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FABRICATION OF GRAPHENE OXIDE-BASED MEMBRANE AND DURIAN SHELL-BASED ACTIVATED CARBON FOR WATER TREATMENT APPLICATION



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UNIVERSITI PENDIDIKAN SULTAN IDRIS
2020



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THESIS SUBMITTED IN FULFILLMENT OF THE REQUIREMENT FOR THE
DEGREE OF MASTER OF SCIENCE (PHYSICS)
(RESEARCH MODE)

FACULTY OF SCIENCE AND MATHEMATICS
UNIVERSITI PENDIDIKAN SULTAN IDRIS
2020



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ACKNOWLEDGEMENTS

All praises and glories to Almighty Allah (SWT) who had bestowed me courage and patience upholding this work. I would like to express my appreciation and immense gratitude to my supervisor Prof. Dr. Suriani Abu Bakar for the continuous support of my study and research, for her patience, motivation, enthusiasm and immense knowledge. Her guidance helped me a lot to develop my skill as well as my interest toward research. Besides my supervisor, I would like to thank my co-supervisor, Assoc. Prof. Dr. Azlan Kamari and Assoc. Prof. Dr. Azmi Mohamed from Chemistry Department, UPSI for their assistance, advice, support, and help me a lot by allowed me to use their laboratory equipment.

My sincere thanks to Dr. Rosiah Rohani and her student, Izzati Izni Yusoff, from Universiti Kebangsaan Malaysia (UKM) for letting me use their laboratory equipment and help me on my project. I would like to express my gratitude to all the staffs of Faculty Science and Mathematics, UPSI for their assistance. I would also express my appreciation to TWAS-COMSTECH Joint Research Grand and Fundamental Research Grand Scheme for financial support on my research.

My appreciation for my lab mates and friends: Dr. Muqoyyanah, Dr. Khayri Zaid Z Al-Zalit, Dr Ali Abdul Ameer Mohammed Al-Saadi, Nur Jannah Idris, and Nur Amira Jamaluddin for the help, comments, and stimulating discussions during my study.

Last but not least, I would like to thank to my family, especially my parent who always supporting, give me courage and always believe on me. Without their help, support, and love, I am nobody. May Allah S.W.T repay all your kindness and bless us always.



ABSTRACT

This study aimed to fabricate polyvinylidene fluoride (PVDF)/graphene oxide (GO)/titanium dioxide (TiO₂)-based nanofiltration (NF) membrane and durian shell based-activated carbon (DAC) for water treatment application. The electrochemical exfoliation assisted by customized double-tail sodium bis(3,5,5-trimethyl-1-hexyl) sulphosuccinate (AOT4) and triple-tail sodium 1,4-bis(neopentyloxy)-3-(neopentyloxycarbonyl)-1,4-dioxobutane-2-sulphonate (TC14) surfactants were used to synthesize GO with N, N-dimethylacetamide (DMAc) as solvent. The fabrication of PVDF/GO/TiO₂-based NF membrane was done via non-solvent induced phase separation method by utilizing DMAc_GO based and TiO₂ as an additives and also PVDF as a main polymer material. On the other hand, durian shell was used to produce DAC by utilizing sulphuric acid as an activator agent via chemical activation method. The NF membrane samples via dead-end cell measurement as membrane separation were used for methylene blue (MB) dye rejection. Meanwhile, the DAC samples via adsorption process as adsorbent were then used for MB dye removal. The findings showed the PVDF/TC14-GO/TiO₂ NF membrane presented the highest dye flux (98.539 L/m²h) and dye rejection efficiency (92.61%) as compared to PVDF/AOT4-GO/TiO₂ membrane. This was due to the higher surfactants' tail number improved the fabricated membrane performance. The DAC sample showed the maximum adsorption percentage (92.05%) by utilizing 0.6 g DAC dosage, 10 ppm initial MB concentration and 15 minutes contact time. In conclusion, the fabricated PVDF/TC14-GO/TiO₂ and durian shell provided a potential to be applied as NF membrane material and activated carbon, respectively. Implication of this study is a novel, simpler, low-cost and greener for the PVDF/GO/TiO₂ NF membrane fabrication and activated carbon from durian shell production.





