



UNIVERSITI PUTRA MALAYSIA

***SYNTHESIS, CHARACTERIZATION AND FILTRATION OF
ADSORPTIVE BIOBASED MEMBRANE FROM JATROPHA OIL
MODIFIED WITH GRAPHENE OXIDE***

NUR HANINAH BINTI HARUN

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ADSORPTIVE BIOBASED MEMBRANE FROM *JATROPHA* OIL
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By

NUR HANINAH BINTI HARUN

**Thesis Submitted to the School of Graduate Studies, Universiti Putra
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Philosophy**

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Doctor of Philosophy

SYNTHESIS, CHARACTERIZATION AND FILTRATION OF ADSORPTIVE BIOBASED MEMBRANE FROM *JATROPHA* OIL MODIFIED WITH GRAPHENE OXIDE

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NUR HANINAH BINTI HARUN

June 2022

Chair : Prof. Zurina Zainal Abidin, PhD
Faculty : Engineering

Industrial discharges containing heavy metals are one of the major concerns globally. Among various method in water treatment, membrane filtration is favored due to its minimal impact on the environment. To move towards green economy, vegetable oil derivatives have been used as a bio-based polymer due to their chemical properties, sustainability, low cost, and ease of access. Therefore, the purpose of this study is to utilize vegetable oil from *Jatropha curcas* that is chemically ideal for bio-based membrane synthesis and a drought-resistance plant, hence is easier to sustain. However, due to the common limitation in organic membrane (e.g., low in hydrophilicity, thermal resistance, separation efficiency), this work aims to develop a new bio-based adsorptive membrane filter from *Jatropha* oil derivatives modified with graphene oxide (GO) to improve its features and performance.

Jatropha oil-based polyol (JOL) was mixed with hexamethylene diisocyanate (HDI) to produce *Jatropha* polyurethane (JPU) in different conditions (HDI:JOL ratio, cross-linking and curing temperature). HDI: JOL ratio was optimum at 5:5 (v/v) with cross-linking and curing temperature at 90 °C and 150 °C, respectively. GO was added into JPU polymer matrix at different weight percent (0.35 wt%, 0.50 wt%, and 0.65 wt%) to form *Jatropha*/graphene oxide membrane (JPU/GO). JPU and JPU/GO were evaluated and characterized. Glass transition temperature (T_g) and onset temperature (T_o) were increased from 58°C to 69°C and from 170°C to 202°C, respectively due to the presence of GO that promotes crystallization, thus improve thermal stability. Additionally, contact angle decreased from 88.8° to 52.1°, which signifies higher hydrophilicity due to GO oxygenated functionalities that assist in absorbing water. Besides, JPU/GO 0.50wt% demonstrates higher water flux followed by JPU/GO 0.35wt%, JPU/GO 0.65wt%, and JPU at 523 L/m².h, 406 L/m².h, 260 L/m².h, and 233 L/m².h, respectively. In filtration, JPU/GO exhibited higher Cu(II) rejection followed by

JPU/GO 0.35 wt%, JPU/GO 0.65 wt%, and JPU with rejections at 71.60 %, 67.56 %, 63.58 %, and 33.51%, respectively due to the interaction between GO and Cu(II) ions.

Besides, JPU/GO 0.50 wt% optimization for Cu(II) ions removal was done using response surface methodology (RSM) by central composite design (CCD). The experiments were done in different factors (60 - 140 ppm, 1.5 - 2.5 bar, pH 3 - 5). Optimum removal of Cu(II) ions was predicted at 116 ppm, 1.5 bar, and pH 3.7 with 87% rejection. The result obtained within 95% of prediction intervals at 80% rejection. Meanwhile, mass transfer mechanism for JPU/GO 0.50wt% was depicted by combined film theory/solution-diffusion (CFSD) and combined film theory/Spiegler-Kedem (CFSK). From the findings, CFSK model exhibited better prediction than CFSD. Additionally, resistance-in-series model was proposed to study the cause of foulant. Fouling JPU/GO 0.50wt% has reduced rejection to 59% with permeate flux at 291 L/m².h. After backwashing using pure water, the fouling membrane had achieved 67% rejection with flux at 441 L/m².h. The fouling studies recorded 52% of flux recovery, 20% reversible fouling ratio, and 48% of irreversible fouling. Results showed JPU/GO 0.50 wt% exhibited adsorptive characteristics. In conclusion, JPU/GO 0.50 wt% offers a better membrane physicochemical property and achieves a good quality permeate with minimal pollutant content. Thus, JPU/GO 0.50 wt% was proven to have the potential as an alternative for wastewater treatment.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

