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Prediction length of carbon nanotubes in CVD method by artificial neural network

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Abstract: Most features of carbon nanotubes such as electrical, mechanical and thermal properties are depended on the length of them. Thereby, the applications of carbon nanotubes significantly developed by controlling this key factor. In this paper, we predict the length of carbon nanotubes in chemical vapor deposition (CVD) by using an artificial neural network. First, the effective parameters in CVD for synthesizing carbon nanotubes include the thickness of catalyst, temperature and time of heat treatment, rate of reactant gas; collected from various studies and they were determined as the input. Then, the length of carbon nanotube considered as the output of the artificial neural network. A Feed-forward backpropagation network was designed with 16 and 12 neurons in the first and second hidden layers, respectively. The predicted outcomes were very close to the experimental results, and the created model with 5.6% root mean square error was able to predict the length of carbon nanotubes. It is expected that the designed model can be helpful for researchers to adjust and regulate the suitable parameters among different effective variables in the CVD method. Furthermore, the result of the sensitivity analysis showed that the temperature and rate of reactant gas and thickness of catalyst have the highest impact on the length of carbon nanotubes, respectively.

Keywords: Carbon nanotubes, chemical vapor deposition, prediction length of carbon nanotubes, Artificial neural network.

Introduction

Carbon nanotubes (CNTs) are the fourth allotrope of carbon [1] which were observed in 1952 by Radushkevich and et al. [2]. Especial and symmetric structure, nano-metric radius, low density, high special surface area, conductive and semi-conductive type, absorbing and transferring energy, strong π and σ bonds, 1 TP young modulus and 100 GP tensile strength are the unique properties of CNTs [3]. According to these features; carbon nanotubes have large range applications; such as used in medicine [4], agriculture [5], lithium battery [6], solar cells [7], energy storage [8], etc.

There are different methods for synthesizing carbon nanotubes, and the way of approach can specify properties of CNTs [9]. Arc discharge, laser ablation, and chemical vapor deposition are the main methods for fabrication the carbon nanotubes [3]. However, chemical vapor deposition has a superiority than other methods; because in CVD method mechanism is simple, the process is done in low temperature, growth condition is controllable, obtained coating of CNTs is well-aligned, the procedure is inexpensive and suitable for mass production [10-12]. Basically, all of the properties of CNTs are specified by quantum mechanics, but length and diameter are two key major parameters that affect a lot of properties of carbon nanotubes [3]. Hence, exact determining the length of

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CNTs leads to optimizing using the CVD process. There are a huge number of investigations for fabrication CNTs by the CVD method, but a few of them perform a study that focuses on the prediction length of carbon nanotubes base on machine learning. The artificial neural network (ANN) is one of the most powerful modeling tools for approaching different datasets and reaching an exact solution [13]. This modeling technique is based on learning and subsequently the prediction of output responses [14]. ANN has been widely used in CVD and owing to the different significant parameters in the CVD process, it can helpful to predict desired outputs. In this paper, we collect important variables in the CVD method, include the thickness of catalyst, time and temperature of heat treatment, time and rate of gas reactant, and length of CNTs from valid related papers. Then, the gathered data were used to design an artificial neural network for prediction the length of carbon nanotubes. Finally, the Neurosolution software was used for specifying the most important parameters on the length of CNTs in the CVD method.

Result and Discussion

There are different CVD methods for synthesizing CNTs such as plasma and laser-assisted CVD or water or oxygen assisted CVD, etc [3, 15]. In this investigation for homogeneous datasets and precise prediction; dataset only collected from the articles that used simple CVD, thermal CVD or catalytic chemical vapor deposition (CCVD), because they follow a similar process for fabrication CNTs. Most protective gases in CVD are noble such as Ar, He, and H_2. In most experiments, a mixture of these gases, especially Ar and H_2, are used for producing CNTs. These protective gases usually have not important influence in the synthesis of CNTs [3]; as a result, they were not considered in the data collection. Different reaction gases in the CVD method are Methane, Ethane, Acetylene, Ethylene, and Carbon Monoxide [16]. Among these gases, unsaturated hydrocarbon Acetylene was more applied in many articles, due to the appropriate rate for fabrication alignment of carbon nanotubes [17]; therefore Acetylene selected as the reactant gas for data collection. Co, Ni, and Fe are the most useful catalysts that alone or mixed with other metals used for synthesizing of CNT by CVD method [8, 16]. In this study, Fe due to the abundant number of research, more accessibility, and low cost selected for the catalyst. Furthermore, the thickness of Fe catalysts in all collected resources was not more than 50 nm for homogeneity of datasets. All information with the above conditions were collected and listed in Table **1**.