FABRIKASI MEMBRAN BERASASKAN GRAFIN OKSIDA DAN KARBON DIAKTIFKAN BERASASKAN KULIT DURIAN UNTUK APLIKASI RAWATAN AIR

ABSTRAK

Kajian ini bertujuan memfabrikasi membran penapisan nano (PN) berasaskan polivinilidena fluorida (PVDF)/grafin oksida (GO)/titanium dioksida (TiO_2) dan karbon diaktifkan berasaskan kulit durian (KBD) untuk aplikasi rawatan air. Pengelupasan elektrokimia yang dibantu oleh surfaktan buatan rangkaian bercabang dua sodium bis(3,5,5-trimetil-1-heksil) sulfosuksinat (AOT4) dan bercabang tiga sodium 1,4-bis(neopentiloksi)-3-(neopentiloksikarboni)-1,4-dioksobutana-2-sulfonat (TC14) digunakan untuk mensintesis GO dengan N, N-dimetilasetamida (DMAs) sebagai pelarut. Fabrikasi membran PN berasaskan PVDF/GO/ TiO_2 dilakukan melalui kaedah pemisahan fasa induksi tanpa pelarut dengan menggunakan DMAs_GO dan TiO_2 sebagai bahan tambahan dan PVDF sebagai bahan polimer utama. Selain itu, kulit durian digunakan untuk menghasilkan KBD dengan menggunakan asid sulfurik sebagai agen pengaktif melalui kaedah pengaktifan kimia. Sampel membran PN melalui pengukuran hujung mati sebagai pemisahan membran digunakan untuk penyingkiran warna metilena biru (MB). Manakala, sampel KBD melalui proses penjerapan sebagai bahan penjerap digunakan untuk penyingkiran warna MB. Dapatan kajian mendapati fluks warna ($98.539 \text{ L/m}^2\text{h}$) dan kecekapan penyingkiran warna (92.61%) bagi membran PN PVDF/TC14-GO/ TiO_2 adalah paling tinggi berbanding membran PVDF/AOT4-GO/ TiO_2 . Ini adalah disebabkan oleh bilangan rangkaian yang lebih tinggi meningkatkan prestasi membran yang telah dihasilkan. Sampel KBD menunjukkan bahawa peratusan maksimum penjerapan (92.05%) dengan menggunakan 0.6 g dos optimum KBD, 10 ppm kepekatan warna awal MB dan 15 minit masa hubungan. Kesimpulannya, PVDF/TC14-GO/ TiO_2 yang telah difabrikasi dan kulit durian, masing-masing berpotensi untuk digunakan sebagai bahan membran PN dan karbon diaktifkan. Implikasi kajian ini adalah sebuah pendekatan baharu, lebih mudah, kos rendah dan hijau untuk fabrikasi PVDF/GO/ TiO_2 PN membran dan penghasilan karbon diaktifkan daripada kulit durian.



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LIST OF ABBREVIATIONS

A	Area
AC	Activated Carbon
Al ₂ O ₃	Aluminium Oxide
AOT4	Sodium Bis(3,5,5-Trimethyl-1-Hexyl) Sulphosuccinate
BJH	Barret-Joyner-Halenda
BET	Brunauer-Emmet-Teller
C	Carbon
C _o	Initial Dye Concentration
C _f	Final Dye Concentration
C _p	Permeate Dye Concentration
CNT	Carbon Nanotubes
D	Defect and Disorder Raman Peak
DAC	Durian Shell-based Activated Carbon
DI	Deionized
DMAc	N, N-Dimethylacetamide
DMF	Dimethylformamide
DS	Durian Shell
DSSCs	Dye-Sensitized Solar Cells
EDX	Energy Dispersive X-Ray
F	Fluorine
FESEM	Field Emission Scanning Electron Microscopy





FRR	Flux Recovery Ratio
G	Crystalline Graphitic Raman Peak
GO	Graphene Oxide
HNO ₃	Nitric Acid
H ₃ PO ₄	Phosphoric Acid
H ₂ SO ₄	Sulphuric Acid
I_D/I_G	Ratio of Raman's D- and G-Peak Intensity
J	Permeated Flux
KClO ₃	Potassium Chlorate
KOH	Potassium Hydroxide
m	Mass
MB	Methylene Blue
MF	Microfiltration
NaOH	Sodium Hydroxide
NF	Nanofiltration
NMP	N-Methyl-Pyrrolidinone
NIPS	Non-Solvent Induced Phase Separation Process
O	Oxygen
PA	Polyamide
PAN	Polyacrylonitrile
PES	Polyethersulfone
PMMA	Poly(methyl methacrylate)
PVDF	Polyvinylidene Fluoride
ppm	Parts Per Million
q_e	Adsorption Capacity



