AEROGEL COATING ON THE COPPER MESH AS SUPERHYDROPHOBIC POROUS LAYER

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AEROGEL COATING ON THE COPPER MESH AS SUPERHYDROPHOBIC POROUS LAYER

by

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LIST OF SYMBOLS

Δm	Mass of permeate	g OR L
Δt	Process duration	min OR h
Å	Length	10^{-10} m
A	Surface area per mass	m^2/g
A_m	Membrane active area	cm^2 OR m^2
Cf	Final concentration of the oil in oil-water mixture	mg/L OR wt.%
Ci	Initial concentration of the oil in oil-water mixture	mg/L OR wt.%
d	Diameter	m OR cm OR nm
E	Dielectric strength	kV/mm
J	Permeate flux	L/m^2h OR $g/cm^2.min$
k	Thermal conductivity	W/mK OR mW/mK
T	Temperature	°C OR K
V	Dielectric voltage	kV
v	Velocity	m^2/s
ρ	Density	g/cm^3 OR mg/cm^3
σ	Tensile strength	N/mm^2

LIST OF ABBREVIATIONS

3D	Three-Dimensional
AFM	Atomic force microscopy
AR	Modified acrylate resin
C	Carbon
C/PPy	Collagen/polypyrrole
$CaCO_3$	Calcium carbonate
CF	Copper foam
CNFA	Ceramic fiber component
CNO	Carbon nano-onion
CNTs	Carbon nanotubes
CO_2	Carbon dioxide
Cu	Copper
Cu(OH)2	Copper (II) hydroxide
$CuSiO_3$	Copper (II) metasilicate
CVD	Chemical vapor deposition
DCMD	Direct contact membrane distillation

DI	Deionised water
DSC	Differential scanning calorimetry
EDX	Energy dispersive X-ray spectroscopy
F	Fluorine
FADETA	(4-((4-((11-ferroceneundecyl)oxy)phenyl)diazenyl)phenoxy)-diethylene triamine
$FeCl_3$	Iron (III) chloride
FTIR	Fourier transform infrared spectroscopy
HEA	2-hydroxyethyl acrylate
IPA	Isopropyl alcohol
LTCVD	Low-temperature chemical vapor deposition
MD	Membrane distillation
MED	Multi-effect distillation
$MgCl_2$	Magnesium chloride
MgF_2	Magnesium fluoride
MSF	Multi-stage flash distillation
MWCNTs	Multi-walled carbon nanotubes

NaCl	Sodium chloride
NMP	N-Methyl-2-pyrrolidone
O	Oxygen
OPU	Optically active polyurethane
PANI	Polyaniline
PdI	Polydispersity index
PDMS	Polydimethylsiloxane
PE	Polyethylene
PEI	Poly-ethyleneimine
POSS	Polyhedral oligomeric silsesquioxane (POSS)
PP	Polypropylene
PTFE	Polytetrafluoroethylene
PUR	Polyurethane resin
PVA	Polyvinyl alcohol
PVDF	Polyvinylidene fluoride
SBMA	Sulfobetaine methacrylate
SEM	Scanning electron microscopy

Si	Silicon
Si_3N_4	Silicon nitride
SiAG	Silica aerogel
SiO_2	Silicon dioxide
SMA	Styrene-maleic anhydride
Sn	Tin
SWCNTs	Single-walled carbon nanotubes
VC	Vapor-compression
VLS	Vapor-liquid-solid
VMD	Vacuum membrane distillation
VS	Vapor-solid
WCA	Water contact angle
WSA	Water sliding angle

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SALUTAN AEROGEL PADA JARINGAN TEMBAGA SEBAGAI LAPISAN BERLIANG SUPERHIDROFOBIK

ABSTRAK

Sejak kebelakangan ini, rawatan air sisa menggunakan pelbagai teknik pengasingan telah menjadi tumpuan ramai penyelidik. Pembangunan perindustrian yang drastik menyebabkan peningkatan dalam penggunaan jumlah minyak yang membawa kemudaratan besar kepada kesihatan awam dan alam sekitar. Walau bagaimanapun, ketinggalan perkembangan dalam aspek teknikal dan pengurusan telah menyebabkan pencemaran air sisa berminyak. Proses pengeluaran minyak dari industri minyak, penapisan minyak, penyimpanan minyak, pengangkutan, dan industri petrokimia menghasilkan banyak air sisa berminyak di mana cara merawat sumber air sisa berminyak adalah sangat luas. Pengapungan, pembekuan, rawatan biologi, dan teknologi pemisahan membran merupakan kaedah rawatan konvensional yang sering digunakan (Yu et al., 2017). Memandangkan perkara ini, sebuah proses pemisahan baharu menggunakan penapis jaringan tembaga berliang superhidrofobik telah diwujudkan untuk memisahkan air daripada campuran minyak dan air. Dalam kajian ini, aerogel telah disalut pada jaringan tembaga dengan menggunakan teknik salutan celup di mana aerogel diuraikan dalam larutan etanol dan etanol/air. Aerogel juga telah dicampur ke dalam larutan polimer polyvinylidene fluoride (PVDF) dan didepositkan pada jaringan tembaga menggunakan teknik salutan tuang. Antara kaedah yang digunakan, aerogel yang diuraikan dalam larutan PVDF telah menawarkan salutan permukaan yang paling seragam. Dalam kaedah ini, saiz liang jaringan tembaga (jaringan bersaiz ‘500-mesh’ dan ‘80-mesh’), kepekatan aerogel, dan kepekatan polimer (PVDF) merupakan parameter yang telah dikaji. Penapis yang terbaik telah

