

**EFFECT OF CATALYSTS AND PRECURSORS IN THE SYNTHESIS OF
CARBON NANOTUBES BY CATALYTIC CHEMICAL VAPOUR DEPOSITION**

YUSRAN SULAIMAN

UNIVERSITI TEKNOLOGI MALAYSIA

EFFECT OF CATALYSTS AND PRECURSORS IN THE SYNTHESIS OF
CARBON NANOTUBES BY CATALYTIC CHEMICAL VAPOUR DEPOSITION

YUSRAN SULAIMAN

A thesis submitted in fulfilment of the
requirements for the award of the degree of
Master of Science (Chemistry)

Faculty of Science
Universiti Teknologi Malaysia

JULY 2006

Dedicated to my beloved father, mother and family ...

*Allah tidak akan mengubah nasib sesuatu kaum
melainkan kaum itu sendiri yang mengubahnya...*

ACKNOWLEDGEMENT

In The Name of Allah, Most Gracious and Most Merciful

I would like to take this opportunity to express my token of appreciation to those who have helped me a lot throughout this project. First, I would like to express my sincere thanks to Assoc. Prof. Dr. Nor Aziah Buang and Dr. Zaiton Abdul Majid for their encouragement, help, advice, guidance, constructive criticism and valuable suggestion besides sharing their knowledge with me.

I would like to acknowledge Dr. H.Kataura for the attachment program in AIST, Tsukuba, Japan and discuss my research result. I am also grateful to Mr. M.Suzuki, Mr. S.Yoshioka and Mr. T.S.Tan and NanoProcessing Partnership Program (NPPP) staff for their help in TEM work and training.

I also want to express my appreciation to all CNT members such as Prof. Dr. Ahmad Fauzi Ismail, Mrs. Suhaila, Razis, Tee Jia Chee, Azni and Mohd Nuruddin for their help, support, sharing the knowledge and useful discussion throughout this project. Special thanks to Mr. Jefri for an easy access to the SEM analysis. Finally to my family and those who helped me directly or indirectly. This research would not have been possible to complete without the support from everyone and each one of you.

Once again

Thank you

ABSTRACT

Since the pioneering report of discovery of carbon nanotubes (CNTs) in 1991 by Iijima, scientists and researchers worldwide have carried out in depth investigations in this new family of carbon because of its myriad properties and potential applications. The synthesis of novel nanoscale material is the main target in current material science. This study investigates the effect of different types of carbon source and catalyst on the type of CNTs formed via catalytic chemical vapour deposition (CCVD) method. Three types of carbon source *i.e.* acetylene, methane and ethanol were used for the synthesis of CNTs. The catalysts used in the synthesis of CNTs are monometallic, bimetallic and trimetallic derived from Fe, Co and Ni salts using wet impregnation method. The catalysts were characterized by scanning electron microscope (SEM) and energy-dispersive X-ray analysis (EDX). The analysis confirmed the presence of Fe, Co and Ni. The as-synthesized CNTs were characterized using SEM/field emission-scanning electron microscope (FE-SEM), EDX, Raman spectroscopy and transmission electron microscopy (TEM). This analysis also confirmed that all the prepared catalysts were active for the production of CNTs. SEM/FE-SEM analysis revealed different morphologies of CNTs were formed when different catalysts and carbon source were used. Raman spectra revealed that acetylene and methane precursor produced multi-walled carbon nanotubes (MWNTs) as indicated by the presence of G-band and D-band peaks. However, their structures were different depending on the catalysts used. Meanwhile, the presence of RBM peaks along with the G-band and D-band revealed that single-walled carbon nanotubes (SWNTs) are produced using ethanol as the carbon source. TEM micrographs obtained confirmed that acetylene and methane produced MWNTs and ethanol produced SWNTs with diameter in the range of 14.74-34.59 nm, 10.19-37.61 nm and 0.96-2.52 nm, respectively. However, Fe/Al₂O₃ catalyst selectively produced double-walled carbon nanotubes (DWNTs) when ethanol was used as the carbon source. Generally, this research has been successful in producing various types of CNTs depending on the catalyst used and the carbon source selected.

ABSTRAK

Sejak daripada penemuan tiubnano karbon (CNT) yang pertama oleh Iijima, ramai saintis dan penyelidik serantau dunia sangat berminat untuk mengkaji dengan lebih mendalam kumpulan carbon yang baru ini kerana sifat dan aplikasinya yang sangat meluas. Sintesis bahan baru dalam skala nano merupakan tumpuan utama dalam kajian sains bahan sekarang. Kajian ini menyelidik kesan penggunaan pelbagai sumber karbon dan mangkin kepada jenis CNT yang terbentuk melalui kaedah pemangkinan pemendapan wap kimia (CCVD). Tiga jenis sumber karbon iaitu asetilena, metana dan etanol digunakan untuk sintesis CNT. Mangkin yang digunakan dalam sintesis CNT ialah monologam, dwilogam dan trilogam yang dihasilkan daripada garam Fe, Co dan Ni dengan menggunakan kaedah pegisitepuan basah. Mangkin dicirikan dengan mikroskopi imbasan electron (SEM) dan analisis penyerakan tenaga sinar-X (EDX). Analisis ini menunjukkan kehadiran Fe, Co dan Ni dalam sampel mangkin. CNT yang disintesis dicirikan dengan SEM/mikroskopi imbasan elektron-sinaran medan (FE-SEM), EDX, spektroskopi Raman dan mikroskopi elektron penyerakan (TEM). Analisis SEM/FE-SEM menunjukkan pelbagai morfologi CNT terhasil apabila pelbagai mangkin dan sumber karbon digunakan. Analisis ini juga membuktikan bahawa semua mangkin yang disediakan aktif terhadap penghasilan CNT. Spektrum Raman menunjukkan bahawa asetilena and metana menghasilkan tiubnano karbon dinding berganda (MWNT) seperti yang ditunjukkan dengan kehadiran puncak jalur G dan jalur D. Walaubagaimanapun, strukturnya berbeza bergantung kepada mangkin yang digunakan. Sementara itu, kehadiran puncak RBM bersama-sama dengan jalur G dan jalur D menunjukkan tiubnano karbon berdinding tunggal (SWNT) dihasilkan apabila etanol digunakan sebagai sumber karbon. Mikrograf TEM yang diperolehi membuktikan bahawa asetilena and metana menghasilkan MWNT dan etanol manghasilkan SWNT dengan diameter 14.74-26.08 nm, 10.19-37.61 nm and 0.96-2.52 nm. Walaubagaimanapun, mangkin Fe/Al₂O₃ secara selektifnya manghasilkan tiubnano karbon berdinding dua (DWNT) apabila etanol digunakan sebagai sumber karbon.