R	Dye Rejection Efficiency
RO	Reverse Osmosis
SDS	Sodium Dodecyl Sulphate
t	Time
TC14	Sodium 1,4-Bis(Neopentyloxy)-3-(Neopentyloxycarbonyl)-1,4-Dioxobutane-2-Sulphonate
TEP	Triethyl Phosphate
Ti	Titanium
TiO ₂	Titanium Dioxide
TIPS	Thermally Induced Phase Separation
UF	Ultrafiltration
UV-Vis	Ultraviolet-Visible
V	Voltage
V	Volume
VIPS	Vapour-Induced Phase Separation
W	Weight
H ₂ O	Water
wt%	Weight Percentage
ZnCl ₂	Zinc Chloride
ZnO	Zinc Oxide
ε	Membrane Porosity
ρ_w	Density of Water
ρ_p	PVDF Density



LIST OF APPENDICES

A Academic Journal

B Presentation





CHAPTER 1

INTRODUCTION



1.1 Introduction

This chapter discusses the research background of graphene oxide (GO)-based membrane and durian shell-based activated carbon (DAC) for water treatment application. The research problem, objectives, scope and limitation of this study are also described in this chapter. This chapter is ended with a description of the thesis organization.



1.2 Research Background

The rapid growth of various industries such as paints, textile, printing inks, paper and plastics causes both of high polluted water and high demand of clean water, simultaneously. These industries, especially textile and dyeing industries utilized a large amount of synthetic dyes to colour their product which resulted large amount of polluted wastewater and released to the environment. Dyes presence can affect water quality since they are toxic and non-biodegradable (H. Liu et al., 2019; Tran, Kumar, & Lue, 2019). Therefore, the removal of this kind of pollutant is crucially needed.

Numerous approaches have been attempted to find an effective method to treat the polluted water in order to protect human health and the environment. Among various dye removal methods, membrane separation is one of the versatile and effective technologies to remove polluted water, such as dye contamination (T. Liu, Yang, Graham, Yu, & Sun, 2017). This was due to its several advantages, such as no phase changes, simple in operation and relatively low energy consumption (Kang & Cao, 2014). Over the past few decades, it becomes popular and plays an important role in major industries for water purification, metal recovery and protein separation (Escobar & Van Der Bruggen, 2015).

Membrane separation technology has become a promising in many type of filtration such as reverse osmosis (RO), ultrafiltration (UF), nanofiltration (NF) and microfiltration (MF). All of these types of filtration can be classified based on its membrane properties and separation principle. Recently, the NF membrane has widely applied for water treatment due to its small pore size (1-5 nm), low operating

pressure (4-20 MPa), high water permeability and can retain ions and low material's molecular weight (S. J. Park et al., 2012; Shon, Phuntsho, Chaudhary, Vigneswaran, & Cho, 2013). The polymeric membrane can be fabricated by a different technique such as phase inversion, sintering, track-etching and stretching (Liao, Loh, Tian, Wang, & Fane, 2018). Among them, phase inversion method is commonly used due to its several advantages, such as applicable for various polymers, simple in preparation and easy optimization of membrane thickness and pore size (Ladewig & Al-Shaeli, 2017).

Synthetic polymer membrane namely polyvinylidene fluoride (PVDF), a semicrystalline polymer with repeating units of $-\text{CH}_2\text{-CF}_2-$ has been extensively used in water treatment processes due to its outstanding physical and chemical properties (Kang & Cao, 2014). However, the hydrophobic nature of the PVDF membrane causes some critical problems which limit its application due to the membrane fouling during the separation process (Li et al., 2015; Wu, Zhang, Wang, Du, & Yang, 2018). The membrane fouling causes the declining in water flux permeability, shortening membrane lifetime and altering membrane selectivity thus decreased the membrane performance (R. Zhang et al., 2016). Therefore, several approaches have been developed to increase the hydrophilic properties of PVDF membrane such as physical and chemical modification with hydrophilic organic and inorganic materials (Kang & Cao, 2014; Xia & Ni, 2014).

Among these approaches, a physical modification of blending with organic or inorganic materials has been widely used due to its advantage of facile preparation method through phase inversion (non-solvent induce phase separation (NIPS)).

