

DEPOSITION OF REDUCED GRAPHENE OXIDE FILM USING DROP
CASTING FOR FLEXIBLE ELECTRONIC

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ABSTRACT

Graphene and its derivatives such as reduced graphene oxide (rGO) are regarded as promising materials for electrode in flexible electronics platform. Deposition of rGO film onto flexible substrate is the key technology to enable the application of rGO into the flexible electronics system. In this work, the feasibility of a simple drop-casting technique to deposit graphene oxide (GO) onto polyimide film substrate was investigated. The polyimide film is widely used in electronics and can withstand relatively high temperature. This work started with preparation of GO dispersion with different concentration. Prior to drop-casting process, polyimide surface was treated with 1 molar Potassium Hydroxide. The prepared dispersion was drop-casted onto polyimide film which was heated at 80°C. Then, the formed GO film on the polyimide was reduced by means of ascorbic acid and thermal reduction processes at 300°C. The relationship between rGO volume with rGO film properties, namely coverage, thickness and uniformity was investigated using Raman Microscope, Atomic Force Microscopy and 3D Microscope. Drop casting of 1.5 ml GO dispersion produced GO film that covered 50mm x 50mm area. By varying the GO dispersion concentration from 0.025 to 0.1 mg/ml, film with thickness from 16 to 30 nm could be deposited. The average film roughness is around 9.82 nm. After the reduction process, film sheet resistance was analyzed by means of four-point probe measurement. rGO film produced from 0.1 mg/ml dispersion showed sheet resistance as low as 1.36 k Ω /sq. The obtained result showed process feasibility to obtain rGO film with controllable thickness and acceptable conductivity. The developed technique can be further utilized in electronic device fabrication at least at lab scale.

ABSTRAK

Graphene dan terbitannya seperti *reduced graphene oxide (rGO)* dianggap sebagai bahan yang menjanjikan untuk elektrod dalam platform elektronik fleksibel. Pemendapan filem *rGO* ke substrum yang fleksibel adalah teknologi utama untuk membolehkan penggunaan *rGO* ke dalam sistem elektronik fleksibel. Dalam kajian ini, kemungkinan teknik *drop-casting* mudah untuk meletakkan *GO* ke substrum filem *polyimide* telah disiasat. Filem *polyimide* digunakan secara meluas dalam elektronik dan boleh menahan suhu yang tinggi. Kajian ini bermula dengan penyediaan *GO* dengan kepekatan yang berlainan. Sebelum proses *drop-casting*, permukaan *polyimide* dirawat oleh 1 Molar *Potassium Hydroxide*. *GO* yang disiapkan telah di *drop-cast* ke filem *polyimide* yang dipanaskan pada 80°C. Kemudian, *drop-casted GO* pada *polyimide* dipanaskankan dengan asid askorbik dan melalui proses pengurangan haba pada 300°C. Hubungan antara kuantiti *rGO* dengan sifat filem *rGO*, iaitu liputan, ketebalan, keseragaman dan kekonduksian elektrik telah disiasat menggunakan Raman Mikroskop, *Atomic Force Microscope* dan Mikroskop 3D. 1.5 ml *drop cast GO dispersion* dihasilkan filem *GO* yang meliputi kawasan 50mm x 50mm. Dengan mengubah kepekatan penyebaran *GO* dari 0.025 hingga 0.1 mg / ml, filem dengan ketebalan 16 hingga 30 nm boleh didepositkan. Purata bagi kekasaran filem adalah sekitar 9.82 nm. Selepas proses pengurangan, rintangan lembaran filem dianalisis dengan cara pengukuran kuar empat titik. Filem *rGO* yang dihasilkan daripada penyebaran 0.1 mg / ml menunjukkan rintangan lembaran serendah 1.36 k Ω / persegi. Dari hasil yang diperoleh, prosedur dan resipi proses untuk mendapatkan filem *rGO* dengan ketebalan yang dikawal dan kekonduksian yang boleh diterima telah dibangunkan. Teknik yang dibangunkan ini boleh digunakan lebih dalam fabrikasi peranti elektronik sekurang-kurangnya pada skala makmal.

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LIST OF ABBREVIATIONS

AFM	-	Atomic Force Microscope
Cm	-	centimeter
DI	-	Deionized water
GO	-	Graphene Oxide
KOH	-	Potassium hydroxide
m	-	meter
M	-	Molar
ml	-	milliliter
mg	-	milligram
Nm	-	nanometer
PET	-	Polyethylene terephthalate
PCB	-	Printed Circuit Board
PI	-	Polyimide
Ra	-	Roughness
rGO	-	reduced Graphene Oxide
s	-	second
S	-	Siemens
SEM	-	Scanning Electron Microscopy
Sq	-	square
SrGO	-	Silicon exfoliated reduced Graphene Oxide
TEM	-	Transmission Electron Microscopy
V	-	Voltage
XPS	-	X-ray Spectroscopy
XRD	-	X-ray Diffraction

LIST OF SYMBOLS

Ω	-	Ohm
$k\Omega$	-	Kilo Ohm
$^{\circ}$	-	degree

CHAPTER 1

INTRODUCTION

1.1 Project Background

In order to reach the best result in the semiconductor and electronics industries, there always been research development regarding the material that been used. Graphene is one of the materials that is widely investigated due to the unique properties. Graphene also conducts heat and electricity efficiency and is nearly transparent. Compared to copper, graphene is not easily oxidized and is chemically stable[1]. There are many ways to synthesize graphene such as mechanical exfoliation, epitaxial growth, liquid-phase exfoliation of graphite, chemical vapor deposition and chemical reduction of graphene oxide (GO) [2].

