UNIVERSITI PUTRA MALAYSIA

SYNTHESIS AND CHARACTERZATION OF CARBON NANOTUBE AND GRAPHENATED CARBON NANOTUBE SUPERFIBRE VIA FLOATING CATALYST CHEMICAL VAPOUR DEPOSITION METHOD

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By

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November 2020

Chairman : Ismayadi bin Ismail, PhD
Institute : Institute of Advanced Technology

The objective of this research is to develop carbon nanotubes (CNTs) sheet superfibre materials with comparable or exceed properties of existing ones. Superfibres is formed by synthesizing long CNTs using direct-spinning method in horizontal furnace and make them into sheet form. Presently, the superfibres has modest properties but potentially to be strongest fiber known and commonly synthesized using arrays of aligned CNTs and limited studies using direct-spinning method. The interest of producing CNT sheet superfibres is due to no ready industrial format of macroscopic CNTs, since individual CNTs cannot grow longer. Hence, one of the proposed methods to overcome this hindrance is to assemble CNTs into continuous fibres as nanotube superfibre material. Many routes to attain this nanotube superfibre material with outstanding properties but mostly through the post-processing method. In this study, macro assembly of CNTs sheet superfibres was achieved directly from floating catalyst chemical vapour deposition process without post-processing methods. Herein, our interest is to synthesis CNTs and graphenated CNTs sheet superfibres and study their properties. The CNT sheets were directly spun from a hot reactor of the horizontal furnace using floating catalyst chemical vapour deposition. To obtain CNT sheets, three main parameters were used as variables which were reaction temperature (from 1050 °C to 1250 °C), injection rate of precursors (1 ml/hr to 20 ml/hr), and gas flow rate of hydrogen gas (200 sccm to 400 sccm). The spinnability of CNT sheets was observed to highly dependable on the gas flow rate of carrier gas followed by other parameters. The morphological characteristics of CNT sheets showed the synthesized CNTs were multi-walled with a diameter of 17 nm to 49 nm. It was also revealed that, high reaction temperature at 1250 °C and low gas flow rate of hydrogen, (200 sccm and 250 sccm) led to the formation of g-CNTs. TGA analysis showed that high decomposition temperature indicated the presence of multi-walled CNT with purity as high as 98%. Furthermore, high degree of graphitization of the CNT sheets increases with increases of temperature. The highest graphitization of CNT sheet was obtained at temperature 1200 °C with a ratio of I_D/I_G of 0.37. Besides, the bulk conductivities of

CNT sheets were measured and found that CNT sheet synthesized at 350 sccm has highest electrical conductivity due to the highest packing density of CNTs with a value of 10.72 S cm^{-1}. This research is important because it will enable industries to manufacture new fiber materials which will change engineering designs of potential application such as supercapacitor.

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

SINTESIS DAN PENCIRIAN *SUPER*-SERAT KARBON NANOTIUB DAN PENGRAFENAN KARBON NANOTIUB MELALUI KAEDAH PEMANGKIN APUNGAN PEMENDAPAN WAP KIMIA