Recently, organic materials-blended PVDF (Poly(methyl methacrylate) (PMMA), Polyvinyl acetate, Poly(ether sulfones)) (PES) (Freire, Bianchi, Martins, Monteiro, & Forte, 2012; Vo & Giannelis, 2007), metal oxide particles (aluminium oxide (Al_2O_3), titanium dioxide (TiO_2) and zinc oxide (ZnO)) (J. Li et al., 2015; Q. Liu, Huang, Zhang, & Zhao, 2018), carbon nanotubes (CNT) (Sivakumaran, Kundu, Kumaran, Mishra, & Pandian, 2016; Zheng et al., 2009) and graphene oxide (GO) (Wu et al., 2018) have been utilized to enhance the PVDF membrane properties by increasing antifouling effect, permeability and membrane selectivity.

In particular, a potential candidate to effectively reinforce PVDF polymeric materials is TiO_2 due to its excellent properties such as commercial availability, chemical stability, hydrophilicity, and low toxicity (Ayyaru & Ahn, 2018; Behboudi, Jafarzadeh, & Yegani, 2016). TiO_2 is considered as the best metal oxide candidate to overcome the fouling issues by increasing the membrane hydrophilicity (Q. Liu et al., 2018; Wu et al., 2018). In addition, it also possesses small size particle and a large surface area (H. J. Park et al., 2018). Several reports have demonstrated that the utilization of TiO_2 could improve the membrane performance in dye separation.

Wang, Wang, Wang, Huang, and Wang (2013) showed that the introduction of TiO_2 into the PVDF membrane drastically increased membrane permeability ($190 \text{ L/m}^2\text{h.MPa}$) as compared to the pure PVDF membrane ($70 \text{ L/m}^2\text{h.MPa}$). Such improvement in the membrane permeability was believed due to the uniform deposition of the hydrophilic TiO_2 along the membrane pores. The result obtained by J. Zhang, Wang, Wang, Pan, and Wu (2017) showed an improvement in antifouling properties when TiO_2 was used as additive. Higher O element was observed which

indicated hydrophilicity enhancement with the TiO_2 presence. Méricq, Mendret, Brosillon, and Faur (2015) showed the improvement of membrane structure, hydrophilicity properties and permeability when TiO_2 nanoparticles were used as an additive for membrane fabrication.

On the other side, GO was also utilized as an additive to fabricate hydrophilic membrane. GO offers many advantages for water treatment due to its large surface area (calculated up to $2630 \text{ m}^2\text{g}^{-1}$) (X. Huang, Qi, Boey, & Zhang, 2012), good chemical stability and high mechanical strength (Young modulus $\sim 1\text{TPa}$) (C. Lee, Wei, Kysar, & Hone, 2008). As exfoliated GO contains the considerable number of oxygen-functional groups, such as carboxyl, carbonyl, epoxy, and hydroxyl groups, GO are highly hydrophilic (Miao, Li, Yan, Guo, & Lang, 2017; J. Zhang, Xu, et al., 2013). GO can be synthesized by using chemical approach, such as Hummers' method and electrochemical exfoliation. The common Hummers' method resulted a high quality of the synthesized GO. However, this method involved high hazardous chemical consumption, long time production and complicated procedures (Zaaba et al., 2017).

A low cost and simpler GO synthesis approach, which is electrochemical exfoliation method assisted by surfactant then offers several advantages such as greener, simpler and low cost production to synthesize GO (Nurhafizah et al., 2015; Parvez, Li, Puniredd, & Hernandez, 2013; Suriani, Muqoyyanah, Mohamed, Othman, Rohani, et al., 2019). Many reports of GO-incorporated membranes for wastewater treatment systems have also successfully increased membrane hydrophilicity, rejection for different heavy metal (Kochameshki, Marjani, Mahmoudian, & Farhadi,



2017) and dye pollutant (Safarpour, Vatanpour, & Khataee, 2016; M. Yang, Zhao, Zhang, Li, & Hou, 2017; Zhu et al., 2017). Nonetheless, the agglomeration of GO is the major drawback in the development of PVDF/GO membrane, which can reduce its performance (Sun & Li, 2018). In order to prevent GO from agglomeration, the utilization of surfactant for better dispersion of GO in the polymer matrix is essentially needed.

