MONTMORILLONITE CLAY-BASED MEMBRANE DISTILLATION FOR SEAWATER DESALINATION

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ABSTRACT

According to the World Health Organization (WHO), water shortage affects more than 66% of the world population. Seawater membrane distillation (MD) is one of the promising ways to solve water scarcity. Montmorillonite is the least explored low cost clay with unique properties that can act as an alternative material in membrane fabrication. This research focused on the development of hydrophobic montmorillonite clay-based hollow fibre MD for seawater desalination. The membranes with three different weight percentages of ceramic loading (36 wt. %, 38 wt. %, and 40 wt. %) were produced via phase inversion technique at a constant air gap distance and bore fluid flow rate before sintered at the temperatures of 1100, 1200, and 1300 °C. The clay hydrophilicity nature was transformed into hydrophobic property by surface modification through dip-coating with 1H, 1H, 2H, 2H-perfluorooctyltriethoxysilane solution. The physical and chemical properties of the prepared membranes were characterised using several analyses, including morphologies, mechanical stabilities, water permeability and hydrophobicity. The membrane performance was further evaluated using membrane distillation test with different concentrations of sodium chloride (NaCl) solution varied from 10,000 to 40,000 ppm and different feed temperatures varied from 50 to 80 °C under a steady condition of 20 °C permeate temperature at 1 atm. The characterization had shown that this material is equipped with the qualities to produce good clay-based hollow fibre MD for seawater desalination. The results also showed that the best performing membrane was spun at 40 wt. % ceramic loading, 5 cm air gap, 9 ml/min extrusion rate, 10 ml/min bore fluid flow rate and sintered at 1200 °C. The hydrophobic surface modification had been successfully proven by the results of the contact angle at the average of 132°. The salt rejection performance demonstrated that the montmorillonite clay-based hollow fibre membrane achieved 95% rejection. The outcome of this study is a novel and beneficial approach, especially in term of material selection, fabrication technique, and the performance in removing salt from seawater to produce fresh water.

ABSTRAK

Menurut Pertubuhan Kesihatan Sedunia (WHO), isu kekurangan air menjejaskan lebih daripada 66% penduduk dunia. Penyulingan membran (MD) air laut merupakan salah satu cara untuk menyelesaikan isu kekurangan air. Montmorillonit adalah tanah liat berkos rendah yang paling kurang diterokai dengan ciri-ciri unik yang mampu bertindak sebagai bahan alternatif dalam fabrikasi membran. Kajian ini memberi tumpuan terhadap pembangunan penyulingan membran gentian berongga hidrofobik berasaskan tanah liat montmorillonit untuk penyahgaraman air laut. Membran dengan tiga muatan seramik berbeza (36 wt. %, 38 wt. %, dan 40 wt. %) telah dihasilkan melalui kaedah penyongsangan fasa pada jarak sela udara dan kadar aliran cecair larik yang tetap sebelum disinter pada suhu 1100, 1200, dan 1300 °C. Sifat hidrofilik tanah liat diubah suai kepada sifat hidrofobik melalui pengubahsuaian permukaan melalui proses salutan celupan 1H, 1H, 2H, 2H-perfluorooktiltrietoksisilana. Ciri-ciri fizikal dan kimia membran yang disediakan dicirikan menggunakan beberapa analisis termasuk morfologi, kestabilan mekanik, ketelapan air dan kehidrofobikan. Prestasi membran seterusnya dinilai menggunakan ujian penyulingan membran dengan larutan natrium klorida (NaCl) yang berbeza kepekatan daripada 10,000 hingga 40,000 ppm pada suapan yang berbeza daripada 50 hingga 80 °C dan suhu bahan telap 20 °C pada 1 atm. Pencirian menunjukkan bahan ini dilengkapi dengan kualiti yang diperlukan untuk menghasilkan penyulingan membran gentian berongga MD berasaskan tanah liat yang bagus untuk penyahgaraman air laut. Hasilnya turut menunjukkan membran berprestasi terbaik dihasilkan pada muatan seramik 40 wt. %, sela udara 5 cm, kadar penyemperitan 9 ml/min, kadar aliran larik cecair 10 ml/min, dan disinter pada suhu 1200 °C. Pengubahsuaian permukaan hidrofobik telah berjaya dibuktikan oleh keputusan sudut sentuh pada purata 132°. Prestasi penolakan garam menunjukkan membran gentian berongga berasaskan tanah liat montmorillonit mencapai 95% penolakan. Hasil kajian ini adalah pendekatan baharu dan bermanfaat terutamanya dalam pemilihan bahan, teknik fabrikasi, dan prestasi dalam menyingkirkan garam daripada air laut untuk menghasilkan air tulen.