Table. 1: The collected dataset with detail of major parameters in the CVD method for producing CNTs.

No	Source	Thickness of catalyst (nm)	Temperature of heat treatment (C°)	Time of heat treatment (min)	Rate of reactant gas (sccm)	Time of reactant gas (min)	Temperature of reactant gas (C°)	Length of CNTs (nm)
1	[18]	30	750	20	30	10	950	5000
2	[18]	30	850	20	30	10	950	10000
3	[18]	30	950	20	30	10	950	20000
4	[19]	2	800	40	10	10	800	114
5	[19]	2	800	40	30	10	800	157
6	[19]	2	800	40	20	10	800	150
7	[19]	2	800	40	40	10	800	97
8	[19]	2	800	40	50	10	800	57
9	[20]	5	750	8	100	10	750	20780
10	[20]	5	750	12	100	10	750	19000
11	[20]	5	750	10	100	10	750	17000
12	[20]	5	750	14	100	10	750	20780
13	[21]	2.8	750	15	12	15	750	24.5
14	[21]	2.8	900	15	12	15	900	23.8
15	[22]	50	950	120	30	10	950	18000
16	[22]	50	950	20	30	3	950	3600
17	[22]	50	950	20	30	20	950	20000
18	[22]	50	950	20	30	30	950	28000
19	[22]	50	950	20	30	10	950	12000

20	[22]	50	950	240	30	10	950	8000
21	[23]	1	725	15	3	15	650	149000
22	[23]	1	725	15	3	15	700	290000
23	[23]	1	725	15	3	15	725	340000
24	[23]	1	725	15	3	5	725	175000
25	[23]	1	725	15	3	10	725	229000
26	[23]	1	725	15	3	15	725	350000
No	Source	Thickness of catalyst (nm)	Temperature of heat treatment (C˚)	Time of heat treatment (min)	Rate of reactant gas (sccm)	Time of reactant gas (min)	Temperature of reactant gas (C˚)	Length of CNTs (nm)
27	[23]	1	725	15	3	15	725	349000
28	[24]	8	700	30	120	30	800	6500
29	[24]	8	700	30	120	30	900	1500
30	[25]	5	580	30	400	60	750	100000
31	[26]	7	700	25	10	20	750	3460
32	[26]	5	700	25	10	20	750	3460
33	[27]	27	900	5	30	20	900	35000
34	[28]	1	750	3	5	30	750	100000
35	[29]	2	650	30	75	40	750	5000
36	[29]	10	650	30	75	40	750	80000
37	[29]	20	650	30	75	40	750	35000
38	[29]	20	650	30	44.4	40	750	44500
39	[20]	20	650	30	109.2	40	750	29000
40	[30]	50	950	20	30	10	950	10000
41	[31]	30	850	20	30	5	850	15830

ANN Results

In order to test the accuracy of the network performance, regression analysis was performed for training and testing data sets. The result of the regression analysis is shown in Figure **2**. According to this graph, the total regression (total regression of test, train, and validation) was 0.99973. Better regression leads to less scattering between datasets and predicted values. Thereby, it is completely reasonable that the error percentage of the measured regression will be less and the obtained relation would be more accurate, due to being very close to 1. It can be seen that the artificial neural network has been able to reach a very close relation between the experimental variables and the prediction values and the network has been able to find an appropriate equation. For verification of the network, a comparison between experimental and predicted values datasets was carried out. Regarding equation 4, the average error of the network was calculated by 5.6%. Based on this result, it can be expected that the modeled ANN network can predict other similar results in order to predict the length of CNT with such high accuracy and reliability. Moreover, with attention to the many variables involved in the CVD method, the proposed model reduces the time and costs of experimental research.