**PENGHASILAN, PENCIRIAN DAN PENAPISAN MEMBRAN PENAPIS
BERPENJERAP BERASASKAN BIO DARIPADA MINYAK *JATROPHA*
DIUBAHSUAI MENGGUNAKAN GRAFENA OXIDA**

Oleh

NUR HANINAH BINTI HARUN

Jun 2022

Pengerusi : Prof. Zurina Zainal Abidin, PhD
Fakulti : Kejuruteraan

Bahan buangan perindustrian yang mengandungi logam berat adalah salah satu kebimbangan global. Antara kaedah yang digunakan dalam merawat air, membran penapisan menjadi pilihan utama kerana memberikan kesan yang minimum terhadap alam sekitar. Untuk menuju ke arah ekonomi hijau, derivatif minyak sayuran telah digunakan sebagai polimer berasaskan bio kerana sifat kimianya, kelestariannya, kos yang rendah, dan mudah untuk didapati. Oleh itu, tujuan kajian ini adalah untuk menggunakan minyak sayuran dari *Jatropha curcas* yang secara kimia sesuai untuk membuat membran berasaskan bio serta merupakan tumbuhan tahan kemarau, lantas lebih mudah untuk dilestarikan. Walau bagaimanapun, disebabkan oleh batasan ciri yang biasa ada pada membran organik (cth., rendah hidrofilik, rintangan haba, dan kecekapan pemisahan), fokus kajian ini adalah untuk menghasilkan membrane tapisan berpenjerap berasaskan bio baharu daripada derivatif minyak *Jatropha* yang diubah suai dengan grafena oksida (GO) untuk menambah baik ciri dan prestasinya.

Poliol berasaskan minyak *Jatropha* (JOL) telah dicampur dengan heksametilena diisosianat (HDI) untuk menghasilkan poliuretana *Jatropha* (JPU) dalam keadaan berbeza (nisbah HDI:JOL, suhu ikatan sambung silang, dan suhu pengerasan). Nisbah optimum heksametilena diisosianat (HDI) ke JOL adalah 5:5 nisbah isipadu (cc:cc) dan suhu ikatan sambung silang serta suhu pengerasan masing-masing pada 90°C dan 150°C. GO ditambah ke dalam matriks polimer JPU pada peratusan berat yang berbeza (0.35% bt, 0.50% bt, and 0.65% bt) untuk menghasilkan membran *Jatropha*/grafena oksida (JPU/GO). JPU dan JPU/GO telah dianalisa dan dicirikan. Suhu peralihan kaca (T_g) dan suhu permulaan (T_o) masing-masing meningkat dari 58°C kepada 69°C

dan dari 170°C kepada 202°C kerana adanya GO menggalakkan penghabluran, lantas meningkatkan kestabilan haba. Tambahan lagi, sudut kontak membran berkurangan daripada 88.8° kepada 52.1°, yang menunjukkan hidrofilik membran yang lebih tinggi kerana fungsian beroksigen GO yang membantu dalam menyerap air. Selain itu, JPU/GO 0.50wt% menunjukkan tapisan air tulin yang lebih tinggi diikuti JPU/GO 0.35% bt, JPU/GO 0.65% bt, dan JPU masing-masing pada 523 L/m².jam, 406 L/m².jam, 260 L/m².jam, dan 233 L/m².jam. Dalam penapisan, JPU/GO menunjukkan penurasan Cu(II) yang lebih tinggi diikuti JPU/GO 0.35% bt, JPU/GO 0.65% bt, dan JPU dengan nilai penurasan masing-masing pada 71.60%, 67.56% , 63.58%, dan 33.51% disebabkan interaksi antara fungsian beroksigen GO dan ion Cu(II).