Reduced graphene oxide (rGO) is a graphene derivative with promising potential for applications in modern electronics[3]. The interest on chemically derived graphene has been increased in view of its potential applications due to their good mechanical, electrical and thermal properties[4]. The rGO possesses improved electrical conductivity after the reduction process of GO[5]. Thin film of rGO is usually deposited using wet process from GO dispersion. The GO is its easy disposability in water and other organic solvents, as well as in different matrixes, due to the presence of the oxygen functionalities.

. In this project, feasibility of using rGO for flexible electronics is investigated. Selection of suitable substrate is a crucial step to ensure the reliability and robustness of the fabricated device. Polyimide is one of materials used as the substrate in order to apply in flexible electronic purposes. The advantages of

polyimide materials are lightweight, flexible, resistant to heat and chemicals. Examples of polyimide films include Apical, Kapton, UPILEX, VTEC PI, Norton TH and Kaptrex. Due to its large range of temperature stability, and its electrical isolation ability, Kapton tape is usually used in electronic manufacturing as an insulation and protection layer on electrostatic sensitive and fragile components. As it can sustain the temperature needed for a reflow soldering operation, its protection is available throughout the whole production process.

Deposition techniques is an important to enable the formation of uniform rGO film with desired thickness. In order to produce uniform graphene films, there are techniques such as spin coating, spray coating, dip-coating, vacuum filtration transferring and drop-casting[6]. The drop-casting method was effectively utilized in the self-assembly of reduced graphene oxide (rGO) materials by evaporation of the solution to form flexible, transparent and conductive films [6]. Process condition to obtain rGO film with good uniformity and conductivity can be archived after the drop-casting method is implemented. In this project, the deposition of rGO film on polyimide film using drop-cast method is investigated.

1.2 Problem Statement

The effectiveness and performance of the rGO film (i.e. electrical conductivity) is determined by the deposition method used. The drop-cast technique was considered in this project due to its simplicity, where the deposition process does not require any sophisticated equipment and setup. This technique is suitable for lab-scale production for the purpose of fundamental and proof-of-concept studies.

Since the technique is not favorable for mass production, less attention was given and less report could be found on systematic study on the characteristics of the drop-casted rGO film on polyimide film. Nevertheless, these information is worth being investigated. Several significant rGO film properties are film uniformity, coverage and electrical conductivity. The volume and the number of drop casting of

the rGO water dispersion is varied too in a way to get the better uniformity and coverage.

1.3 Objectives

The objectives of this research are as follows:

- 1) To develop process condition for deposition of rGO thin film onto polyimide film by using drop cast method.
- 2) To characterize morphology in example uniformity, coverage and electrical properties of the formed rGO film.

1.4 Scope

In order to achieve the objective of this project, there are some guidelines need to be referred in this project. The theory involved in constructing the device is investigated before constructing the device. First of all, the characteristic of the reduced graphene oxide has been study. GO aqueous dispersion purchased from Graphenea Inc. which the concentration was 1mg/ml is used to obtain GO water dispersion with four other concentration. The reduction process is done using ascorbic acid and thermal with process condition developed[2].

The polyimide is use to be the substrate so that the GO water dispersion can be drop casted onto 50mm x 50mm area. The amount of GO water dispersion and number of drop casting is varied to produce different GO film thickness. The effect of the heating during the drop casting is also investigate. Kapton was chosen due to the great characteristic of this polyimide compared to Polyethylene Terephthalate (PET)in terms of temperature resistance[7].

The measurement of the conductivity of the rGO film is characterized from four point probe measurement. The morphology of the film also will be characterized by using optical microscope and atomic force microscope. Next after studying and reviewing all the information about the electrical characteristics of reduced graphene oxide and the polyimide, the process conditions were designed to obtain the reduced graphene oxide with good uniformity and good conductivity as well as the relationship of the drop casting condition and rGO film properties.

1.5 Report Outline

This report is organized in five chapters. Chapter one gives an overview and the introduction of the project including the problem statement, objectives and research scope.

Chapter Two discusses the literature review on the definition and characteristics of graphene oxide, followed by the method to produce reduced graphene oxide. The types of deposition techniques are discussed under the topic of reduced graphene oxide. Drop-casting was the technique that has been used on the polyimide substrate and will be discussed more deeply. Next, the advantages of using this technique compared to other techniques have been also discussed. Before the summaries of this chapter, the surface treatment of Kapton on the polyimide and the previous study on the drop-casting method have been discussed.

Chapter Three covers the design methodology of the project. In this chapter, the overview of the design and all the tools and modules used in this project are discussed. Chapter Four shows the results of the method that was applied to the rGO film. The results of the measurements from the microscope are also discussed in this chapter. In the last chapter, it will cover the conclusion and the future works for this study.

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