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RESEARCH PAPER

Optimization of Continual Production of CNTs by CVD Method using Radial Basic Function (RBF) Neural Network and the Bees Algorithm

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ABSTRACT

Optimization of continuous synthesis of high purity carbon nanotubes (CNTs) using chemical vapour deposition (CVD) method was studied experimentally and theoretically. Iron pentacarbonyl ($Fe(CO)_5$), acetylene (C_2H_2) and Ar were used as the catalyst source, carbon source and carrier gas respectively. The synthesis temperature and flow rates of Ar and acetylene were optimized to produce CNTs at a large scale. A flow rate of 30-120 sccm of acetylene and 500-3000 sccm of Ar at temperatures between 650-950 ˚C were examined. Using the fundamental trial and error method it was found that the maximum yield of pure CNTs can be produced at 750 ˚C with flow rates of 40-45 sccm of acetylene and 1500 sccm of Ar. In theoretical part, an artificial neural network (ANN) and the Bees Algorithm (BA) were used to model and optimize the CNTs production, based on the experimental data. The Bees Algorithm used the ANN as the fitness function and the optimum variables found as 60 sccm for acetylene, 555 sccm for argon and 759 ˚C for temperature. The computational results have relatively good agreement with the experimental results.

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INTRODUCTION

Nowadays, the optimization process can play an important role in the experimental studies. Finding the optimum conditions using the conventional trial and error method is very time consuming and costly. So, optimization process based on experimental data is very useful to achieve such a goal. Here, we report the optimization of CNTs production conditions at a relatively large scale using continuous thermal CVD and confirmation of the production conditions using the artificial neural network.

Carbon nanotubes (CNTs), formed by rolling of graphene sheets into a tube shaped structure are unique 1D nanostructures. CNTs have been subjected to intense experimental investigations due to their novel mechanical and electrical properties. These properties accompanied by their high aspect ratio make them ideal for various potential applications such as electron field emitters, single molecular transistors, scanning probe microscope tip, hydrogen and energy storage, sensitive gas sensors, reinforcement agent of composites, etc. [1-6].

To date, many sintering methods have been developed for the production of CNTs such as the arc discharge method, laser ablation [7-9] and chemical vapour deposition (CVD) [10-12]. Among these methods, the CVD is more promising due to its large scale and continuous production potential,

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low price, controlled synthesis conditions and the possibility of dense arrays of CNT growth [13-17]. In this method at certain temperatures, the nanostructured metal catalysts like Fe, Co and Ni decompose a gas phase carbon source like hydrocarbons or other carbon precursors. The decomposed carbon atoms then are formed into the CNTs structures.

In order to find the optimal conditions for CNT production, different flow rates of C_2H_2 between 30 and 120 sccm and Ar between 500-3000 sccm were tried. The reaction temperature was chosen between 650 and 950˚C. The reaction time for all experiments was 30 minutes afterward the furnace was cooled down to room temperature under a small flow of Ar. The samples were characterized using a scanning electron microscope (SEM, LEO 1455VP) and a transmission electron microscope (TEM, LEO 906E). The XRD pattern of samples was taken using a Philips diffractometer (PW 1840) at room temperature utilizing Cu Kα radiation wavelength of λ = 1.5418 Å.

The artificial neural network (ANN) was used to optimize the production conditions. Neural network models [18], assume many simplifications over actual biological neural networks. Such simplifications are necessary to understand the intended properties and to attempt any mathematical analysis. An artificial neural network (ANN) tries to model a living system by attempting to replicate its description from observation of the input/output behaviour. Many different internal descriptions can capture the input/output behaviour over the domain of observation, but the property of autopoiesis can be satisfied only by the internal states and intricate connections and dynamics of a living system. For this to happen in an ANN, the system must incorporate the feature of structural adaptation [19].

In order to solve many complex multi-variable optimization problems, it is necessary to use search algorithms that they can find optimal solution in the reasonable running times. The Bees Algorithm is one of the relatively new population based optimization techniques. It is inspired by the natural foraging behaviour of honey bees to find the optimal solution. Successful applications of the BA to a wide range of optimization problems, like benchmark test functions [20], mechanical design problems and other optimization problems [21] have demonstrated its potential and established it as an efficient optimization tool.