Selain itu, pengoptimuman JPU/GO 0.50% bt untuk penyingkiran ion Cu(II) telah dilakukan menggunakan rangsangan permukaan (RSM) melalui rekabentuk komposit pusat (CCD). Eksperimen telah dilakukan dalam faktor yang berbeza (60 – 140 ppm, 1.5 - 2.5 bar, pH 3 - 5). Penyingkiran optimum ion Cu(II) telah dijangka pada 116 ppm, 1.5 bar, dan pH 3.7 dengan 87% penyingkiran. Keputusan diperoleh dalam 95% selang jangkaan untuk penyingkiran 80%. Sementara itu, mekanisma pemindahan jisim JPU/GO 0.50% bt telah ditunjukkan melalui teori gabungan filem/ larutan-penyebaran (CFSD) dan teori gabungan filem/ Spiegler-Kedem (CFSK). CFSK menunjukkan jangkaan yang lebih baik berbanding CFSD. Tambahan lagi, model siri-rintangan diusulkan untuk mengkaji punca terbentuknya kotoran. Prestasi penyingkiran JPU/GO 0.50% bt yang telah kotor merosot kepada 59% dengan tapisan 291 L/m².jam. Selepas pembilasan berbalik menggunakan air tulin, JPU/GO 0.50% bt telah mencapai 67% penyingkiran dengan tapisan pada 441 L/m².jam. Kajian kotoran pula kemudian merekodkan 52% pemulihan pengeluaran tapisan, 20% kotoran yang boleh dikeluarkan, dan 48% adalah kotoran yang tidak dapat dipulihkan. Hasil kajian menunjukkan JPU/GO 0.50% bt mempunyai ciri-ciri penjerapan. Kesimpulannya, JPU/GO 0.50% bt menawarkan sifat fizikokimia membran yang lebih baik dan telah mendapat kualiti pengeluaran tapisan yang baik dengan kandungan pencemar yang minimum. Oleh itu, membran JPU/GO 0.50% bt telah terbukti mempunyai potensi sebagai membran penapis alternatif berasaskan bio untuk rawatan air sisa.

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Zurina binti Zainal Abidin, PhD

Professor
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Abdul Halim bin Abdullah, PhD

Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

Mohamad Rezi bin Abdul Hamid, PhD

Senior Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Mohd Yusof bin Harun, PhD

Senior Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Rizafizah bin Othaman, PhD

Associate Professor
Faculty of Science and Technology
Universiti Kebangsaan Malaysia
(Member)

ZALILAH MOHD SHARIFF, PhD

Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 10 November 2022

Declaration by Members of the Supervisory Committee

This is to confirm that:

- the research and the writing of this thesis were done under our supervision;
- supervisory responsibilities as stated in the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2015-2016) are adhered to.

Signature: _____
Name of Chairman
of Supervisory
Committee: Prof. Zurina Zainal Abidin

Signature: _____
Name of Member of
Supervisory
Committee: Assoc. Prof. Abdul Halim Abdullah

Signature: _____
Name of Member of
Supervisory
Committee: Assoc. Prof. Rizafizah Othaman

Signature: _____
Name of Member of
Supervisory
Committee: Dr. Mohamad Rezi Abdul Hamid

Signature: _____
Name of Member of
Supervisory
Committee: Dr. Mohd Yusof Harun

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LIST OF ABBREVIATIONS

PU	Polyurethane
JPU	<i>Jatropha</i> oil-based polyurethane membrane
JPU/GO	<i>Jatropha</i> oil-based polyurethane membrane/graphene oxide
GO	Graphene oxide
EJO	Epoxide crude <i>Jatropha</i> oil
JOL	<i>Jatropha</i> oil-based polyol
HDI	hexamethylene diisocyanate
RSM	Response Surface Methodology
CFSD	Combined film-theory/solution-diffusion model
CFSK	Combined film-theory/spiegler-kedem model
FESEM	Field emission electron microscopy
XRD	X-ray diffraction
FTIR	Fourier-transform infrared spectrometer
DSC	Differential scanning calorimetry
TGA	Thermogravimetric analysis
CP	Concentration polarization
RIS	Resistance-in-series model
TMP	Transmembrane pressure
CFV	Cross-flow velocity
IRFR	Irreversible fouling ratio
RFR	Reversible fouling ratio
eNRTL	Electrolyte non-random two liquids
NG	Nucleation and growth
SD	spinodal decomposition

