min, and 5gr) is almost high. Present findings were also confirmed with previous observations. For instance, Niu (Niu Fang, 2008) reported that the optimum temperature of synthesizing CNTs via FBCVD over Mo-Co-MgO bimetallic is about 727°C. Also, Venegoni (Venegoni and Serp, 2002) remarked the process operates is between 550 and 750°C. At higher temperature deposition yield of CNTs decrease, due to catalyst particles sintering. It notes worthy that residual mass after reaction is a combination of catalyst particles such as Fe and Mo.

### 3.1. Influence of Process Variables on Carbon and CNT Yield

A full factorial design was employed to verify the influence of (A) synthesis temperature, (B) deposition time and (C) catalyst loading on carbon and CNT yield. The factorial design was also used to examine the effect of higher order process interactions. ANOVA was conducted at a 95% confidence interval to verify the statistical significance of process parameters. Carbon yields were used as the parameter for optimization. In majority of the literature on parametric studies of CNT synthesis, researchers employed the "change one factor at a time approach" (Venegoni and Serp, 2002, Corrias and Caussat, 2003). However, they did not provide enough data to analyze the effect of interactions. The results of the ANOVA are given in Table 2 and values of $p < 0.05$ indicate a statistically significant variable. Results from ANOVA also imply that the interaction effect of reaction temperature, deposition time and catalyst loading is enormous. Obviously, the influence of reaction temperature on the percentage of the carbon yield depends on the time of deposition and amount of catalyst. To test differences between levels, a number of post hoc comparison techniques were used. The comparison tests showed that four combination of temperature, time and catalyst leads to significant carbon yield percentage. These interactions are (750 °C, 40 min, and 5 gr), (750 °C, 50 min, and 5 gr), (950 °C, 40 min, and 5gr) and (950 °C, 50 min, and 5 gr). The main effects were significant because the P values are less than 0.005 (Venegoni and Serp, 2002).

Table 2. ANOVA for Selected Factorial Model Analysis of Variance Table [a]

source	response 1 : carbon yield	
	F-value	*P-value> F*
model	3877.19	<**0.0001**
A, temperature	12627.09	<**0.0001**
B, deposition time	2310.84	<**0.0001**
C, catalyst loading	10333.50	<**0.0001**
AB	51.77	**0.0004**
AC	220.52	<**0.0001**
BC	734.27	<**0.0001**
R^2	0.999	

[a] ANOVA of carbon yields. A value of $P < 0.05$ indicates a statistically significant variable. These are highlighted in bold.
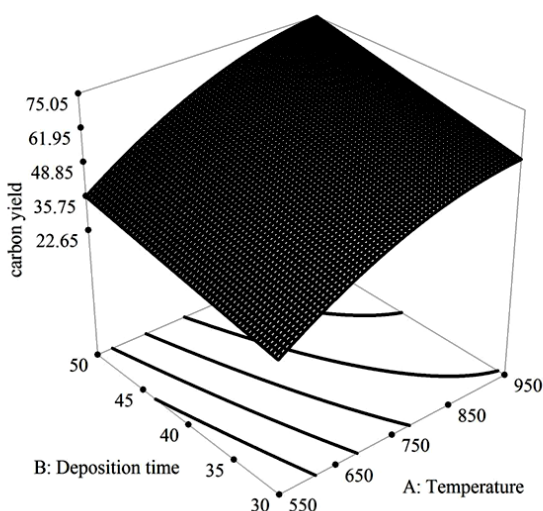
Figure 5. 3D response surface graph for carbon yield: time vs. temperature
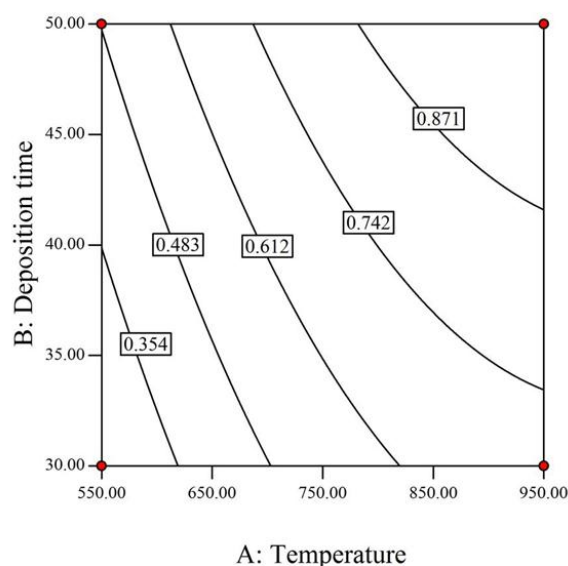
Figure 6. Contour plot for desirability: time vs. temperature

The result suggests that temperature, time and catalyst have considerable effect on the carbon yield. On the other hand, it was shown that the reaction temperature has great influence on the carbon yield and this influence was determined with time of deposition and catalyst loading. Also the effect of deposition time on carbon yield is a result of the reaction temperature and catalyst loading parameters. Similarly, reaction temperature and deposition time dramatically change the effect of catalyst loading on carbon yield. In figure 5, the overall interaction effects are displayed as a 3D representation of the carbon yield. Figure 6 shows contour plot for desirability versus time and temperature which is obtained from the experimental data. According to the graphs, the carbon yield increased proportional to temperature as expected. It confirmed the positive

effect of temperature, time and catalyst loading on the carbon yield. At higher catalyst loading a slight increase was observed due to higher amount of available active sites.