MATERIALS AND METHODS

The experimental system consists of a tube furnace with a heating zone of 20 cm and a quartz tube with an inner diameter of 3 cm as the reaction media. The quartz tube was connected from one end to the gases' entrance and from the other end to the exhaust. The CNTs were formed by introducing the C_2H_2 as the reaction gas, Ar as the carrier gas and ($Fe(CO)_5$) as the catalyst source into the reaction media. It was observed that the CNTs were formed everywhere on the inner surface of the quartz tube. Because of the liquidity of ($Fe(CO)_5$) at room temperature, it was entered into the reaction media through a bubbler by direct bubbling of $C_2H_2$with various flow rates. The flow rates were controlled by local flow meters with an accuracy of one sccm. The bubbler was kept at 0 °C in order to control a uniform evaporation of iron pentacarbonyl. This way of introducing the nanostructured metal catalyst into the furnace, was a key to have a continuous production. Indeed, as long as the flowing of the acetylene was last, the production of CNTs was continued. The parameters that were to be optimized were the reaction temperature and the flow rates of Ar and C_2H_2. Any changes in these parameters could affect the purity and the yield of CNTs. Table 1 shows the conditions for the preparation different batches. In spite of producing CNTs with all conditions,

Table 1. Different conditions for CNTs production

Sample	Furnace temperature (˚C)	Flow rate of C_2H_2 (sccm)	Flow rate of Ar (sccm)	CNT formation
1	750-800	100-120	1000-1500	light
2	700-750	40-45	1000	dense
3	750	40-45	1500	high dense
4	750	40-45	2000	dense
5	750	40-45	3000	light
6	850	40-45	1500	dense
7	900	40-45	1500	dense
8	750	30-35	500-1500	light
9	650	40-45	1500	medium

based on SEM and TEM results, the quality and yield were different.

By trial and error it was found that the flow rate of 40-45 sccm of C_2H_2, 1500 sccm of Ar and sintering temperature of 750 ˚C are the best choice for a dense production of CNTs. Figs. 1 and 2 show the SEM, TEM and XRD of the best sample.

RESULTS AND DISCUSSION

The neural network used in this study was a Radial Basic Function (RBF) neural network and has some specific characteristics. Fig. 3 shows the schematic representation of the RBF neural network structure. It has an input layer that represents the input variables to the neural network model which are Argon, Acetylene and temperature. This layer does not analyse the date. Also the RBF has an output layer that shows the result of the process, while the output layer is the quality of produced CNTs from the experiments. The hidden layers make a nonlinear correlation between input and output layers. In the RBF networks, the Gaussian functions were used as the transmission functions. The structure of ANN which was used in this paper

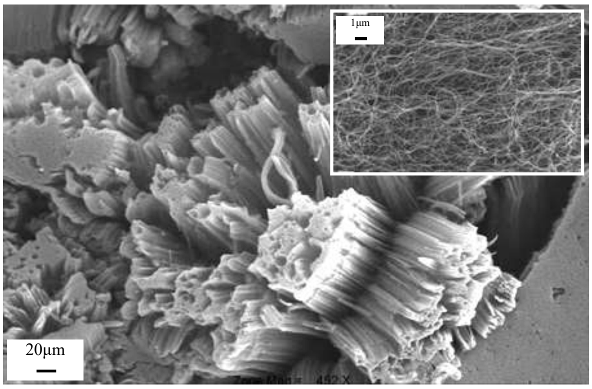

Fig. 1. SEM image of the best sample (sample 3). The inset shows SEM of the sample dispersed before imaging