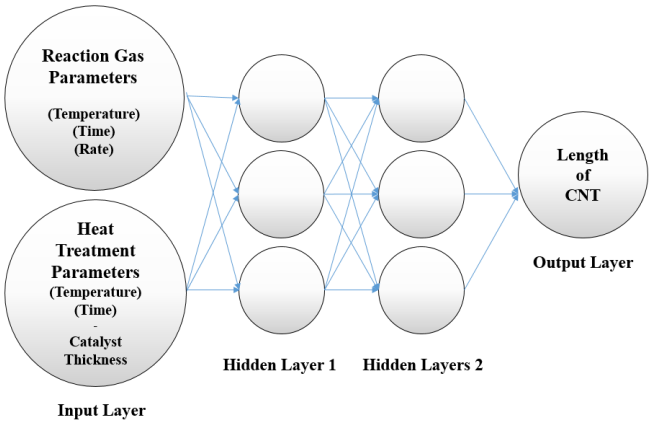

Figure 1: Diagrams of the designed ANN architecture.

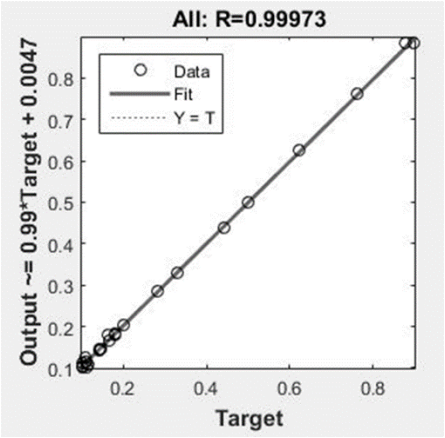

Figure 2: Schematic of regression based on the designed ANN model for prediction length of CNTs in the CVD method.

Sensitivity Analysis

Although neural network modeling can predict the outcome parameter, it is not able to determine the effect of each parameter. In this study, due to the use of Neurosolution, we can find and specify the least and the most important factors [40]. Indeed, the influence of each parameter can be determined by using SA. This analysis explains which of input more important than the others and so it is very useful and acts as the supplement of the ANN. Figure **3** provides information about the result of Neurosolution from the sensitivity analysis of collected datasets. According to the results, the temperature and rate of reactant gas, and the thickness of the catalyst have the highest impact function, respectively.

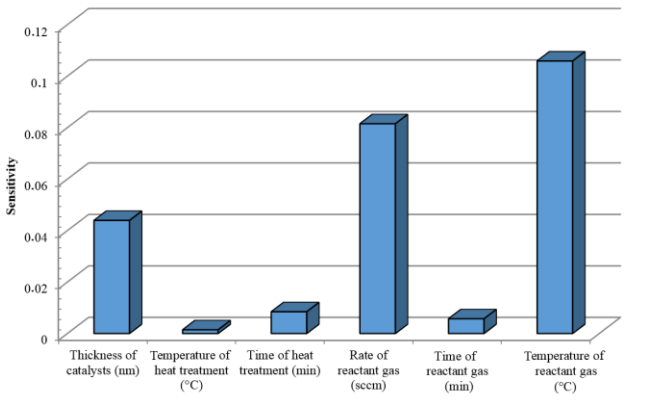

Figure. 3: Sensitivity analysis of synthesized CNTs via the CVD method based on collected data.

According to these results, the temperature of the reaction gas is the most important parameter in the sensitivity analysis of the CVD process. Similarly, some studies reports [18, 41] that the temperature of reactants has a major effect on the growth of CNTs. Based on the nucleation and the growth mechanism for fabrication carbon nanotubes in the presence of catalyst [11, 42], nucleation and growth of nanotubes perform on the catalytic particles. By reducing the solubility and deposition of carbon atoms in the solution of carbon atoms and metal catalysts; carbon atoms by SP^2 bonds are bonded. Generally, the temperature of reactant gas prepares a suitable condition for connecting carbon atoms in the substrate. Likely, the temperature of acetylene increases the solubility of carbon and the small change in it leads to a transformation in the length of the carbon nanotube.
The second important parameter is the rate of reactant gas (C_2H_2). According to the other researches [43, 44], the rate of reactant gas influenced the structure of CNTs and can handle the rate of connecting carbon atoms for production carbon nanotubes. Indeed, acetylene acts as a precursor and rate of it characterize when and where bonds of carbon atoms could be extended or cease, with attention to the supersaturated condition in the solution of carbon atoms and Fe catalyst.