Previous report showed that the highest CNT dispersion was achieved by using the triple-tail sodium 1,4-bis(neopentyloxy)-3-(neopentyloxycarbonyl)-1,4-dioxobutane-2-sulphonate (TC14) surfactant as compared to single- or double-tail surfactant (Mohamed et al., 2014). Suriani, Nurhafizah, et al. (2016) showed that the utilization of triple-tail TC14 surfactant has presented smooth surface with low agglomeration of GO when the synthesized GO was intermixed with natural rubber latex. The triple-tail surfactant offers triple interaction during electrochemical exfoliation which resulted higher dispersion of GO. This result was also in a good agreement when the synthesized GO utilizing triple-tail TC14 surfactant was used to fabricate thin film and resulted higher DSSCs efficiency as compared to the single-tail surfactant (Suriani, Fatiatun, et al., 2018; Suriani, Muqoyyanah, Mohamed, Mamat, et al., 2018; Suriani, Muqoyyanah, Mohamed, Othman, et al., 2018). These confirmed that the surfactant's tail number essentially affect the quality of the synthesized GO. Previous report has also proved that the direct GO synthesis utilizing single-tail sodium dodecyl sulphate (SDS) surfactant and N, N-dimethylacetamide (DMAc) as solvent was successfully achieved and can be used to fabricate NF membrane which resulted high dye rejection of 92.76% (Suriani et al., 2019).



In this work, the utilization of both customized double-tail sodium bis(3,5,5-trimethyl-1-hexyl) sulphosuccinate (AOT4) and triple-tail TC14 surfactants were then carried out in order to improve membrane performance. In addition, it is believed that better dispersion and homogeneity of GO-based membrane sheet were achieved and resulted better performance for water treatment application. To the best of our knowledge, the novelty of this study lies on utilization of customized double-tail AOT4 and triple-tail TC14 surfactants for the GO synthesis by electrochemical exfoliation method for dye rejection application. It is believed that the utilization of both surfactants in the fabrication of the PVDF/GO-based hybrid membrane will increase the hydrophilicity, water flux, dye rejection and antifouling performance.

The second method to overcome dye contamination is via adsorption.

Adsorption process is considered as a versatile and effective method for dye removal due to its advantages such as low cost, high efficiency, eco-friendly and ease in operation (Cheng et al., 2017; Narvekar, Fernandes, & Tilve, 2018; Novais, Caetano, Seabra, Labrincha, & Pullar, 2018). A number of different adsorbents type have been employed for adsorption process including activated carbon (AC) (Marrakchi, Ahmed, Khanday, Asif, & Hameed, 2017; Novais et al., 2018), metal oxide (Taman, Ossman, Mansour, & Farag, 2015), bio sorbents (Gurung et al., 2013) and zeolites (Mateen et al., 2015). Among these adsorbents, AC becomes the most promising material and widely employed in adsorption process due to its convenient properties such as large surface area, high adsorption capacity (q_e) and possesses microporous structure (Rashid, Tehreem, Rehman, & Kumar, 2019; Solgi, Najib, Ahmadnejad, & Nasernejad, 2017).

Nevertheless, the commercial AC are uneconomical which hinder their widely used as adsorbent since they are fabricated from expensive precursors and non-renewable material such as coal and lignite (Rafatullah, Sulaiman, Hashim, & Ahmad, 2010). Regarding the aforementioned limitation, the development of a low cost and renewable carbonaceous material (Geçgel, Özcan, & Gürpınar, 2013) but possesses equivalent q_e as commercial AC has widely done (Rashid et al., 2019). This has led to the production of AC by utilizing agricultural waste variety as a precursor including pumpkin peels (Rashid et al., 2019), palm kernel shell (Nicholas, Hussein, Zainal, & Khadiran, 2018), banana stem (Misran, Bani, Situmeang, & Purba, 2018), lapsi seed stone (Sahira, Mandira, Prasad, & Ram, 2013), papaya peel (Musa, Alwi, Ngadi, & Abbaszadeh, 2017) and *Ficus carica* bast (Pathania, Sharma, & Singh, 2013).