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Objektif penyelidikan ini adalah untuk membangunkan serat lembaran karbon nanotuib. dengan sifat-sifat setara atau mengatasi yang sedia ada. *Super*-serat nanotiub dibentuk dengan sintesis KNTs panjang menggunakan kaedah putaran langsung dalam relau memanjang dan dibuat dalam bentuk lembaran. Pada masa kini, *super*- serat menpunyai sifat-sifat yang sederhana tapi berpotensi menjadi serat yang kuat dan kebiasaannya disintesis menggunakan KNTs susunan berjajar dan kajian terhad menggunakan kaedah putaran langsung. Kecenderungan untuk menghasilkan lembaran serat KNTs kerana ketiadaan format tersedia makroskopik CNTs, kerana individu KNTs tak boleh tubuh secara panjang. Oleh itu, salah satu cara dicadangkan adalah untuk mengatasi halangan untuk himpunan KNTs kepada serat berterusan sebagai bahan *super*-serat. Pelbagai cara juga telah dibangunkan untuk mencapai sifat luarbiasa ini tetapi kebanyakan melalui keadah rawatan-selepas. Dalam kajian ini, himpunan makro serat lembaran karbon nanotuib dicapai secara langsung menerusi proses pemangkin apungan pemendapan wap kimia tanpa kaedah rawatan-selepas. Di sini, adalah menjadi tujuan untuk sistesis lembaran KNTs dan g-KNT dan mengkaji sifat-sifatnya. Lembaran KNT itu diputar daripada rektor panas relau melintang menggunakan keadah pemangkin apungan pemendapan wap kimia. Untuk mendapatkan lembaran KNTs, tiga parameter utama telah digunakan sebagai pembolehubah iaitu suhu tindakbalas, (dari 1050 °C hingga 1250 °C), kadar suntikan prekursor (1 ml/hr hingga 20 ml/hr), dan kadar aliran gas hidrogen (200 sccm hingga 400 sccm). Kebolehan-berputar lembaran KNT didapati sangat bergantung kepada kadar aliran gas pembawa, diikuti oleh parameter yang lain. Ciri-ciri morfologi lembaran KNT menunjukkan KNT yang disintesis merupakan KNT yang berbilang dinding berdiameter di antara 17 nm hingga 49 nm. Ianya juga menunjukkan bahawa, suhu tindak balas yang tinggi iaitu 1250 °C dan kadar aliran gas hidrogen yang rendah, 200 sccm dan 250 sccm menyebabkan pembentukan karbon nanotiub bergrafen (g- CNT). Analisis TGA menunjukkan suhu penguraian yang tinggi menyatakan kehadiran KNT berbilang dinding dengan ketulenan setinggi 98%. Tambahan lagi, darjah grafitisasi lembaran KNT meningkat dengan peningkatan dalam

suhu tindakbalas sehingga 1200 ^{o}C. Grafitisasi tertinggi lembaran KNT diperolehi suhu 1200 ^{o}C, dengan nisbah I_D/I_G adalah 0.37. Sebagai tambahan, konduksi lembaran KNT diukur dan didapati lembaran KNT yang disintesis pada 350 sccm mempunyai kekonduksi elektrik yang tertinggi kerana ketumpatan KNTs dengan nilai 10.72 S cm^{-1}. Penyelidikan ini adalah penting kerana ianya akan membolehkan industri untuk membuat bahan serat baru yang akan mengubah reka bentuk kejuruteraan untuk potensi aplikasi seperti *super*-kapasitor.

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LIST OF SYMBOLS AND ABBREVIATIONS

C_2H_2	Acetylene
CNS	Carbon nanosheets
CNT	Carbon nanotubes
CVD	Chemical vapour deposition
DWCNT	Double walled carbon nanotubes
eV	Electron volt
EDX	Energy dispersive X-ray
C_2H_5OH	Ethanol
$C_{10}H_{10}Fe$	Ferrocene
FESEM	Field emission scanning electron microscopy
FCCVD	Floating catalyst chemical vapour deposition
FWHM	Full width at half maximum
GPa	Gigapascal (Standard unit of pressure)
G-CNT	Graphenated carbon nanotubes
HRTEM	High resolution transmission electron microscopy
K	Kelvin (Standard unit of temperature)
MWCNT	Multi-walled carbon nanotubes
Ω/sq	Sheet resistance
S cm^{-1}	Siemen per centimeter
SWCNT	Single-walled carbon nanotubes
Sccm	Standard cubic centimetres per minutes
TGA	Thermogravimetric analysis
C_4H_4S	Thiophene
VLS	Vapour-solid-liquid

VSS	Vapour-solid-solid
VACNT	Vertically aligned carbon nanotubes
wt. %	Weight percentage