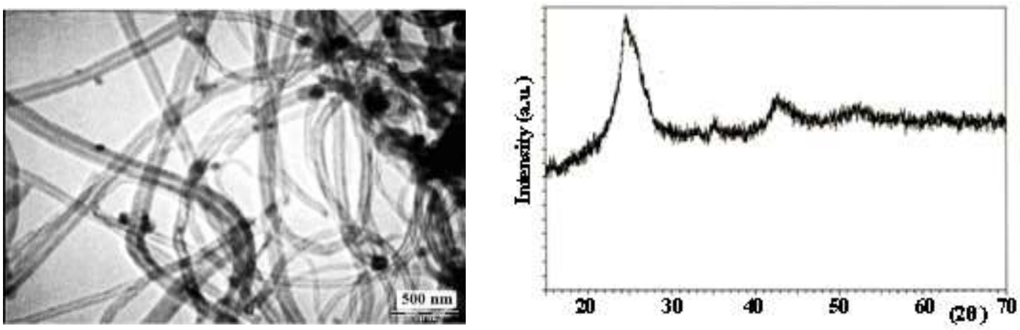

Fig. 2. TEM image (left) and XRD pattern (right) of the best sample

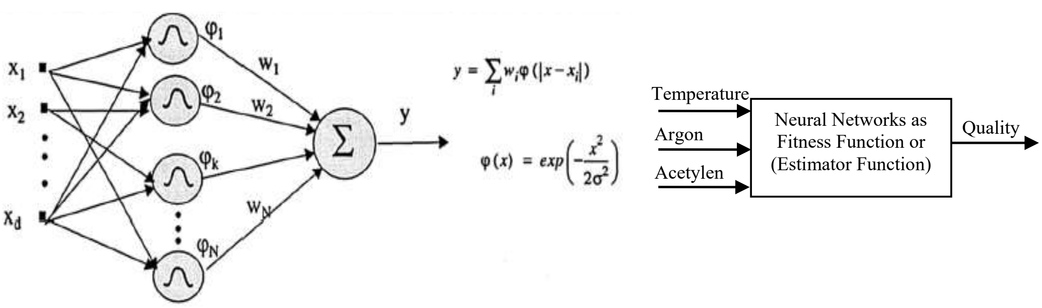

Fig. 3. Schematic representation of the RBF network

consist of 3 hidden layers that number of neurons in each layer is 10, 10 and 4 respectively. In order to train this network, cross validation method was used. 2/3 of total experimental data were selected to train while the rest of data were used to test the network. To make a good train, the data should select randomly among all regions of data. So, the network can be abled to have an acceptable interpolation and extrapolation. The train procedure repeated until the mean square error (MSE) of the network and the experimental data become less than specified value (eta). Fig. 4 shows the schematic representation of the training process.

ANN was used to estimate the fitness function calculator for the optimization problem. The BA requires a number of parameters to be set, namely: number of scout bees (n), number of

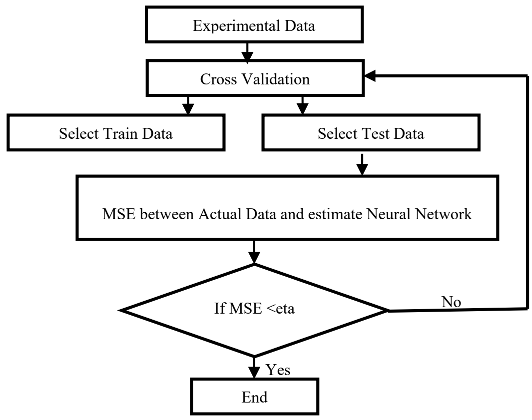

Fig. 4. Schematic representation of training process of NNs

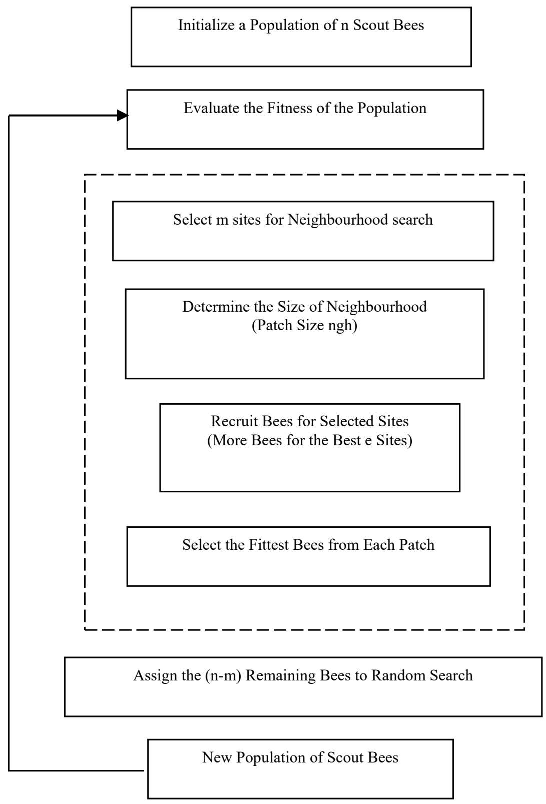

Fig. 5. Flowchart of the BA

sites selected out of n visited sites (m), number of the best sites out of m selected sites (e), number of the bees recruited for the best e sites (nep), number of the bees recruited for the other (m-e) selected sites (nsp), initial size of patches (ngh) which includes site and its neighbourhood and stopping criterion. Fig. 5 shows flowchart of the Bees Algorithm. For more details, the reader is referred to [23].

The optimization problem formulated as follows:

Maximize J = ANNs (MLP) = f (T, Ar, Ac)
Subject to $650 < T < 950$
$500 < Ar < 3000$
$30 < Ac < 120$

The resulting test of this network is shown in Fig. 6. In this graph, O, represents the experimental data and +, represents the estimated value by ANN. According to this graph, the estimated values are in a good agreement with the experimental data.

Then, by use of this trained ANN as a fitness function, the convergence of quality graph was obtained and is shown in Fig. 7. The BA method is evaluated on experimental dataset according to Table 1 and compared against the state of the experimental results. According to this convergent, the designed variables that consist of argon, acetylene and temperature were obtained 555 sccm, 60 sccm and 759 °C respectively.

