UNIVERSITI PUTRA MALAYSIA

LOW-TEMPERATURE SYNTHESIS OF CARBON NANOTUBES VIA FLOATING CATALYST-CHEMICAL VAPOR DEPOSITION METHOD

MAHMOOD RASHID ATIYAH

LOW-TEMPERATURE SYNTHESIS OF CARBON NANOTUBES VIA FLOATING CATALYST-CHEMICAL VAPOR DEPOSITION METHOD

By

MAHMOOD RASHID ATIYAH

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

April 2010

DEDICATION

بسم الله الرحمن الرحيم

وَمَنْ يَتَّقِ اللَّهَ يَجْعَلْ لَهُ مَخْرَجًا وَيَرْزُقْهُ مِنْ حَيْثُ لا يَحْتَسِبُ

وَمَنْ يَتَوَكَّلْ عَلَى اللَّهِ فَهُوَ حَسْبُهُ إِنَّ اللَّهَ بَالِغُ أَمْرِهِ قَدْ جَعَلَ اللَّهُ

لكل شَيْءٍ قَدْرًا

صدق الله العظيم

I dedicate this work to my late parents

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

LOW-TEMPERATURE SYNTHESIS OF CARBON NANOTUBES VIA FLOATING CATALYST-CHEMICAL VAPOR DEPOSITION METHOD

By

MAHMOOD RASHID ATIYAH

April 2010

Chairman: Dayang Radiah Awang Biak, PhD

Faculty : Engineering

Carbon nanotubes (CNTs) are widely synthesized at high temperatures via floating catalyst chemical vapor deposition (FC-CVD) method. It is important to reduce the synthesis temperature of CNTs to allow better control of the reactor's conditions, and to eliminate the formation of carbon by-products as well as reduce the overall cost. Therefore, the main objectives of this work were (i) to synthesize carbon nanotubes at low temperatures using some improvements to the CVD technique, (ii) to investigate the effects of temperature on the synthesis of CNTs and (iii) to simulate the temperature and velocity profile of the FC-CVD system in the absence of reaction.

The synthesis temperature of CNTs was examined in the range between 500^{o}C and 600^{o}C at 10^{o}C interval. The preheating set temperature was varied between 150^{o}C and 400^{o}C at 50^{o}C interval. All experiments were conducted at 1 atm and exposed sections were insulated with glass wool covered with aluminum foil. Three O-ring heating

mantels were used as a preheater and three ceramic boats were used to collect the product. Temperature in-situ monitoring device was used to monitor the temperature profile in the reactor and provide the exact time to heat up the catalyst and, thus, initiate the reaction. A single heat source for both catalyst and reactor was employed to enhance the growth of CNTs. COMSOL Multiphysics was used to simulate the velocity and temperature profiles of the system in the absence of reaction. The morphology and internal structures of the CNTs formed were analyzed via Scanning Electron Microscope (SEM), Transmission Electron Microscope (TEM) and High Resolution Transmission Electron Microscope (HRTEM). The thermal stability and the purity of the products were analyzed by Thermal Gravimetric Analyzer (TGA).

The results of the work showed that CNTs were formed at the synthesis temperature ranging from 530^{o}C to 600^{o}C. Both quality and quantity of CNTs synthesized were increased with the increase in synthesis temperature. The highest amount of carbon deposit was obtained when the preheating and the synthesis temperatures were set at 300^{o}C and 600^{o}C, respectively. However, based on the TGA results, the highest purity of CNTs was achieved when the preheating and the synthesis temperatures were set at 200^{o}C and 600^{o}C, respectively. Well aligned CNTs were found when the preheating temperature was set at 400^{o}C for synthesis temperatures of 570^{o}C and 580^{o}C. The use of single heat source for both catalyst and reactor was found to induce the formation of well aligned CNTs at synthesis temperature of 550^{o}C. The simulation results indicated some regions in the reactor have variation in both temperature and velocity profiles. This, indirectly, affects the formation of CNTs. To conclude, this work has successfully

achieved the outlined objectives. The amount of product produced and its quality depend both on the preheating values and the synthesis temperature.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

SINTESIS NANOTIUB KARBON(CNT) PADA SUHU RENDAH DENGAN KAEDAH PEMANGKIN TERAPUNG PENGENDAPAN WAP KIMIA

Oleh

MAHMOOD RASHID ATIYAH

April 2010

Pengerusi: Dr.Dayang Radiah Awang Biak, PhD

Fakulti : Kejuruteraan

Sintesis nanotiub karbon (CNTs) dilaksanakan secara meluas pada suhu tinggi. Melalui kaedah pemangkin terapung pengendapan wap kimia. Ini adalah bertujuan untuk mengurangkan suhu bagi sintesis nanotiup karbon (CNTs) selain untuk memberikan pengawalan keadaan reaktor yang lebih efisien serta mengurangkan pembentukan karbon ketulenan. Tujuan utama penyelidikan ini adalah untuk (i) mensintesis nanotiub karbon pada suhu rendah dengan menggunakan yang telah ditingkatkan (ii) menyelidik kesan suhu pra-pemanasan dalam proses sintesis nanotiub karbon dan (iii) menghasilkan profil suhu dan halaju (tanpa tindak balas) bagi bendalir di dalam sistem tersebut dengan menggunakan perisian simulasi.