In Malaysia, about 209,343 metric tons of durian fruit are produced in year 2017 and its production are estimated to be increased in year 2020 (M. C. Lee et al., 2018). The durian fruit consists of 40% of flesh and 60% of waste. Durian shell (DS) has no economic value thus usually burned without taking care of the surrounding environment. DS is suitable for preparing AC due to the abundance number and its availability which makes them a good source as a raw material in AC production. On the other hand, this AC conversion using agricultural waste would enhance the value of these agricultural commodities and at the same time decrease the waste disposal cost and provide potentially alternative on economic AC. AC from agricultural waste can be produced via two methods which are physical and chemical activation (Nicholas et al., 2018; Solgi et al., 2017; Xianlong Zhang, Cheng, Wu, Tang, & Wu, 2015).



Physical activation involves the carbonization of the precursors in an inert gas before activated it by using oxidizing agents such as steam and carbon dioxide. Meanwhile, chemical activation is a single step method which includes the process of carbonization and activation simultaneously occurs. In chemical activation, the impregnation process of the precursor can be done with chemical activator such as sulphuric acid (H_2SO_4) (Narvekar et al., 2018; Pathania et al., 2013), zinc chloride (ZnCl_2) (Xianlong Zhang et al., 2015), potassium hydroxide (KOH) (Musa et al., 2017; Novais et al., 2018) and phosphoric acid (H_3PO_4) (Misran et al., 2018; Nicholas et al., 2018). Comparing both methods, chemical activation offers several advantages including less energy usage, low operating temperature and higher carbon yields (Solgi et al., 2017). Furthermore, pore structure development can be modified by changing the parameters such as temperature during carbonization process, impregnation ratio and type of activating agent (Xianlong Zhang et al., 2015).

The q_e of AC is mainly depends on the type of activator agent, pyrolysis temperature and activation time (Rafatullah et al., 2010). Among several activator agents, H_2SO_4 is known as a super oxidizing agent that has been widely used for the preparation of carbonaceous adsorbents (Jawad, Razuan, Appaturi, & Wilson, 2019). Jawad et al. (2019) has proved that watermelon peels were successfully converted into AC by utilizing H_2SO_4 and presented excellence performance on methylene blue (MB) removal from aqueous solution. Meanwhile, Sahira, Mandira, Prasad, and Ram (2013) showed that AC was successfully prepared from lapsi seed stone which had been initially impregnated with H_2SO_4 . High q_e was also achieved as compared to the utilization of others activator agent due to a well-developed mesoporosity of H_2SO_4 (Sahira et al., 2013).



AC with the different surface area can be synthesized from the same precursor with different carbonization time and temperature. Recently, Nicholas et al. (2018) showed that carbonization temperature of 500°C produced the highest AC's surface area. However, lower surface area was observed when the carbonization temperature was increased up to 700°C. This result was in good agreement with the data presented by Srinivasakannan, Zailani, and Bakar (2004). They found that optimized carbonization temperature was around 400-500°C depends on impregnation ratio and time of carbonization. They also found that the optimum carbonization process time was around 1-3 hours. However, by increasing carbonization time, lower AC product and higher surface area was achieved.

Most studies reported the production of AC from DS by utilizing the various activator agent with different carbonization temperature and time (Chandra, Mirna, Sunarso, Sudaryanto, & Ismadji, 2009; Ismail, Sudrajat, & Jumbianti, 2010; Tham, Latif, Abdullah, & Taufiq-Yap, 2010; Mahmood, Samsuddin, & Deris, 2015). However, there is insufficient information of obtaining the durian shell-based AC (DAC) by utilizing H₂SO₄. Therefore, in the present study, the DS as an agricultural waste is used as a precursor to produce AC by utilizing H₂SO₄ as the activator agent via chemical activation. Furthermore, the synthesized DAC is further used to investigate its performance for MB removal in the adsorption process.

1.3 Problem Statement

PVDF polymer has widely been applied in membrane separation process for water treatment application since it has outstanding physical and chemical properties. However due to its hydrophobic nature, the application of the PVDF membrane is limited and becomes the main causes of membrane fouling. The membrane fouling causes a decline in water flux permeability, shortening membrane lifetime, altering membrane selectivity and affecting its dye rejection performance. Consequently, several approaches have been developed to increase the hydrophilicity of the PVDF membrane including physical and chemical modification with organic and inorganic material. Among those approaches, physical blending with organic and inorganic material has been widely used due to its advantage of facile preparation method which is through phase inversion. In addition, phase inversion method commonly used since it is applicable for various polymers and easy optimization of membrane thickness and pore size.