In order to test the results of the ANN and BA experimentally, an experiment was performed at predicted conditions. Fig. 8 shows the SEM image of this sample which confirms that the CNTs have been successfully synthesized by the predicted conditions and the computational results have relatively in a good agreement with the experimental ones.

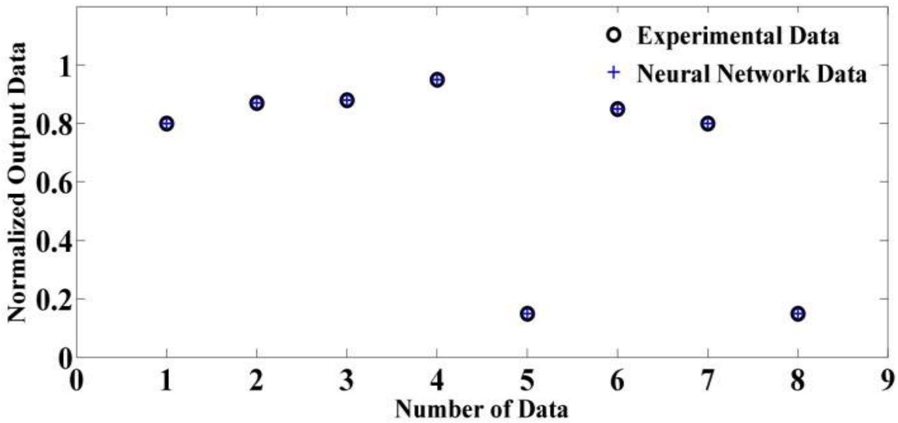

Fig. 6. Comparison between the experimental data and the estimated value by ANN

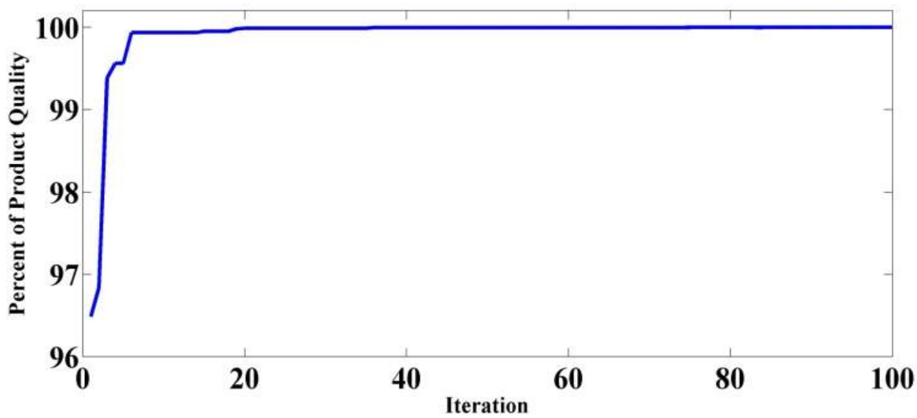

Fig. 7. Converge of quality graph by use of trained ANN as a fitness function

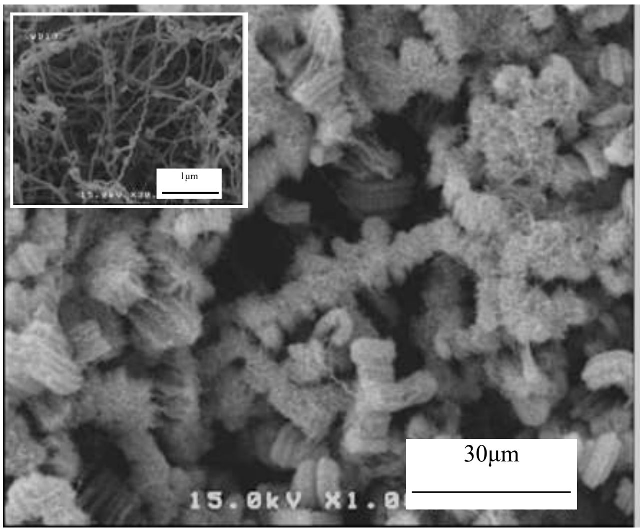

Fig. 8. SEM image of the sample that was produced using predicted conditions by the ANN and BA

CONCLUSIONS

In this study, CNTs at a large scale were grown using CVD under different conditions. The flow rates of 1500 sccm of Ar and 40-45 sccm of acetylene at 750˚C were the optimal conditions for large scale production of nearly pure CNTs. Combination of artificial neural networks (ANN) and the Bees Algorithm (BA) was applied for optimization of CNTs production by use of the experimental data. So, the optimum variables were obtained as 60 sccm for acetylene, 555 sccm for Ar and 759˚C for temperature. The conditions which were found by the BA have good agreement with the conditions deduced from the experiments.

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CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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