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SYNTHESIS OF ZEOLITE TEMPLATED GRAPHENE (ZTG) FROM METHANOL VIA CHEMICAL VAPOR DEPOSITION (CVD) METHOD

MOHD SYAFIQ BIN ELIAS

(MS112098)

ASSOC. PROF. DR. NOR AZIAH BUANG

DEPERTMENT OF CHEMISTRY
FACULTY OF SCIENCE
UNIVERSITI TEKNOLOGI MALAYSIA

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For my respectful supervisor, my beloved mother and father, my siblings andmy best friend forever

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ABSTRACT

A new synthesis route in the production of graphene by template synthesis technique using zeolite as the host materials has successfully produced graphene. The highly regular ordered and highly crystalline structure of zeolite was successfully utilized for the formation of ordered sp^2 graphitized graphene structure in the zeolite porous framework. Graphitic carbon structure of zeolite template graphene (ZTG) has been synthesized via catalytic chemical vapor deposition (CVD) method from methanol as the carbon precursor. The influence factors of types of zeolite used in the template synthesis and CVD reaction temperatures have been investigated to obtain the optimum experimental condition for producing high quality of ZTG. The results show acid sites of the zeolite plays an important role in the synthesis of ZTG in porous framework of zeolite structure. CVD reaction temperatures at 500°C is considered as the best reaction temperature for the production of graphene using zeolite as template with high quality of carbon graphitic structure. UV- Visible spectroscopy and Raman spectroscopy analysis further proven the existence of sp^2 character of graphene structure with small amount of defect in the ZTG produced.

ABSTRAK

Kaedahsintesisbaruuntukmenghasilkangrafindengantekniksintesisacuanmenggunakan zeolite sebagaibahanacuantelahberjayamenghasilkangrafinberkualititinggidenganjumlahkuantiti yang mencukupi.Struktur yang sangattersusundanstrukturhablur zeolite telahberjayadigunakanuntukmembentukstruktur sp^2karbongrafin yang tersusun di dalamstrukturbingkaianberliang zeolite. Strukturkarbongrafinacuan zeolite (ZTG) telahdisintesisdengankaedahpemendapanwapkimia (CVD) daripada methanol sebagaisumberkarbon.Faktorpengaruhjenis zeolite yang digunakandalamtekniksintesisacuandansuhutindakbalas CVD telahdikajiuntukmendapatkankeadaaneksperimen yang optimum untukmenghasilkan ZTG yang berkualititinggi.Keputusaneksperimenmenunjukkanbahagianasid zeolite memainkanperananpentingdalamsintesis ZTG di dalamstrukturbingkaianberliang zeolite.Suhutindakbalas CVD pada 500°C dianggapsebagaisuhutindakbalas yang terbaikuntukmenghasilkangrafinmenggunakan zeolite sebagaibahanacuandenganstrukturkarbon yang berkualititinggi.Analisis UV spektroskopidan Raman spektroskopimembuktikanlagikewujudankriteriasp^2strukturgrafindengansedikitkecacatandalam ZTG yang dihasilkan.

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LIST OF ABBREVIATIONS

ZTG	-	Zeolite Template Graphene
GO	-	Graphene Oxide
CVD	-	Chemical Vapor Deposition
AFM	-	Atomic Force Microscopy
FESEM	-	Field Emission Scanning Electron Microscopy
FTIR	-	Fourier Transform Infra-Red
TGA	-	Thermogravimetric Analysis
XRD	-	X-ray Diffractometer
XPS	-	X-ray Photoelectron Spectroscopy
TEM	-	Transmission Electron Microscopy
BET	-	Brunauer-Emmett-Teller
DME	-	Dimethyl Ether
NaOH	-	Sodium Hydroxide
HCl	-	Hydrochloric Acid

CHAPTER I

INTRODUCTION

1.1 Introduction

In the latest rising frontier materials in the area of materials science research since its discovery in 2004 [1], graphenes have attracted an intensive intention in recent years [2]. A novel two-dimensional [3] graphitic honeycomb lattice carbon structure [4] of graphene with sp^2 hybridization[5, 6] which only one atom thick [2, 3] possesses very fascinating electronic, thermal and mechanical properties [7]. Graphene has ultrahigh electron mobility[8], zero electronic band gap with [4], tunable band gap, quantum electronic transport[8], high thermal stability and conductivity[9], high elasticity, and electromechanical modulation[8].

In addition to its remarkable and outstanding sheets resistance, ballistic transport, fascinating flexibility[4] and transparency of graphene-based thin film[10] have raised the eyebrows of the researchers in applying this unique special material in advanced technologies such as nanoelectronics, transparent conducting electrodes, nanocomposites, supercapacitors, sensors, batteries, and other area of technologies [4]. The encouraging aspects of graphene is to use it in the touch panels in electronic devices, e-paper, organic light-emitting display (OLED), and solar cells[4].

