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Optimization of catalyst formation conditions for synthesis of carbon nanotubes using Taguchi method

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ABSTRACT

A growth of Carbon Nanotubes (CNTs) suffers many difficulties in finding optimum growth parameters, reproducibility and mass-production, related to the large number of parameters influencing synthesis process. Choosing the proper parameters can be a time consuming process, and still may not give the optimal growth values. One of the possible solutions to decrease the number of the experiments, is to apply optimization methods to the design of the experiment parameter matrix. In this work, Taguchi method of designing experiments is applied to optimize the formation of iron catalyst during annealing process by analyzing average roughness and size of particles. The annealing parameters were: annealing time (t_{AN}), hydrogen flow rate (f_{H2}), temperature (T_{AN}) and argon flow rate (f_{Ar}). Plots of signal-to-noise ratios showed that temperature and annealing time have the highest impact on final results of experiment. For more detailed study of the influence of parameters, the interaction plots of tested parameters were analyzed. For the final evaluation, CNT forests were grown on silicon substrates with AlO_X/Fe catalyst by thermal chemical vapor deposition method. Based on obtained results, the average diameter of CNTs was decreased by 67% and reduced from 9.1 nm (multi-walled CNTs) to 3.0 nm (single-walled CNTs).

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1. Introduction

In the last years, since discovery by Iijima in 1991 [1], Carbon Nanotubes (CNTs) have received much attention due to their extraordinary physical, mechanical, electrical and optical properties [2,3]. CNT forests with high density and vertical alignment may provide uniform CNTs with preferable length, diameter and structure for mass production of CNTs [4].

It is known that the physical properties and structure of CNT forests directly affect mechanical, thermal, electrical and optical properties of CNTs. On account of possession of such extraordinary properties, CNTs are suitable for use in many applications such as nanoelectronic devices [5], water and gas filters and sensors [6], actuators and polymeric composites for electronic devices [7]. Electrical properties are utilized in bulk shaped CNT forests as electrodes with large surface area [8], high emission current density emitters [9] and interlayer connectors for large-scale integrations (LSIs) [10]. Due to high absorbance and low reflectivity [11] CNTs exhibit properties close to that of blackbody, which is claimed to absorb all spectra of incident light [12].

To achieve the desired properties of CNTs many various methods of CNTs synthesis are used. The most popular are arc-discharge, laser ablation and chemical vapor deposition (CVD), among which CVD method is the most common [13]. Catalytic CVD method is a simple and cheap way to grow CNTs at low temperature and ambient pressure. In comparison to the other two methods, the catalytic CVD provides higher yield and higher purity of grown nanotubes and allows for easier control of growth parameters and structures of CNTs [13]. Also, the equipment used for growth is easy to modify, causing an expansion and further development of CVD methods to one of the following types: thermal CVD, Plasma Enhanced CVD (PECVD) [14], water-assisted (super growth) CVD [15] and many others. However, despite of all the advantages of CVD method, still many problems connected to high density growth of Single Walled CNTs (SWNT) can be found [16].

Even though many years have passed since the discovery of CNTs, it is still difficult to fabricate high-density and high-length SWNT forests, due to the influence of many process parameters on the growth process [17–20]. One of the most important factors is the annealing process, which allows for the formation of the catalyst nanoparticles which govern the diameter of CNTs [19,21].

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Also, the density of formed catalyst particles influence the density of CNTs forest. Although many experiments have been made, the mechanisms of catalyst formation are not clear. For example, Sakurai et al. [22] explained the formation of the catalyst particles by interpretation of the role of subsurface diffusion and Ostwald ripening phenomena. In their work, the Ostwald ripening was shown to be the main cause of elimination of small particles and subsurface diffusion was the cause of reduction in the size of larger particles, giving high uniformity of catalyst size. However, to utilize those phenomena in the catalyst formation process, complicated calculations are needed [23]. These complex calculations take more time and make modification of the process even more difficult.

The growth of CNT forest may cause many difficulties in finding optimum growth parameters, reproducibility and mass-production, related to the large number of parameters influencing the growth during synthesis process. Testing possible parameters separately and choosing the proper values of the CNTs growth can be a time consuming process, which can give us a local minimum. However, there might still be a possibility of the existence of better values, which would require a larger number of experiments by standard procedures to check all the possibilities. One of the many possible solutions to decrease the number of experiments is to apply optimization methods to the design of the experiment parameter matrix [24]. This method allows to screen the factors and determine which are important for explaining process variation. The statistical approach allows for the systematic investigation of the process and the parameters that influence the quality of final product [25–27]. Identification of significant factors shows the direction of improvement to enhance manufacturability, reliability and quality of a product. Recently, Taguchi method was also successfully used in carbon nanotube field, in order to optimize CNT growth [28–31], properties [32] and their use in various applications [33–35]. In those works, Taguchi method was applied to increase a yield of CNTs, tune field emission properties [32] or optimize use of CNTs for applications; however, none of this works regard formation of catalyst particles, which determines structure of CNTs in the forest.