CHAPTER 1

INTRODUCTION

1.1 General Overview

Carbon nanotubes (CNTs) was first discovered by Iijima in 1991, and this discovery piques great interest among researchers to date due to their important physical properties such as mechanical strength, electrical and thermal conductivities (Baughman et al., 2002). Although CNTs exhibit excellent features, however, the ability to transfer the mechanical and electrical properties of individual CNTs into macroscopic assemblies is still a challenge to the research world. Presently, the general synthesis methods of CNTs include arc discharge, laser ablation, floating catalyst, and chemical vapour deposition. In this work, a floating catalyst chemical vapour deposition (FCCVD) method was used where catalyst travels through a quartz reactor,and resulted in the nucleation of CNTs. Usually, ferrocene is used as an iron catalyst, while sulfur is used as a reaction promoter. This method provides high yield, material efficiency, and continuous production. Thus, large scale production is easier to achieve (Su, 2015; Ma et al., 2016; Hoecker et al., 2017). This method is chosen used due to its simplicity as a single-step synthesis method production of CNT sheets superfibre with high capability of producing a continuous collection of CNTs in macroscale formfor industrial. Most studies of CNT sheets superfibre have been carried out using powder-like CNTs or supported on different substrates. Meanwhile, to our best knowledge limited studies on sheet form of CNT using FCCVD which usually focused on fibre form of CNT.

In comparison to FCCVD, directly synthesized CNT sheets in gaseous feedstock mixture formed in a quartz tube are seldom discussed. Currently, catalytic chemical vapour deposition (CVD) method has become a popular choice in both research and industry field due to its many advantages such as simple processing, low cost, high degrees of control, and scalability (Hoecker et al., 2016). Large scale production of macroscopic assemblies of CNTs for industrialization is essential for industrial applications of CNTs.

The current issues is to control CNT synthesis and processing to fabricate CNT superfibre material with properties that exceed those existing fiber materials. The technical challenge for manufacturing of CNTs superfibers is defect occur during growing long nanotubes. The properties of the superfibre material is relatively low and the process is not continuous and repeatable. Hence, technical efforts and improvements are needed to produce extraordinary properties of short nanotubes to bulk fibrous materials. To remove the defect during process by controlling processing conditions. The fabrication of macroscopic assemblies or superfibre material such as sheets and fibres is an essential approach toward industrial applications (Schulz et al.,2013).

However, there is the complexity to grow macroscopic assemblies of these long continuous CNTs, and the growth of ultralong carbon nanotubes (>1 mm) required basic

comprehension of experimental factors on CNTs growth (Joshi, 2010). This ultralong CNTs is also defined as superfibre material, whereby include both CNT aerogel and CNT sheet in this study. The ultralong CNTs were first proposed by Kim et al. (2002), where ultralong single-walled carbon nanotube (SWNT) with a length of 0.6 millimetres was grown from the mixture of methane and ethylene carbon source in chemical vapour deposition CVD. In order to grow growing ultralong CNTs, lifetime of catalysts are one of the crucial aspects to consider, while hydrogen is used during the growth process to keep the activity of catalysts for a long time to get macroscale long CNTs (Zhang et al., 2014).

A number of establish research have been developed in the literature regarding CNT superfiber. A research team led by Koziol et al., (2007) where he used direct-spinning techniques, where high speed ring machine is used and to develop CNT fibre, a form of superfibre material. Another research by Hou et al., (2017) study on formation of CNT sheet at high temperature, injection rate and gas flow rate. Apart from that, Wang and researchers studied on CNT superfibers include CNT fibre and CNT sheet and their properties (Wang et al., 2014; Luo et al., 2016). The studies discussed on properties of macro scale CNTs however, lack of understanding of mechanism on formation of ultra-long CNTs and their morphological and structural study. Hence, the effect of main variables were manipulated to discussion their mechanism and their effect on formation of g-CNTs in sheet superfibre form.

CNT sheets superfibre is a macro-assembly of individual CNTs in the dimension (2D) layer of bulk form. The term ‘sheets’ is referring as films, flat papers made of CNTs with their dimension arranged in the plane of papers and along the length of sheets (Lekawa-Raus et al., 2014). Meanwhile, superfibre material is long and continuous fibrous material such as ribbon, yarns and sheet, which has length > 1 mm (Joshi, 2010; Schulz et al., 2014). This CNT sheet is a lightweight material with remarkable properties. Thus, it is makes a great interest to fabricate it on a larger scale. Manufacturing aligned 2D sheets is worthy due their long individual nanotubes characteristic.