From the discoveries of graphene until now, many researchers have done several attempts especially in preparing high quality of graphene and higher yield with low cost operation procedures [4]. In addition, the outstanding properties of graphene are hugely depending on the synthesis method, surface roughness and also graphenesubstrate interaction [8, 11]. Recently, the synthesis of graphene in large scale have been reported including liquid suspension graphene oxide (GO) followed by reduction by chemical treatment[3], thermal reduction of graphite oxide, epitaxial growth on SiC, liquid-phase exfoliation by organic solvents, epitaxial growth by chemical vapor deposition (CVD), solvothermal synthesis, and unzipping of carbon nanotubes [4, 12].

Besides that, despite the general top-down approach of preparation of graphene, some researchers also have introduced the bottom-up chemical approach towards the synthesis of the graphene-based film. It has been achieved by the thermal reaction of synthetic nano-graphene molecules of giant polycyclic aromatic hydrocarbons (PAHs) which are cross-linked together and fused further to form larger graphene sheets [13].

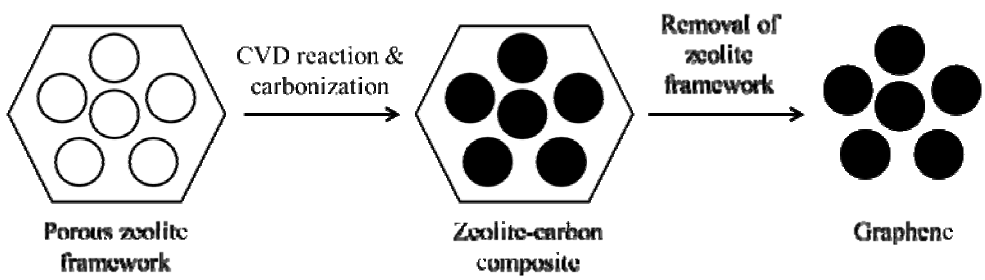

Figure 1.1: The steps involved in synthesis of graphene in zeolite framework

Herein different approach has been done in order to prepare the remarkable unique graphene materials. In this research, the graphene was synthesized inside some host materials using chemical vapor deposition (CVD) method from alcohol as the carbon precursor. Several host materials which is Zeolite H-Y type, Zeolite H-SAPO-34 (SA001) and Zeolite H-SAPO-34 (SA002) have been used to synthesis zeolite template graphene (ZTG) via CVD method. Those zeolite host materials act as the template for the growth of graphene as shown in Figure 1.1. Different CVD reaction temperatures that contribute to the preparation of graphene in zeolite pores have also been studies in this research.

1.2 Problem Statement

As describes before, the encouraging part of the use of graphene is its application in touch panel electronic devices. A transparent electrically conductive material is one of the critical components as electronic devices specifically in photoelectronic devices [9]. Presently, the state-of-the-art in transparent conductive electrode uses the indium tin oxide (ITO) due to its high electrical conductivity and optical transparency [10].

However, ITO appears to be increasingly problematic because of incompatibility with large scale productions[5].Besides that, the ITO becomes increasingly expensive due to limited availability of the indium element on earth[6, 13]. Furthermore, the devices problematic like susceptible ion diffusion into the polymer layers and mechanical brittleness limit the applicability of ITO as the flexible photoelectronic devices[5].Thus, thin graphene-based film is one of the best candidates which offer similar or better performances as ITO with cost-efficient method as the transparent conductive material.

Besides that, the problem statement of this research is the possibilities to synthesis zeolite template graphene (ZTG) in zeolite pores as the host materials via CVD method approach. Different host materials will leads to different physical and chemical properties of graphene produced. The CVD reaction parameters also play important roles in synthesizing graphene in the zeolite host materials. In this research, reaction parameters of CVD system which is the reaction temperatures have been studied to obtain better quality of graphene.

1.3 Objectives

The objectives of the research are:

1. To synthesis zeolite template graphene (ZTG) from different types of zeolite via chemical vapor deposition (CVD) method.

2. To investigate the effect of reaction temperature in the synthesis of zeolite template graphene (ZTG) during chemical vapor deposition (CVD) process.

These objectives will lead to find the best condition for producing high quality graphene sheets and transparent graphene film.

1.4 Scope of Study

Different types of zeolites were chosen as the host materials to synthesis the zeolite template graphene (ZTG) in the zeolite pores materials which is Zeolite H-Y type, Zeolite H-SAPO-34 (SA001) and Zeolite H-SAPO-34 (SA002). The synthesis of ZTG was then carried out in an in-house built chemical vapor deposition (CVD) reactor system from methanol as the carbon precursor. Reaction temperatures was varied and studied in order to obtain the ZTG with different quality and properties according to the different types of zeolite host materials. The ZTG produced was characterized using Thermal Gravimetry Analysis (TGA) to measure the different thermal range effect on the ZTG.