According to the statistical analysis in previous section, two set of experiments exhibited best percentage of carbon deposit. However, image analysis beside TGA results illustrated that the combination of 750°C, 40min, and 5gr is the best for CNT yield percentage. After screening the factors and determining their interactions, the optimization of the main parameters was carried out. Optimization was performed on the basis of desirability function, in order to find the optimal conditions for the carbon yield. The numerical optimization part of the SPSS software was used to locate the maximum desirability function. The desired goal was selected by adjusting the weight or importance of a goal. The goal fields for response have five options: none, maximum, minimum, target and within range. By using all above settings and boundaries, the software optimized 98.1 % carbon yield with calculating

the optimized model factors of temperature at 950 °C, deposition time of 50 min and catalyst loading of 5 gr, respectively (Figure 7).

4. CONCLUSION

We have investigated for the first time, the best operating condition for CNTs synthesis via FBCVD. The process involved acetone as a carbon source in presence of catalyst comprising Fe and Mo supported on alumina. Considering the interaction of listed parameters, the best operating condition (reaction temperature, catalyst loading, and deposition time) was determined based on full factorial design of experiments. The result of ANOVA was used to find the best conditions for carbon yield which confirmed by SEM, TEM and TGA results. On the other hand, SEM and TGA results showed that only the combination of 750 °C, 40 min, and 5 gr leads to the highest amount of CNT yield. It is clearly the best condition to synthesize the CNTs.
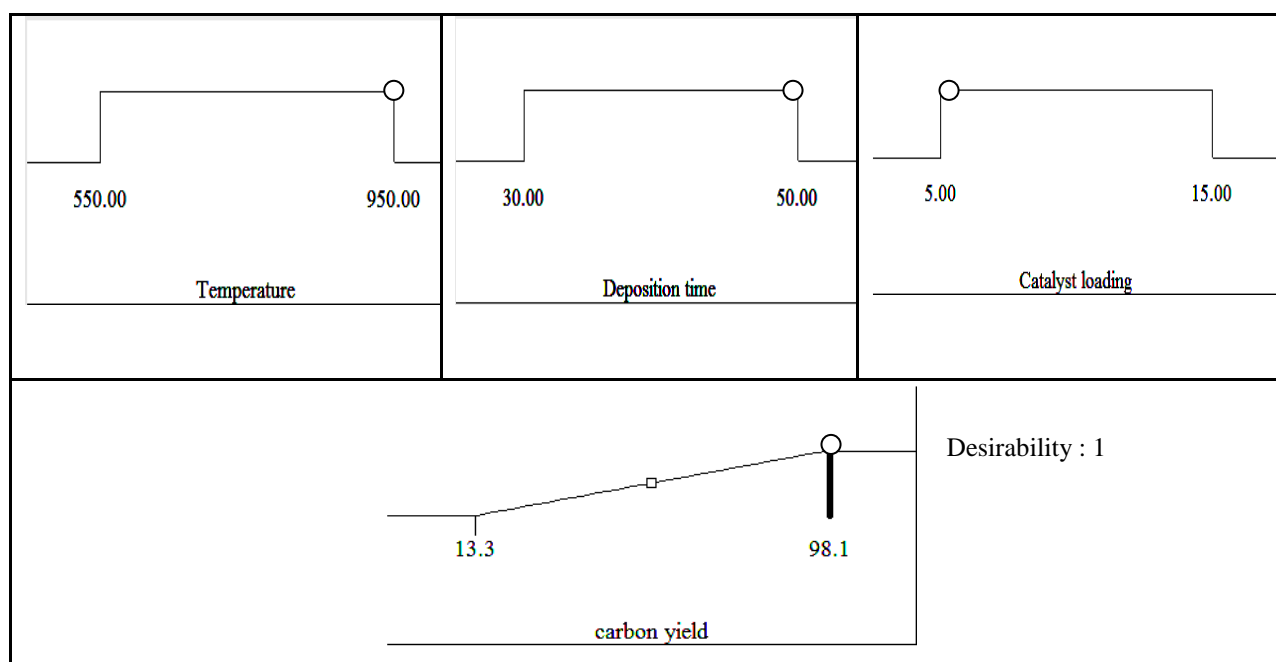

Figure 7. Desirability ramp for numerical optimization.

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