To date, there are three methods to synthesis ultralong CNT sheets superfibres. The first method is spinning from CNT solutions such as vacuum filtration and solution. Secondly, CNT sheets superfibres can be produced via the spinning of vertically aligned CNT array. The last method is the spinning of CNT aerogel which is formed in a high temperature reactor. The advantages and disadvantages of the methods are summarized in table 1.1.

Table 1.1: Methods to synthesis CNT superfibers and their advantages and disadvantages.

Method	Steps	Advantages	Disadvantages
Spinning from CNT solutions	Consists of more than two steps process; 1) synthesis of nanotubes and 2) combining them into macroscopic sheets followed by pressing treatment via vacuum-filtered buckypapers to densify it into 2D layer structure bulk form (Xu et al., 2016).		Limited due to short CNTs and residual micro molecular surfactants on the tube surface, a random orientation, and weak intertube van der Waals interaction which lead to low mechanical and physical properties. The highest electrical conductivity measured by filtered MWCNT buckypapers is only at 72.40S cm^{-1} (Mansfield et al., 2015).
Spinning from vertically aligned CNT array	1) CNT array was synthesis in chemical vapour deposition process. The process directly spun from CNT array and result in CNT sheets.	High electrical conductivity along the CNTs measured at room temperature with a measurement of $10^2 – 10^3$ S cm^{-1}, owing to their highly aligned orientation of the CNTs (Yang et al., 2011).	The production of nanotube sheets is limited by the volume of the corresponding array produced.
Spinning of CNT aerogel	1) Directly synthesis macro-assembly of CNT sheets using a rotating spindle attached at the end of the furnace to collect the CNTs.	Single step process where synthesized CNT is collected to therotating spindle as CNT sheets. Depending on the geometries of rotating spindle. The CNT sheets could reach up to 2026 S cm^{-1} (Liu et al., 2011).	

Continuous improvements on macroscopic fibres made up of CNTs are necessary for future performance maximization and for the potential of CNT assemblies for high end uses. The applications include energy storage systems with higher efficiency and higher density. These CNTs have high potential performance in energy storage due to1 TPa stiffness and tensile strains as high as 6%. While using theoretical stress-strain models, strain in defect-free CNTs is predicted to have a maximum energy density (energy stored in system per unit volume or mass) of 7.7×10^6 kJ, which is three orderslarger a magnitude than steel (Wu and Wang, 2016). Apart from that, CNTs combine properties of best materials from polymers, carbon fibres, and metals. The unique properties of CNTs, including high tensile strength at 50 GPa and 11 – 63 GPa for individual single-walled CNTs and multi-walled CNTs, respectively. Other properties include high electrical conductivity, 10^6 S cm^{-1} for single-walled CNTs and 3×10^4 S cm^{-1} for multi-walled CNTs (Yakobson et al., 1997; Collins and Avouris, 2000; Yu etal., 2000., Li et al., 2007; Motta et al.,2007). These excellent properties can be obtained, when CNTs are assembled into fibres material such as sheets and fibres for real application and still a great challenge for researchers.

1.2 Problem Statement

In the 2010s, interest in macroscopic fibres of CNTs brought new interest because of their strength, electrical conductivity, and thermal conductivity. The developments on the synthesis of high-quality CNTs and the assembly of macroscopic CNT aligned and fibres provide advances in properties. However, information on the dependence of macroscopic properties on intrinsic CNTs such as diameter, length, number of walls, graphitic, and purity of CNTs is limited.