diperolehi pada jaringan tembaga bersaiz ‘500-mesh’ yang disalut dengan 15g PVDF+NMP+1g aerogel. Jaringan yang bersalut mempamerkan tenaga permukaan yang rendah dengan sudut sentuhan air 150.40±2.31°. Purata fluks resapan bagi jaringan tulen dan jaringan bersalut ialah 218.91±3.85 and 351.90±3.93 $L/m^2.h$ masing-masing menunjukkan bahawa apabila hidrofobisiti meningkat, fluks resapan minyak tulen (heptana) juga meningkat. Walau bagaimanapun, keberkesanan penapisan air berminyak bagi jaringan bersalut adalah lebih rendah daripada jaringan tulen iaitu 61.45% dan 87.09% penolakan air masing-masing. Keberkesanan penapisan membran bersalut aerogel 15g PVDF+NMP+1g adalah lebih rendah daripada jaringan tulen kerana saiz liang telah dikurangkan yang mengurangkan keberkesanan pemisahan minyak dan air.

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ABSTRACT

Wastewater treatment using various separation techniques has become the focus of many researchers for some time now. There is an increase in the amount of oil used due to the escalating industrial development causing major harm to public health and environment. The lagging of the technical and management developments leading to oily wastewater pollution. Since the oil industry, oil refining, oil storage, transportation, and petrochemical industries in the production process produce a lot of oily wastewaters, treatment of oily wastewater sources becomes very broad. The conventional treatment methods include flotation, coagulation, biological treatment, and membrane separation technology (Yu et al., 2017). In view of this, a novel separation process using a superhydrophobic porous copper mesh filter was developed to separate the water from oil-water mixtures. In this study, aerogel was coated on the copper mesh via dip coating with the aerogel being dispersed in the ethanol and ethanol/water solution. Aerogel was also mixed into the PVDF polymer solution and cast on the copper mesh. Amongst the method, aerogel embedded within the PVDF solution offered the most uniform coating on the surface. Under this method, the other studied parameters include pore sizes of the copper mesh (500-mesh and 80-mesh sized meshes), aerogel concentration, and polymer (PVDF) concentration. The best filter was obtained on a 500-mesh sized coated with 15g PVDF+NMP+1g aerogel. The coated mesh exhibits low surface energy with the water contact angle of 150.40±2.31°. The average permeate flux for the pristine mesh and coated mesh are 218.91±3.85 and 351.90±3.93 L/m^2.h respectively indicating that when the hydrophobicity increases, the

permeate flux of pure oil (heptane) also increases. However, the oil-water filtration efficiency for the coated mesh is lower than the pristine mesh which are 61.45% and 87.09% water rejection respectively. The filtration efficiency of the 15g PVDF+NMP+1g aerogel coated membrane is lower than that of the pristine mesh due to the pore sizes being reduced, thus, reducing the efficiency of the oil-water separation.

CHAPTER 1

INTRODUCTION

A novel separation process can be carried out by developing a porous superhydrophobic copper mesh as a membrane or filter to separate the solutions of different polarity. Polymer coating such as aerogel can be used to coat the porous copper mesh resulting in a superhydrophobic porous membrane/filter. Depending on the size of the polymer nodules, its hierarchical structure could help to change the surface energy. The deposition of polymer coating on the copper mesh with different openings can be done via numerous coating methods such as sol-gel coating, spray coating, spin coating, and dip-coating as well as casting (Mekonnen et al., 2021). Thus, choosing the right coating material and coating method is vital to ensure the hydrophobicity and strength of the membrane for applications such as membrane distillation and oil-water separation.

1.1 Research Background

Filtration and adsorption are among the simplest and feasible methods in separation processes. The material is easy to be modified to perform separation on complex components (Tong, Q. et al., 2021). For example, membrane distillation (MD) is a membrane process in which the water vapour molecules from the wastewater can be selectively flow through the microporous hydrophobic membrane. Membrane distillation is a thermally driven separation process in which its driving force being the vapour pressure difference originated from the temperature difference across the hydrophobic membrane. It is highly sustainable that makes it widely applied in a plethora of industries such as wastewater treatment, desalination, and in the food industry (Alkhudhiri et al., 2011). As for the oil-water separation, it is a process of separating oil from water in wastewater treatment for which a superhydrophobic membrane is developed due to their high oil affinity and water repellence, thus, making the separation of mixture of various organic solvents with water into individual components easily (Singh et al., 2018).