The ZTG produced then were undergone further chemical treatment with acid and base to remove the zeolite framework that act as the host for the production of graphene. The graphene obtained was dispersed in water as the medium to investigate the dispersion ability of the graphene produced. The dispersed solutions were than characterized using UV-spectrometer. The graphene obtained were characterized using Fourier-Transform Infrared Spectrophotometer (FT-IR) and Raman Spectroscopy.

1.5 Significance of Study

When the graphene sheets are successfully synthesized inside the zeolite pores as the host materials, it can open a new frontier of research regarding the synthesis approach of graphene sheets. Smaller size range of graphene sheets can be engineered as the growth of graphene take place in the pores of zeolites which the size of graphene sheets formed will depend on the pore size of the zeolite as the host materials.

In addition, graphene that has been successfully synthesized in this research can be used to be applied in the fabrication of transparent conductive film. The transparent conductive film prepared from graphene is ableto provide similar or better performances in conductivity as compared to indium tin oxide (ITO) which is widely used as the transparent electrode in touch panel devices. Besides that, graphene are the best candidates to replace ITO in the transparent electrode technology due to the low cost manufacturing of this remarkable material.

1.6 Outline of the Research

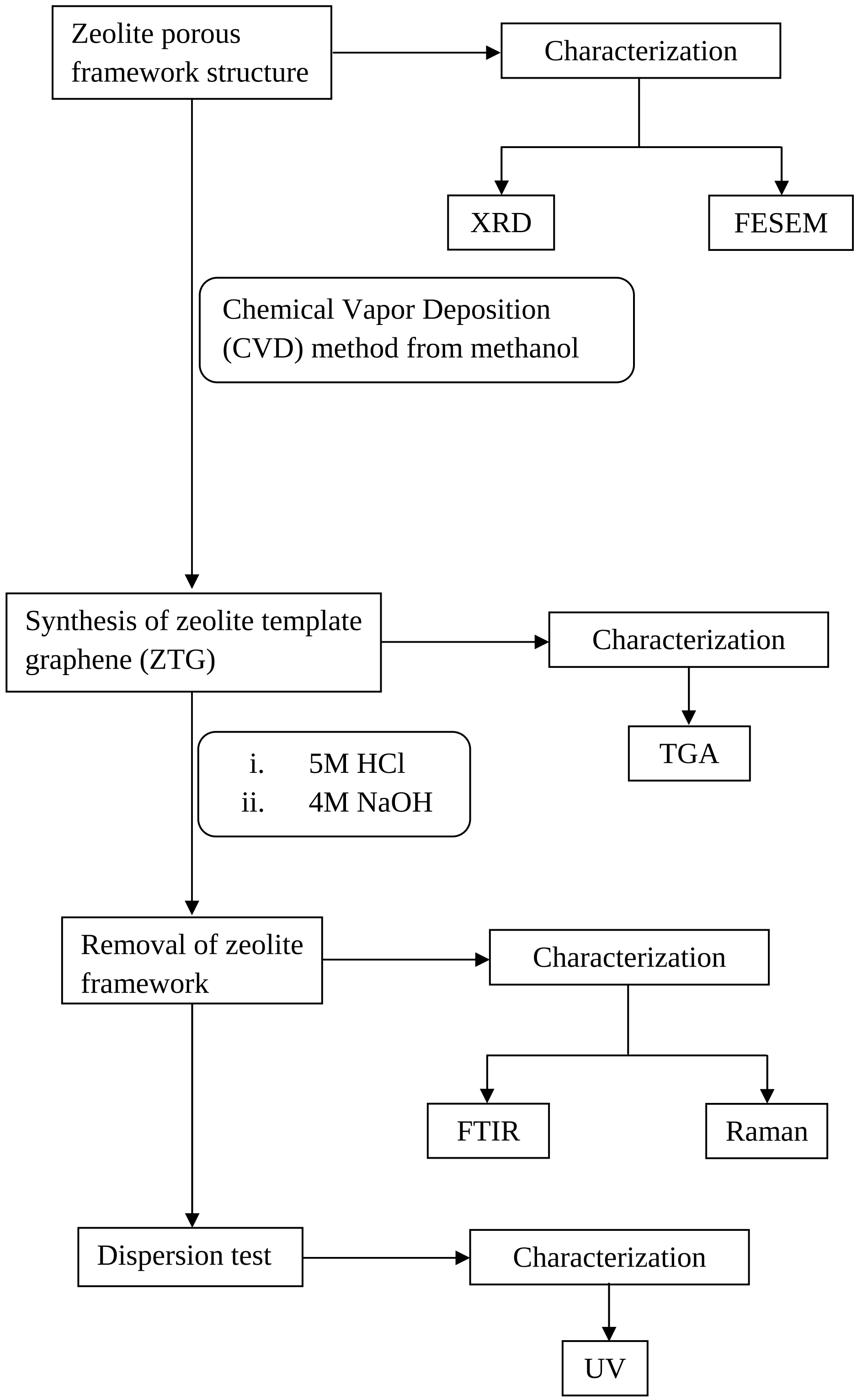

Figure 1.2: The flow chart of the research

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