Research related to the synthesis of CNTs almost come to a saturation where thousands of literature were reported. Many methods were proposed, and parameters tested to obtain useful CNTs. Even though individual CNTs displays magnificent and impressive properties over other nanomaterials, improvement still needed for industrial application. The CNTs possesses terrific properties at the nano level, but when it was turned into a bulk form consist of sheet, fibre, cotton, and others, the properties would deteriorate which deserved further study to improve its functional properties. This nanomaterial does not fully utilize into the consumer market due to one problem: there is limited macro-assemblies CNT superfibre which makes it difficult for the industry player to adopt in their application. Therefore a method is required to synthesis a bulk form of CNTs in a ready format for the industry. Macroscopic CNT assemblies with controlled orientation and configurations such as 1D CNTs fibre, 2D CNTs sheet/film, and 3D aligned CNTs array is another approach to employ instead of pursuing chirality and defect-free CNTs to obtain excellent electronic properties (Zhang et al., 2015). Thus simple fabrication processes to produce these macroscopic assemblies such as CNT fibres and CNT sheets are essential study.

Apart from that, these macroscopic assemblies of CNTs have longer nanotubes, thus able to overcome the disadvantage of short CNTs and able to fill void since it can entangled and form long CNT. The disadvantage is due to short CNTs required high

concentrations to form a network within other materials. CNT superfibres fabricated from the wet spinning method own numerous short CNTs since starting material typically CNT powders. While, CNT sheets/fibres from aligned array have limited size and length of CNT sheets and usually too fragile to handle on traditional textile equipment (Wu and Wang, 2016). Thus, a direct spinning method from FCCVD enables overcome these limitations since it can produce CNTs continuously based on synthesis parameters in mass production.

In this work, CNT sheets were synthesized using the FCCVD method where the direct spinning process was involved. The common problems when synthesizing the CNT sheets using this method are discontinuity of formation of aerogel, thus understanding on growth mechanism and their properties are necessary for this field. This failure of aerogel formation happened due to many factors that are interlinked to each other hence it is quite a challenging and complicated subject. The significance of this study will lead to further research projects involving CNT sheets superfibre as starting material for numerous potential applications on a large scale.

1.3 Objectives of the Study

This research is carried out based on several objectives that are mentioned below:

I. To synthesis CNT sheet superfibres in a single-step process using a floating catalyst chemical vapour deposition method.
II. To optimize the reaction temperature, the injection rate of precursors, and gas flow rate of hydrogen gas on morphological, structural, and electrical properties of synthesized CNT sheet superfibres.
III. To investigate the effect of synthesis parameters such as reaction temperature, the injection rate of precursors, and the gas flow rate of hydrogen gas on g-CNT sheet superfibres formation.

1.4 Scope of the Study

In this study, CNT sheets were synthesized in a single-step process using FCCVD method where the ratio of precursors was fixed at 96.4 wt. % of ethanol, 2.4 wt. % of ferrocene and 1.2 wt. % of thiophene meanwhile other parameters were varied including reaction temperature, (in the range of 1050 °C to 1250 °C), injection rate (from 1 ml/hr to 20 ml/hr), and hydrogen gas flow rate (from 200 sccm and 400 sccm).CNT sheets were synthesized and continuously spun onto a spindle using these variables parameters. The probabilities of CNT sheets to be continuously spun were observed and characterized using field emission scanning electron microscopy (FESEM), high resolution transmission electron microscopy (HRTEM), thermogravimetric analysis (TGA), Raman spectroscopy and its electrical conductivity was measured using a surface resistivity meter. The analyses were carried out, and theobtained results were discussed in detail.

1.5 Chapter Organization

This thesis consists of five chapters, including introduction, literature review, methodology, results and discussion, and conclusion. The first chapter is the introduction of this research include problem statements, the objectives, and the scope of the study conducted. In the second chapter, study related to CNT superfibres were reviewed. The third chapter focuses on methods in this work, where procedures, methods to synthesis CNT sheets superfibres, and their characterization include FESEM, HRTEM, TGA, Raman spectroscopy, and surface resistivity meter are stated. Then, chapter four discussed the obtained CNT sheets, and their results based on characterization are analyzed and discussed. Lastly, chapter five concludes the results acquired in this work and the recommendations for future research.

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