In conjunction of achieving a good separation in membrane distillation as well as oil-water separation, utilising superhydrophobic materials may realise the performance required for it. Although majority of superhydrophobic materials face significant difficulties in achieving large-scale production in the preparation processes, a porous hydrophobic copper mesh with different openings is commonly utilised as oil-water separator. The copper mesh with uniform pore size comes as a chemically stable supporting material that exhibits good performance in separation processes. It is to be noted that the water contact angle of the modified copper mesh could be more than 160° (Tong, Q. et al., 2021). The hydrophobic microporous copper meshes are also widely established due to their large-scale production, high filtration flux, and low cost as well (Li et al., 2019).

In the process of making a superhydrophobic system for the separation processes, a superhydrophobic polymer coating is deposited on the hydrophobic copper mesh. The conventional polymer coating materials utilised for the separation processes are such as epoxy resin, polydimethylsiloxane (PDMS), polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), polyethylene (PE), and polypropylene (PP) (González et al., 2017). Apart from that, other insulating materials such as aerogel can be used for coating of copper mesh as it has low thermal conductivity and low surface energy. The aerogel, in particular, is highly focused as it is a superhydrophobic and thermally insulating coating material due to its highly porous structure. Due to this, it is also known as the best thermal insulator in the world. This property exists because it contains only air particles in the absence of water which makes it very light as well (Thapliyal & Singh, 2014). In this study, due to the focus being on the hydrophobicity of the coating material, aerogel is preferred. There are numerous types of aerogels, the most common ones being silica aerogel, carbon aerogel, and metal oxide aerogels. Silica aerogel has a great thermal insulating property, and due to its high availability in market, it is largely favoured for filtration processes such as membrane distillation and oil-water separation. It is also largely applied in various industries such as space science, as adsorbents, sensors, catalysts, storage media, antifouling materials, and as host for biomaterials or drugs. It has large porosity which is greater than 95%, density of 3-350 mg/cm^3, large surface area, as well as low thermal conductivity ranging from 0.004-0.03 W/mK (Duan et al., 2012).

In order to produce the aerogel coated copper mesh, the superhydrophobic aerogels must be first dispersed in organic solvents before it is deposited on the hydrophobic copper mesh, thus, making the mesh more superhydrophobic. As far as the coating method is concerned, out of all the coating methods available such as casting, spray coating, dip coating, spin coating, and sol-gel coating, spray coating, dip coating, and casting are relatively popular due to its simple operation. Dip-coating and casting does not contain any complicated steps which makes it more advantageous (Mekonnen et al., 2021). The coating methods differ according to the type of solvent used to disperse the aerogel and more importantly, the viscosity of the coating solution prepared.

Meanwhile, separation processes such as filtration and membrane distillation are widely used in industries like the wastewater industry. In order to ensure the sustainability of the wastewater industry such as in the removal of oil from the oil-water mixtures, filtration can be done by using copper mesh as filter with the required modifications making it more superhydrophobic. Since aerogel has superhydrophobic and thermal insulating properties, it is utilised as a coating material for the copper mesh. The persistent superhydrophobic property enables the reusability of the membrane and makes them cost-effective. Moreover, aerogels are flexible and could withstand high temperatures that makes it suitable for coating. Aerogels are also highly biocompatible making them compatible with various components. In addition, the whole process of dispersing the aerogels and coating can be carried out under ambient conditions, thus, it is classified as an eco-friendly approach. Overall, the simplicity of the process, and high availability of the materials utilised make it a sustainable process.

1.2 Problem Statement

In order to make a copper mesh to be used as a superhydrophobic insulating material for separation processes, the material chosen as a membrane should be coated with polymeric coating. The challenge arises when the coating is on the low surface energy material as the only possible interaction is via hydrophobic interactions. The process of choosing a suitable coating material therefore becomes crucial. Although there are numerous conventional coating materials being widely used such as epoxy resin, hydrogel, PDMS, PTFE, PVDF, PE, and PP, aerogels are being studied in more detail due to their exceptional thermally insulating and hydrophobic property (González et al., 2017). Aerogel can be derived from hydrogel with some steps being followed which is started with sol-gel process, aging, and finally drying (Zuo et al., 2015). The aerogel is produced when the water content in hydrogel is dried till there is only air particles present in it, thus, making it very light. Aerogel is favoured due to its ultra-low thermal conductivity which is lower than hydrogel and other coating materials as well