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LIST OF ABBREVIATIONS

WHO	-	World Health Organization
MD	-	Membrane Distillation
SEM	-	Scanning Electron Microscopy
XRD	-	X-Ray Diffraction
PVDF	-	Polyvinylidene Fluoride
PP	-	Polypropylene
PSF	-	Polysulfone
PE	-	Polyethylene
PI	-	Polymide
TGA	-	Thermal Gravimetric Analysis
MSF	-	Multi-Stage Flash Distillation
MED	-	Multi-Effect Distillation
VCD	-	Vapour Compression Distillation
ED	-	Electrodialysis
EDR	-	Electrodialysis Reversal
RO	-	Reverse Osmosis
DCMD	-	Direct Contact Membrane Distillation
PES	-	Polyethersulfone
NMP	-	N-methyl-2-pyrrolidone
PTFE	-	Polytetrafluorethylene
AFM	-	Atomic Force Microscopy
BSA	-	Bovine Serum Albumin
PVP	-	Polyvinylpyrrolidone
GCC	-	Gulf Cooperation Council
VTE	-	Vertical Tube Evaporation

LIST OF SYMBOLS

°C	-	Degree Celsius
%	-	Percent
mm	-	Millimetre
µ	-	Micro
g	-	Gram
mol	-	Molar
wt	-	Weight
rpm	-	Rotation per minute
min	-	Minute
cm	-	Centimetre
N	-	Newton
D_o	-	Outer diameter
D_i	-	Inner diameter
mg	-	Milligram
M	-	Mega
Pa	-	Pascal
n	-	Nano

CHAPTER 1

INTRODUCTION

1.1 Research Background

About 66% of the world population, which is nearly four billion people, live without sufficient access to fresh water for at least one month of the year whereas half a billion people in the world face severe water scarcity all year round (Mekonnen and Hoekstra, 2016). Severe water scarcity is defined as the depletion of water in certain areas. The World Economic Forum has placed the world water crisis in the top three of global problems alongside climate change and terrorism. According to the World Health Organisation (WHO), about 2.4 billion people do not have access to basic sanitation facilities, and more than one billion people do not have access to safe drinking water (Singh, 2015).

The world water crisis becomes serious when 36 countries faced with extremely high levels of baseline water stress. The agricultural sector is by far the highest user of clean water around the globe. About 80%–90% of all fresh water available is consumed by the sector (Hamdy *et al.*, 2003). This means that more than 80% of the water available for agricultural, domestic, and industrial users are withdrawn annually. Therefore, the problem will leave businesses, farms, and communities vulnerable to water scarcity. Unclean water causes diarrhoea, cholera, dysentery, guinea worm infection, typhoid, intestinal worm infection, and trachoma. According to the WHO, four billion people suffer from diarrhoea every year, which kills nearly 1.8 million people, with 90% of them are children under the age of five (Meyer, 2006).

Figure 1.1 shows the average exposure of water users in each country to water stress, which is the ratio of total withdrawals to total renewable supply in a given area. A higher percentage means more water users are competing for limited supplies (Gassert *et al.*, 2013).