CHAPTER 1

INTRODUCTION

1.1 Background

To date, the demand for fresh water is increasing due to the rapid growth of the human population along with the evolution of the industrial sectors (Ramakrishnaiah et al., 2009). Moreover, due to the anthropogenic activities, the surface water quality may also deteriorate that can be very harmful for other uses (Kepner et al., 2004; Simeonov et al., 2003). Without the proper ethics in water treatment, polluted water can be a drawback for any future development. In Malaysia, wastewater from the private sector in urban areas is obliged to undergo tertiary wastewater treatment before being released into the surface waters. The Environmental Quality Act of 1974 and associated rules, such as the Environmental Quality (Industrial Effluent) Regulations 2009, has govern the effluent from the treatment facilities to conserve the quality standard of the treated wastewater.

Various methods regarding the remediation of wastewater treatment have been applied that include the flocculation, membrane filtration, photocatalysis, and coagulation (Aljohny et al., 2020; Yao et al., 2017). However, membrane filtration are favored due to its simplicity, easy to handle, and give lower risks to the environment (Xu et al., 2021). In general, filtration is done by using a thin layer semi-permeable membrane as a barrier to allow only selected species to pass through the membrane while preventing the unwanted substance as the driving force is applied. The effectiveness of the membrane filtration is on the basis of its porosity and selectivity.

Besides, the researcher has been using biopolymers for membrane synthesis to address the importance of sustainability as an alternative to encourage the industry to move towards green technology. In general, biopolymers can be produced from monomers or chemically modified natural polymers such as polylactic acid, (PLA), cellulose, chitosan, starch (Ibrahim et al., 2019; Phan et al., 2021; Russo et al., 2021), and vegetable oil (Ibrahim et al., 2015; Karimi et al., 2017). With different chemical properties, organic polymer precursors are utilized based on their suitability and availability for various applications. Among these, vegetable oil is one of the promising sources that have shown the potential as an alternative for biomaterial synthesis due to its sustainability and readily available in nature.

Remarkably, the usage of vegetable oil from *Jatropha curcas* has shown a growing interest in wastewater treatment due to its outstanding efficiency in treating the effluent. For example, studies have found biocoagulant from *J.*

curcas presscakes and seed extracts to be effective for turbidity removal in kaolin wastewater (Abidin et al., 2011) and in palm oil mill effluent (POME) (Abidin et al., 2019) as well as the removal of Cd(II) from water using *J. curcas* bark, seed peel, and endosperm seed (Nacke et al., 2017). Additionally, *Jatropha curcas* seed (chaff) adsorbent has been successfully removed dye (Congo red) through the chemisorption process (Pudza & Abidin, 2020). In another recent study, *J. curcas* seeds extract has reduced the turbidity in pharmaceutical wastewater (Sibartie & Ismail, 2018). However, to date, no research has been reported on *Jatropha curcas* oil for the synthesis of membrane filter and this work aims to achieve this potential goal.