Sintesis proses dibuat pada suhu antara 500^{o}C – 600^{o}C dengan kenaikan pada setiap 10^{o}C. Suhu pra-pemanasan diubah antara 150^{o}C – 400^{o}C dengan kenaikan pada setiap 50^{o}C. Operasi proses dilaksanakan pada 1 atm dan semua bahagian yang terdedah ditebat menggunakan wul kaca disaluti foil aluminium. Alat monitor suhu digunakan

untuk merekod profil suhu di dalam reaktor. Profil ini dapat member panduan masa yang tepat untuk memanaskan pemangkin dan memulakan proses tindakbalas. Sumber haba tunggal digunakan untuk memanaskan reaktor danpemangkin. Perisian COMSOL Multiphysics digunakan untukmenghasilkan simulasi bagi membetuk profil suhu dan halaju bendalir di dalam system ini dalam keadaan tanpa tindakbalas. Morfologi dan struktur dalam CNTs yang dibentuk telah dianalisa menggunakan Mikroskop Pengimbas Elektron (SEM), Mikroskop Pemancar Elektron (TEM) dan Mikroskop Pemancar Elektron Resolusi Tinggi (HR-TEM). Kestabilan terma dan ketulenan produk yang dihasilkan telah dianalisa menggunakan Analisis Gravimetri Terma (TGA).

Hasil penyelidikan menunjukkan bahawa CNTs boleh dihasilkan pada suhu 530^{o}C – 600^{o}C. Kualiti dankuantiti produk yang bertambah dengan kenaikan suhu sintesis. Kuantiti tertinggi dihasilkan apabila proses dijalankan pada suhu 300^{o}C untuk pra-pemanasan dan 600^{o}C untuk sintesis. Walaubagaimanapun, ketulenan tertinggi dicapai apabila suhu pra-pemanasan sistem ini ditukar kepada 200^{o}C. CNTs yang tersusun secara sejajar telah didapati apabila suhu pra-pemanas diubah kepada 400^{o}C dan hanya berlaku pada suhu sintesis antara 570^{o}C – 580^{o}C. Penggunaan pemanasan tunggal untuk reactor dan pemangkin telah membantu dalam pembentukan CNTs sejajar pada suhu sintesis iditu 550C. Simulasi proses yang menghasilkan profil suhu dan halajau bendalir di dalam sistem ini telah dapat mengenalpasti beberapa seksyen yang sejuk atau mempunyai kepelbagaian suhu.Keadaan ini telah member kesan kepada pembentukan CNTs di bahagian tersebut. Sebagai kesimpulan, penyelidikan ini telah berjaya menghasilkan nanotiub karbon padasuhu yang rendah. Kualiti dan kuantiti produk yang dihasilkan adalah berkadaran dengan suhu pra-pemanasan dan juga sintesis.

ACKNOWLEDGMENTS

In the name of Allah, the Most Beneficent, the Most Merciful

Thanks God for helping me out while I was about to give up and His unbelievably close support any time I needed.

I hereby convey my sincere gratitude and appreciation to my venerable supervisor Dr. Dayang Radiah Awang Biak, who gave me invaluable and worthwhile advice on this research, and gave me a panorama of observance into the knowledge in my field.

I also would like to appreciate the efforts of the honorable Dr. Zurina Zainul Abidin for her supports with fruitful information and giving me a vast visibility in the world of knowledge.

I thank Professor Dr. Fakhru'l-Razi Ahmadun for his providing me with fruitful information. I also thank all other faculty members who somehow helped me prepare this assignment. Deep thankful to our beautiful university (UPM) for the encouragement and support.

Last but not least, I would like to express my gratitude to my brothers and sisters, with their guidance, supports, love and encouragement.

I certify that a Thesis Examination Committee has met on 22/4/2010 to conduct the final examination of Mahmood Rashid Atiyah on his thesis entitled "**Low- temperature synthesis of carbon nanotubes via floating catalyst chemical vapor deposition method**" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the University Putra Malaysia [P.U.(A)] 15 March 1998. The Committee recommends that the Student be awarded the relevant Master of Science.

Members of the Thesis Examination Committee were as follows:

Norhafizah Abdullah, PhD
Associate Professor
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Suraya Abdul Rashid, PhD
Senior Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Mohd Amran Mohd Salleh, PhD
Senior Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Subhash Bhatia
Professor
Faculty of Engineering
Universiti Sains Malaysia
(External Examiner)

BUJANG KIM HUAT, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

Dayang Radiah Awang Biak, PhD
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Zurina Zainal Abidin, PhD
Faculty of Engineering
Universiti Putra Malaysia
(Member)

HASANAH MOHD GHAZALI, PhD
Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 10 June 2010

DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations, which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

_____

MAHMOOD RASHID ATIYAH

Date:

TABLE OF CONTENTS