Recently, inorganic material-blended PVDF such as metal oxides particle, CNT and graphene have been utilized to enhance the PVDF membrane properties which increased antifouling effect, permeability and membrane selectivity. TiO_2 is considered as a potential candidate to reinforce PVDF polymeric material due to its properties such as commercially availability, chemical stability, low toxicity, and hydrophilicity. On the other hand, GO also offers several advantages for water treatment due to its large surface area, high chemical stability and contains abundance number of oxygen-functional group. Therefore, GO is considered as a highly

hydrophilic material. Nonetheless, agglomeration of GO is the major drawback in the development of PVDF/GO-based membrane.

As compared to other methods, electrochemical exfoliation method presents a simpler synthesis process of GO. To date, the utilization of the customized double- and triple-tail surfactants, AOT4 and TC14, respectively has proven the effectiveness for better exfoliation as compared to the commercially available single-tail SDS surfactant. In addition, the utilization of surfactant compound for better dispersion of GO in the polymeric membrane has also been proven. However, the surfactant for GO dispersion in polymeric membrane is limited to single-tail surfactant (SDS). The utilization of both double-tail AOT4 and triple-tail TC14 surfactants were believed resulted in better homogeneity and dispersion of GO in the polymeric membrane.

Furthermore, it also believed that higher surfactant's tail number provided the extra chain to interconnect between GO and PVDF polymer and resulted in increasing of hydrophilic properties of PVDF membrane.

AC for the adsorption process has also gained a lot of interest due to its characteristics such as high q_e , large surface area, and possesses microporous structure. However, the commercial AC is expensive since they are obtained from costly precursor and non-renewable material. In recent year, many researches have been performed on finding the effective and economical source of AC with high q_e which equivalent to the commercially available AC. Variety of agricultural waste has also been employed on AC production. Production of AC from agricultural waste not only provides a potential low cost of AC but overcome the environmental problem since their disposal can cause pollution. AC can be synthesis by two methods which

are physical and chemical activation. However, chemical activation presented more advantages than physical activation, such as require less energy, low operating temperature, resulted high carbon product and controllable of pore formation. DS is a potential candidate to be applied as a precursor for AC production since its abundance presence and availability of agricultural waste. The previous report on DAC is more focused on using KOH and H_3PO_4 as a chemical activator. However, there is insufficient research on using H_2SO_4 as a chemical activator on DAC and its performance on dye removal.

Therefore, in this work, GO-based material synthesized from the simple electrochemical exfoliation assisted by customized double-tail AOT4 and triple-tail TC14 surfactants are used for membrane application. Meanwhile, AC was produced using DS as starting material via chemical activation for adsorption process to investigate its performance on MB removal for water treatment application.

1.4 Research Objectives

The objectives of this study are:

- i. To synthesize GO solution using electrochemical exfoliation assisted by customized double-tail AOT4 and triple-tail TC14 surfactants.
- ii. To fabricate PVDF/GO/ TiO_2 -based hybrid membranes by using phase inversion method for dye rejection application.
- iii. To produce durian-based AC for dye removal application.

1.5 Scope and Limitations of Study

The GO synthesis was performed using graphite rod as a carbon precursor via electrochemical exfoliation method instead of other graphite material. The utilization of the surfactants for GO synthesis is also limited to the customized double-tail AOT4 and triple-tail TC14 surfactants. The scope of this study was to optimize the membrane performances which include the effects of utilizing the various surfactants on dye rejection and antifouling performance.

Meanwhile, H_2SO_4 is utilized as an activator agent instead of others in the production of AC from DS. The production of DAC is limited to 500°C of carbonization temperature for 3 hours of carbonization time. The scope of this research is based on the optimization of the effect of DAC dosage, contact time, and initial MB dye concentration.

1.6 Thesis Organization

This work is focused on the water treatment applications based on carbon-based material, which are GO and AC material. The improvement of dye rejection and antifouling performance by utilizing two types of surfactants are done for membrane separation. Meanwhile, the adsorption process is investigated by utilizing AC produced from DS via chemical activation. Chapter 1 consists of research background, research problem, research objective and scope and limitations of the study. Meanwhile, Chapter 2 presents the previous study and theories about



membrane separation and adsorption process in brief. The fabrication method of membrane and AC and its characterization techniques are discussed in Chapter 3. Chapter 4 explains the results and discussion including the morphology, structural and performance on dye removal for membrane separation and adsorption process. Finally, Chapter 5 covers the conclusion and suggestion for future work.