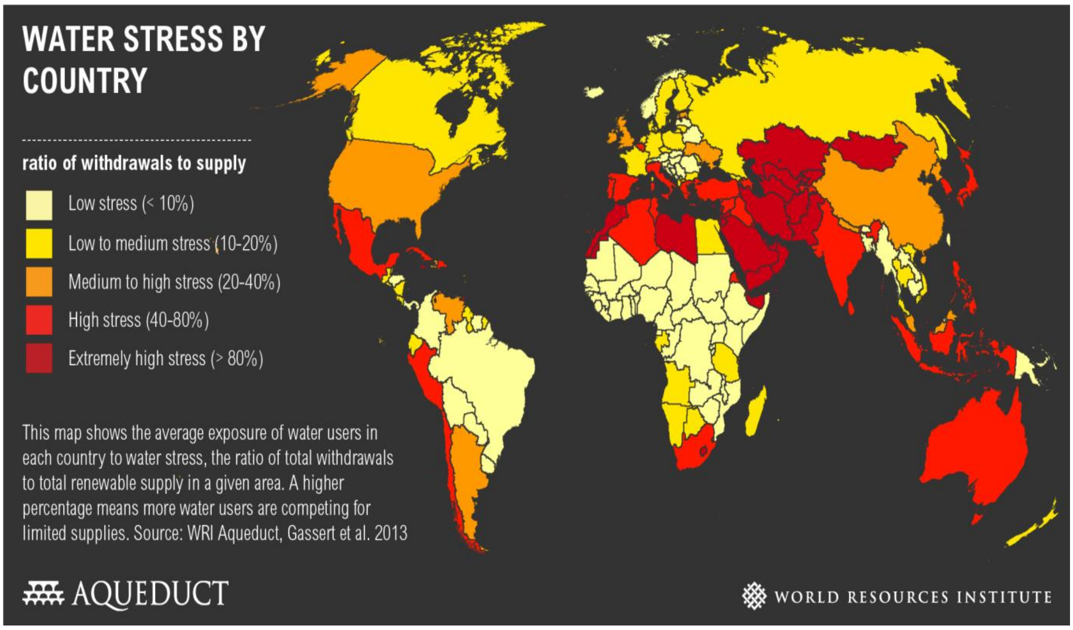

Figure 1.1 Water stress by country (Reig *et al.*, 2014)

75% of the earth is nearly covered by water but 97% of the water is salty ocean water. Most of the remaining fresh water is frozen in glaciers and ice sheets, which makes this source not readily available to use. Only a little less than 1% is all of the fresh water that is available on the earth to be shared with animals, plants, and humans. The availability of water decreases with increasing environmental pollution (Gryta, 2011). The solution to the problem is to fulfil the increasing demand of water supply by developing an efficient technology to turn seawater into fresh water.

Desalination is an available technology that separates salt from saline water to produce fresh water. Desalination process requires energy in the form of thermal, mechanical, or electrical to reject brine or concentrate from seawater or brackish water in order to produce fresh water. Desalination can be conducted through two methods, which are thermal and membrane. The thermal method, such as multi-stage flash, requires energy to heat up the brine until it vaporises at the temperature ranging from 60 to 100 °C. The drawback of this method is the process occurs at the surface of water, which means the efficiency of producing the condensate is based on the surface area involved. The higher the surface area involves, the more condensate forms in a short period. The second method is by using a pressure-driven membrane like reverse osmosis to separate salt from saline water. The drawback of this method is the high pressure required to force water particles to pass through the membrane without allowing solid salt particles to pass through. Operating at high pressure is equal to high energy cost. The end product using this method is called permeate.

Membrane distillation (MD) is basically the combination of both thermal and membrane methods to overcome both individual drawbacks. MD is a thermally-driven process that involves the transport of water vapour through a porous hydrophobic membrane. In MD, salt water is heated until it vaporises and the vapours are allowed to pass through the porous membrane. This process does not require a large surface area and high pressure to force the vapour to pass through the membrane. Theoretically, MD process enables the production of pure water from natural water. This study focuses on the development of a hydrophobic porous membrane for MD process.

1.2 Problem Statement

Recently, MD has become a popular option in the water treatment field (Ashoor *et al.*, 2016). Available polymeric membranes such as polyvinylidene fluoride (PVDF), polysulfone (PSF), and polymide (PI) are low cost and easy to fabricate but this type of membrane suffers the inability to perform in a harsh condition. Another alternative membrane material available is common ceramic membranes, such as alumina, zeolite, and silica. This type of membrane has superior structural, chemical, and thermal stabilities but at very high cost, which means ceramic membranes are not as cheap as montmorillonite.