Despite the substantial achievement of polymeric membrane in wastewater treatment, some inadequacies such as low water flux and inefficient membrane selectivity remain. This has brought the attention towards adsorptive membrane that can remove the emerging contaminants that threaten human's health and the aquatic organisms. It is a method that involves both adsorption and filtration process (Chong et al., 2019). Adsorption process is a separation process in which the substances/solutes are bounded or adsorbed by chemical and physical interactions to membrane surface (Qalyoubi et al., 2021). Adsorptive membrane is commonly known as mixed matrix membrane (modified membrane) with many benefits (e.g., high separation rate and flux, low operating pressure, and small footprint) (Qalyoubi et al., 2021). To produce adsorptive membranes, nano size adsorbents (known as nanoparticles) have been commonly used as membrane additives due to their surface characteristics and many successful reviews have been reported in wastewater treatment (Wang et al. 2012). The type of nanoparticles used usually consist hydroxyl, amino, carboxyl, and sulfonic groups for adsorption functions (Z. Q. Huang & Cheng, 2020). For example, other studies have reported polyethersulfone (PSF) membrane shows a better membrane separation potential for dyes removal after being incorporated with O-carboxymethyl chitosan/Fe₃O₄ nanoparticles (Zinadini et al., 2014) while polyvinylidene fluoride/ZnO (PVDF/ZnO) composite membrane exhibits higher Cu(II) removal than the pristine PVDF (Zhang et al., 2014), which have revealed the removal efficiency of the polymeric membrane can be experimentally controlled with the modification of the membrane building components (Nam et al., 2014). Therefore, from the findings, it is possible to enhance the membrane characteristics with the additional methods which can maximize the foulant rejections.

1.2 Problem statement

Recent advances in urbanization have significantly upset water quality due to the industrial activities such as coal mines or metal processing that release harmful effluent in a form of water-soluble toxic metal waste (e.g., Cu, Zn, Pb, Al etc.). (Rowell & Boving, 2010). This toxic effluent, unfortunately, can easily leach to the earth's surface and groundwater, potentially harming not only the environment but also living organisms. Various methods for metal ion removal have been applied including flocculation, membrane filtration, photocatalysis, and coagulation (Ahmad et al., 2022; Aljohny et al., 2020; Yao et al., 2017).

Among these technologies, membrane filtration is favored due to its small footprint area and chemical usage with minimal impact on the environment (Ahmad et al., 2022). Moreover, the over-use of resources has challenged the researcher to overcome this issue by finding the cost-effective and innovative method corresponding to the environmental and public pressures. Therefore, the utilization of natural bioresources in membrane synthesis has been used as an alternative to encourage the industry to move towards sustainability.

This moves the interest to use the versatile *Jatropha* oil in polymer technology due to its chemical properties and sustainability. *Jatropha* oil contains two types of unsaturated fatty acids (double bond), namely oleic acid and linoleic acid with the value of 41.68% and 35.55%, respectively (Nakpong & Wootthikanikkhan, 2010). This high amount of unsaturated fatty acid is capable to promote further reactions to occur. Apart from that, *Jatropha curcas* has high tolerance with prolonged period of water shortage and manage to survive in abandoned agricultural land (Ong et al., 2011). From its chemical properties, *Jatropha curcas* oil has the potential to be used as a precursor for the synthesis of renewable and environmentally friendly membrane and subsequently used for filtration in environmental applications which has not been explored.

Jatropha oil-based membrane is a promising sustainable membrane filter for water purification such as filtering metal ions from wastewater. However, in some cases of the organic polymer membrane, poor material properties are likely to happen that can cause inefficient water filtration due to its physicochemical properties. Limitations of organic polymer membrane are often due to its large pores and less active functional groups to adsorb and filter the penetrated ions. In membrane technology, synthetic polymer membrane is commonly incorporated with nanoparticles to improve the membrane selectivity as well as reducing the tendency from fouling to occur (Yin & Deng, 2015). Hence, it is desirable to have more active functional groups by embedding nanoparticles into the polymer matrix.

In membrane composite productions, graphene oxide (GO) is commonly used as membrane additives due to its unique structure and surface characteristics. As a typical single atomic two-dimensional nanomaterial, GO exhibits a large specific surface area and high charge mobility, which gives it famous applications in material science (Lu et al., 2012; Stankovich et al., 2006) and wastewater purification (Li et al., 2014; Li & Zhi, 2018; Perreault et al., 2015). Notably, the oxygen-containing functional groups on the surface of GO, such as carboxyl and hydroxyl groups, are capable to adsorb metal cations which become a plus point in wastewater treatment (Yu et al., 2019).