In this work, the statistical approach of designing CNTs growth experiments by Taguchi method in a small number of experiments, developed to control the size and roughness of the catalyst particles after annealing process will be presented. The article is focused on the determination of the optimum parameters and mechanisms of formation of the catalyst, which determines the diameter of CNTs. Growth of CNTs involves many various steps, which cause high noise and low reproducibility of process [36,37]. By employment of Taguchi method, it is possible to remarkably decrease or eliminate negative effects of producing process through statistical approach. Using Taguchi orthogonal arrays, we were able to dramatically decrease the number of experiments from 4^4 (=256) to 16, while keeping high accuracy, at the same time. Due to the fact that all the annealing parameters in the design were weighted equally, we were able to evaluate all factors independently, so the effect of one parameter did not influence other parameters. The aim of the experiments was to reduce average surface roughness and size of the particles after annealing process, based on Taguchi method of designing experiments. Further, the physical properties of CNT forests grown with the optimized parameters were measured and the effect of different annealing parameters on CNTs growth is discussed.

2. Experimental and methodology

Carbon nanotube forests have been grown on an AlO_X/Fe catalyst deposited on p-type (100) silicon substrates. The radio frequency (RF) magnetron sputtering method was used to deposit a 30 nm thick AlO_X support layer and a 0.5 nm thick Fe catalyst layer continuously without breaking vacuum. The deposition chamber was equipped with an Al_2O_3 target of 99.99% purity (4N) and a Fe target of 99.99% purity (4N), 2 inches diameter each, placed horizontally on the top of the chamber. Argon gas of 99.9999% purity (6N) was supplied to the deposition chamber through a mass flow controller (Horiba SEC-400MK3).

Table 1
Deposition parameters of AlOx/Fe catalyst system.

Pressure of residual gases [Pa]	$<5.0 \times 10^{-4}$
Working pressure [Pa]	0.8
Argon flow rate [sccm]	25
RF Discharge Power	50 W for Al_2O_3, 25 W for Fe
Substrate BIAS voltage [V]	floating
Target-substrate distance [mm]	100

Table 2
The optimized parameters and their value level applied in the experiments.

No.	Parameter optimized	Variability interval			
		1.	2.	3.	4.
(1)	Annealing time (t_{AN}) [min]	1	2.5	4	5
(2)	Hydrogen flow rate (f_{H2}) [sccm]	0	35	65	100
(3)	Annealing temperature (T_{AN}) [°C]	730	760	790	820
(4)	Argon flow rate (f_{Ar}) [sccm]	0	20	40	60

Catalyst layers were deposited on electrochemically polished ($R_A \leq 0,15$ nm) p-type 2 × 2 cm Si square plates. Samples were mounted on a substrate holder, introduced to the vacuum chamber, and evacuated to pressure below 5.0×10^{-4} Pa. After evacuation, a sample holder was moved away from the alumina target. In order to remove oxides and impurities from the surface of the cathode, a process of target cleaning was performed under the argon pressure of 0.8 Pa for 10 min. After the cleaning process, alumina sputtering of the support layer was performed (24 min 8 s), followed by cleaning procedure of Fe target (for 5 min) and catalyst deposition (for 10 s) at the same partial pressure. Deposition parameters of AlO_X/Fe layer are shown in Table 1.

CNT growth was performed using thermal chemical vapor deposition (CVD) technique on as prepared Si/AlO_X/Fe samples inserted to vacuum chamber. A CVD chamber −50 cm long, (2 in. outer diameter) quartz tube was evacuated to the base pressure below 5.0×10^{-4} Pa. Then heating in vacuum was performed with a temperature ramping speed of 60 °C/min until annealing temperature was reached. Based on Taguchi orthogonal table [38] of experiments (Table 2), various temperatures, gases flow rates of argon and hydrogen, and annealing times were used. After annealing, growth of CNTs was performed using only acetylene gas (C_2H_2) with a flow rate of 10 sccm at 54 Pa partial pressure for 10 min. No additional gases were used. Based on previous reports [39,40], annealing parameters (ref. standard) used during growth of CNTs were: annealing time of 3.5 min, temperature of 730 °C, hydrogen and argon flow rates of 0 sccm. Those parameters were used as a base for the Taguchi optimization process, as a starting point to decrease diameter of CNTs.

The purities of argon, hydrogen during annealing, and acetylene during growth, were 99.9999% (6N), 99.99999% (7N) and 99.999% (5N), respectively. After the growth, the CVD chamber was cooled by external air flow with 100 °C/min speed, down to 300 °C, when the air flow was stopped.

Taguchi method, which provides an efficient way for designing products that operate consistently and optimally over a variety of conditions, was applied to develop the plan of experiments and to elaborate upon obtained experimental results. Taguchi method allows for the definition of which controllable factors reduce the variation of results and make the product insensitive to changes

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