Studies have shown that clay-based membrane is able to perform in harsh conditions, such as high temperature and highly chemical condition, and there is no significant change in terms of morphologies and properties during long time operation. Clay-based membrane also has superior structural, chemical, and thermal stabilities, which are as good as ceramic membrane but at a cheaper cost. Montmorillonite has not received enough attention as research on clay-based membrane only focuses on materials such as kaolinite, bentonite, and illite. Among these materials, montmorillonite poses the favourable properties to be developed as ceramic membrane. This is due to the abundant source, low cost, low sintering temperature (i.e., around 1,200 °C), and porous structure. These factors lead to the efficient cost of production. However, this material has never been developed as hollow fibre membrane. Therefore, thorough investigation on the effect of fabrication parameters such as ceramic loading and sintering temperature on hollow fibre membrane properties have to be established. Furthermore another limitation of this type of membrane to be used in membrane distillation application is the hydrophilic property due to the nature of surface hydroxyl group (Ren *et al.*, 2015). The hydrophilic problem can be overcome by coating the membrane with a hydrophobic material. A study showed that ceramic membrane subjected to hydrophobic modification possessed the contact angle more than 150° is able to reject almost 100% for desalination application through MD (Kujawski, 2004). This result makes a bold statement that ceramic membrane can be used as an alternative to polymeric membrane in MD.

1.3 Research Objectives

By referring to the problem statement mentioned above, the main goal of this study is to develop hydrophobic montmorillonite clay-based membrane distillation for seawater desalination via phase inversion, sintering, and dip-coating techniques. The specific objectives of this study are as follows:

i. To fabricate and characterise porous montmorillonite hollow fibre membrane via phase inversion/sintering technique in terms of their physical and chemical behaviours.

ii. To graft and characterise a hydrophobic layer onto montmorillonite hollow fibre membrane through silane crosslinking process and examine its physical and hydrophobic properties.

iii. To evaluate the performance of montmorillonite hollow fibre in membrane distillation for seawater desalination.

1.4 Research Scopes

In order to achieve the objectives, the following scopes have been considered:

i. Preparing different loadings of montmorillonite dope suspension from 36 to 40 wt. % for hollow fibre fabrication.

ii. Producing montmorillonite hollow fibre precursor by extruding the dope suspension based on dry/wet phase inversion spinning technique.

iii. Performing heat treatment to produce montmorillonite hollow fibre membrane at different target temperatures from 1,100 to 1,300 °C.

iv. Characterising the resultant membranes by using X-ray diffraction (XRD), X-ray fluorescence (XRF), scanning electron microscopy (SEM), mechanical strength, and water permeation.

v. Modifying the montmorillonite hollow fibre membrane surface by dip-coating with fluoroalkylsilane agent to induce the hydrophobicity property.

vi. Analysing the hydrophobic surface properties using liquid contact angle test and mercury porosimetry.

vii. Preparing the membrane module array as a membrane holder suitable for the membrane distillation system.

viii. Investigating the membrane performance of the membrane distillation system using synthetic saline water to evaluate the effect of feed concentration in range between 10,000 to 40,000 ppm and feed temperature from 50 to 80 °C on flux and salt rejection.

1.5 Research Significance

Although research on the desalination of seawater has been widely published for the past few decades, the outcome to solve the problem is not convenient in terms of cost, since the focus is more on polymeric membrane compared to ceramic membrane. The disadvantages of polymeric membrane including wetting problem, as well as morphological and property changes during long time operation make the research of fabricating clay-based hollow fibre membrane more significant. In contrast, the use of common ceramic membrane in seawater desalination is not convenient due to the high cost issue. Therefore, recent studies on ceramic membrane have been focusing on low-cost kaolin in the attempt to introduce a cheaper ceramic material. In addition, montmorillonite is an alternative clay-based material that can be used to produce ceramic membrane. Furthermore, the study on montmorillonite is less exposed compared to other low-cost ceramic materials. Hence, this study can be a frontier research on the fabrication of montmorillonite hollow fibre membrane, which is new in the scope of ceramic membrane.

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