In recent years, due to the rising concern about environmental issues, the control and proper operation of wastewater treatment to meet the requirement have become crucial. Modeling the treatment process can provide proper operation and better control of wastewater treatment system (Wantala et al., 2012). The ability of response surface methodology (RSM) in modeling complex systems

that include nonlinear characteristics has made them the most popular tool for modeling filtration processes (Mason et al., 2003). Hence, response surface methodology (RSM) was used to design the filtration experiment and to evaluate its performance (Issa et al., 2020). From RSM, an optimum condition for JPU/GO 0.50 wt% membrane to achieve the highest Cu(II) ions rejections can be determined.

On the other hand, there are different theories and models have been developed to model mass transfer describing flux of water and solutes through the membrane. Researchers in recent years have paid a great deal of attention in modeling mass transfer membrane filtration processes that is beneficial to pre-design studies, the operation, and other aspects of the advancements in water treatment. In water filtration process, CFSD and CFSK models are widely used and were based on a few principles of diffusion, convection, and film theory. For mass transfer model, mass transfer coefficients is the important parameter, in which, membrane surface characteristics, solute, and solvent properties are the factors affecting the parameter (Cussler, 2003; Fang, 2013). Therefore, it is crucial to determine membrane mass transfer coefficient that can be determined by solving the model equations.

Apart from that, membrane filtration application in industrial wastewater treatment is often limited due to fouling. Membrane fouling is a condition when the flux started to decline, that is, the loss of permeate flux with time due to the solute deposition towards membrane surface and pores (Tay & Song, 2005). To date, the related studies have focused on understanding the underlying factors that hinder the membrane performance. As reported in other findings, manipulating the operating conditions can lessen the severity of fouling (Tu et al., 2005). Therefore, resistance-in-series model was employed at different conditions to observe the changes in membrane fouling. However, fouling membrane are often linked with irreversible and reversible fouling that is caused by the foulant adsorption towards membrane surface and pores. The phenomena either caused by physisorption (reversible fouling) or chemisorption (irreversible fouling) which determines the membrane lifetime (Ezugbe & Rathilal, 2020; Speth et al., 1998; Zainith et al., 2021). Hence, it is reasonable to undergo membrane cleaning process that determines the reversible and irreversible fouling ratio, which will indicate the occurrence of membrane adsorptive characteristics from the fouling ratio.

1.3 Objectives

There are 5 main objectives:

1. To optimize the synthesis *Jatropha* oil-based polyurethane membrane (JPU) and elucidate its properties.
2. To optimize the synthesis JPU membrane modified with Graphene Oxide (JPU/GO) and evaluate its properties.

3. To investigate the performance of *Jatropha* oil-based membrane using Response Surface Methodology (RSM) for Cu(II) ions removal from aqueous solution.
4. To correlate CFSD and CFSK mass transfer model for removal of Cu(II) ions from aqueous solution by using JPU/GO 0.50 wt % membrane.
5. To study the fouling behavior of JPU/GO 0.50wt% membrane using resistance-in-series model and to evaluate the effectiveness of backwashing by pure water for mitigation process.

1.4 Scope of research

To obtain the above objectives, the studies were divided into the following parts:

1. JPU membrane synthesis.
 - a. *Jatropha* oil was processed into a two-steps method to produce *Jatropha* oil-based polyol (JOL): epoxidation and ring-opening. JOL was blended with hexamethylene diisocyanate (-NCO) to form *Jatropha* oil-based polyurethane (JPU).
 - b. The synthesis of JPU was conducted at three different parameters; JOL: HDI ratio (v/v), the temperature of the reaction, and the curing temperature. The synthesized JPU membranes were observed to determine which sample experiments were solidified.
2. JPU/GO membrane synthesis.
 - a. GO was added into JPU polymer matrix at three different ranges. The difference in JPU/GO membranes were assessed through characterizations.
3. Characterization of the optimized JPU and JPU/GO membrane.
 - a. JPU and JPU/GO membranes were characterized in terms of their morphology, thermal stability, hydrophilicity, and porosity. The membrane properties were studied using field emission electron microscopy (FESEM), thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), Fourier-transform infrared spectrometer (FTIR), contact angle, and porosity.
4. To optimize the performance of *Jatropha* oil-based membrane using RSM for Cu(II) ions removal.
 - a. A sequential factor screening methodology, that is one-factor-at-a-time (OFAT) design was done to determine the range of parameters for every variable before continuing with the design of experiments (DOE).
 - b. Beforehand, an experiment was conducted on JPU and JPU/GO membranes at constant concentration, pressure and pH for Cu(II) rejection in order to select membrane with the best performance. From the experiment, membrane with the highest obtained Cu(II) rejection was chosen for further experiments using OFAT method to determine the suitable range of parameters (concentration,

pressure, pH) to be studied. The concentration of Cu(II) ion was determined using atomic absorption spectroscopy (AAS).