Conclusion

The ANN model with 16 and 12 neurons in hidden layers 1 and 2, respectively, is a useful method for the prediction of length of carbon nanotubes synthesized by the CVD method. In this study, the designed ANN model predicted the length of CNTs with an average error of 5.6%.The results of the network and length of carbon nanotubes have a remarkable agreement with the experimental data. According to the sensitivity analysis, the rate and temperature of reactant gas, and the thickness of the catalyst have the highest impact on the length of carbon nanotubes, respectively.

Experimental

ANN Modeling Procedure

ANN network generally contains interconnected units known as neurons or nods. Neurons are the smallest computing elements which interconnected to weighted links and they aggregate into layers. These layers affect their input information and can be trained by a process [32-35]. Indeed, ANN consists of input layers, output layers, and hidden layers and neuron signals transmitted several times from input to the output. The training process of ANN continuous intermittently by changing weights until the network could approach the desired output and reaches to the

acceptable error. After training, the network can predict the output of untrained data by using the designed model that was learned at the training step.
The relationship of neurons can be expressed by relation (1):

$$x = \sum_{i=1}^{p} w_i x + b \qquad (1)$$

Where the output x produced by the neuron in the layer, p is the number of elements in the layer, $w_i x$ is the weight, and b is the offset or bias.

ANN Architecture

Among the total number of data, 33 and 8 datasets were used randomly for training and verification of the network, respectively. Feed-forward backpropagation (FFBP), which is one of the most suitable ways for the training of the network in ANN [36], was used for training the model. This method presents effective solutions for approaching different factors in order to find a solution [37, 38]. The number of neurons in the hidden layers during the training process was determined by trial and error. This network includes an input layer, two hidden layers and, an output layer. There are 16 and 12 neurons in the first and second hidden layers, respectively. Figure. 1 provides information about the schematic diagrams of the ANN model configuration. As can be seen, there are 6 inputs and 1 output. The input variables are the thickness of catalyst, temperature and time of heat treatment; rate, time, and temperature of reactant gas; and the length of carbon nanotube considered as the output.

The network modeling was written in MATLAB software version R2014a and the Levenberg–Marquardt (LM) algorithm [39] was used to train the network. Furthermore, the log-sigmoid transfer function was applied as an activation function for hidden and output layers. The data sets have been normalized between 0.1 to 0.9 for homogenization according to relation (3):

$$\mathrm{N} = 0.8\left(\frac{x - x_{min}}{x_{max} - x_{min}}\right) + 0.1 \qquad (3)$$

Where x_{max} and x_{min} are the maximum and minimum values of the parameters, respectivelly.

The root mean square errors (RMSE) for the designed network was computed by:

$$\mathrm{RMSE} = \frac{1}{\mathrm{N}}\sum_{1}^{\mathrm{N}}\left(\frac{|\text{Actul value} - \text{Predicted value}|}{\text{Actul value}} \times 100\right) \qquad (4)$$

Where N is the total number of training patterns.

The thickness of the catalyst is the third important factor and commonly mentioned in some reports [45-47] as a key parameter for synthesizing CNT via CVD. Fe catalyst which covered in the surface, decomposed during the process and with Root or Tip growth mechanism, leads to bonding carbon atoms with SP^2 hybridization. Moreover, the thickness of catalyst influence in the growth location and alignment of CNTs and consequently it is so much effective in the length of carbon nanotubes. It is noticeable that despite the limited size of Fe catalyst (< 50 nm), this parameter still has a considerable impact on the Length of CNTs in the CVD method.

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