- c. After obtaining the range for each parameters, DOE was carried out by using RSM for the optimization of Cu(II) rejection. A central composite design (CCD) was used in the experimental design.
5. To investigate the mass transfer mechanisms of JPU/GO 0.50wt % membrane filtration for Cu(II) ions removal from aqueous solution.
 - a. Two types of mass transfer models were considered: (i) the non-porous or homogenous model (solution-diffusion (SD) model) and (ii) the irreversible thermodynamics model (Spiegler-Kedem (SK) model). SD models assumed the membrane is homogenous (non-porous) and the membrane transport is only by diffusion (Khalaf, 2008a; Lonsdale et al., 1965) whereas SK model assumed the membrane transport involves both diffusion and convection while assuming the membrane as a black box which requires no specific knowledge of the membrane structure and its porosity.
 - b. Since the mass transfer model is chosen based on the morphology of the membrane top skin layer, solution-diffusion (SD) model is proposed as the membrane top surface exhibits no observed pores. Additionally, SK model is chosen to have a comparative study with SD model due to the assumption of the transport behavior which involves both diffusion and convection.
 - c. Film theory is included for the concentration polarization, that is, a common occurrence for the filtration process. Thus, the membrane transport behavior was assessed by these two models:
 - (i) Combined film theory/solution-diffusion model (CFSD)
 - (ii) Combined film theory/Spiegler-Kedem model (CFSK)
 - d. For mathematical modeling, the estimation parameters were obtained from nonlinear regression in Aspen plus V11 and Microsoft excel.
 6. Membrane fouling behavior on JPU/GO 0.50wt % membrane.
 - a. The experiment was done by physical cleaning (backwash method) using distilled water to mitigate the foulant membrane.
 - b. Cu(II) ions removal and flux recovery were calculated.

1.5 Hypothesis

Jatropha oil derivative is proposed for membrane synthesis due to its chemical properties that are suitable for further reactions in producing bio-based polymers. However, with common poor bio-based membrane characteristics, *Jatropha* oil-based membrane was modified by adding graphene oxide in the polymer matrix to improve its physicochemical properties and membrane performance for water filtration applications such as in Cu(II) ions removal from aqueous solution.

1.6 Structure of the thesis

Chapter 1 focuses on the background studies, problem statements, objectives, scope of the research, and thesis structure.

Chapter 2 includes a comprehensive literature review on membrane filtration for wastewater treatment, a bio-based precursor for membrane fabrication, and additives for material modification. The utilization of vegetable oil such as *Jatropha* oil as a precursor for bio-based membrane incorporated with graphene oxide as additives, its associated filtration mechanism, optimization on filtration condition using RSM, mass transfer model, and fouling studies are discussed further.

Chapter 3 delivers information on the materials and methods used in this study which contains the preparation of a bio-based membrane using *Jatropha* oil and its modification with graphene oxide. This chapter also includes the characterization methods (FTIR, TGA, DSC, FESEM, contact angle, zeta potential, AAS, XRD), optimization by RSM, mass transfer model, fouling studies, and computational methods for data regression and filtration trials were also discussed.

Chapter 4 discusses the results obtained from the study which comprised of bio-based membrane characterizations, filtration optimization using CCD in RSM, data regression using Aspen plus and Microsoft excel for mass transfer model, and fouling study. The potential of *Jatropha* oil-based membrane incorporated with graphene oxide for heavy metal removal as well as the mechanisms during filtration was discussed in detail.

Chapter 5 presents the conclusion of the studies and the recommendations for future work.

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