FABRICATION AND OPTIMIZATION PERFORMANCE FOR TREATMENT OF SHIP OILY WATER BY NANOCOMPOSITE MEMBRANES USING TAGUCHI METHOD

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TO MY BELOVED HUSBAND

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ABSTRACT

The objectives of this study were to optimize of ultrafiltration (UF) process using novel nanocomposite membrane for oily water treatment in cruise-ship. Three nanocomposite membranes consisted of polyvinylidene fluoride (PVDF) polymer, Zeolite Socony Mobil–5 (ZSM5) nanofiller, and N-methyl-2-pyrrolidion (NMP) polymer solvent have been produced which was denoted as M0, M1 and M2. N-β-(aminoethyl)-ɣ-aminopropyltrimethoxysilane (AEAPTMS) was used to modify ZSM5 to enhance the dispersion of nanocrystals in the polymeric solution. Nanocomposite structures have been characterized via field emission scanning electron microscopes (FESEM) combined with Energy-dispersive X-ray (EDX), thermal gravimetric analysis (TGA), Fourier Transform Infrared (FTIR), and contact angle. Taguchi method was employed to design experiments using three different parameters (A: membrane type, B: temperature, C: pressure) and to optimize the process performance via single response (Y0: selection parameter, 'SP'). Based on the Taguchi design and optimization, a significant model in analysis of variance (ANOVA) was predicted with the highest response (Y0 = 65.22 $L.m^{-2}.h^{-1}$). Taguchi also predicted an optimal operation condition which has achieved the highest oil rejection (99.7%) and promising permeation flux (105.89 $L.m^{-2}.h^{-1}$) by using nanocomposite membrane with 15 gram of PVDF/ 1 gram of M-ZSM5, at temperature 55°C and 2 bar pressure gauge.

ABSTRAK

Kajian ini adalah bertujuan untuk mengoptimakan proses ultraturasan (UF) menggunakan membran nano komposit yang novel untuk rawatan air berminyak dalam kapal pelayaran. Tiga membran nano komposit yang terdiri daripada polimer poliviniliden fluorida (PVDF), zeolit sokoni mobil-5 (ZSM5) sebagai pengisinano, dan N-metil-2-pirolidon (NMP) sebagai pelarut polimer telah disediakan dinamakan sebagai M0, M1 dan M2. N-β-(amino etil)-ɣ-aminopropiltrimetoksilane (AEAPTMS) telah digunakan untuk mengubahsuai ZSM5 untuk meningkatkan penyebaran nanokristal dalam larutan polimer. Struktur nano komposit telah dicirikan melalui mikroskopi medan pengimbas elektron (FESEM) dengan kombinasi sebaran tenaga sinar-X (EDX), analisis termal gravimetrik (TGA), inframerah transformasi Fourier (FTIR), dan sudut hubungan. Kaedah Taguchi telah digunakan untuk mereka eksperimen menggunakan tiga parameter yang berbeza (A: jenis membran, B: suhu, C: tekanan) dan mengoptimumkan prestasi proses berdasarkan tindak balas tunggal (Y0: parameter pilihan, 'SP'. Berdasarkan kepada rekabentuk dan pengoptimuman Taguchi, satu model penting di dalam analisis varians (ANOVA) telah diramalkan pada maklumbalas tertinggi (Y0=65.22 L. $L.m^{-2}.h^{-1}$). Taguchi turut meramalkan keadaan operasi yang optimum yang telah mencapai penyingkiran minyak tertinggi (99.7%) dan kebolehtelapan fluks air yang baik (105.89 $L.m^{-2}.h^{-1}$) dengan menggunakan membran nano komposit 15 gram PVDF/ 1 gram M-ZSM5 pada suhu 55 ^{o}C dan tolok tekanan 2 bar.

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LIST OF ABBREVIATIONS

Sc	-	Schmidt number
Sh	-	Sherwood number
C_P	-	Concentration polarization
CTA	-	Cellulose triacetate
DMF	-	Dimethylformamide
DMSO	-	Dimethyl sulfoxide
ECP	-	External concentration polarization
PMR	-	Photocatalytic membrane reactor
FTIR	-	Fourier Transform Infrared Spectroscopy
HTI	-	Hydration Technologies Inc
ICP	-	Internal concentration polarization
IPC	-	Isophthaloyl chloride
M_W	-	Molecular weight
NF	-	Nanofiltration
PA	-	Polyamide
PAI	-	Polyamide-imide
PAN	-	Polyacrylonitrile
PAS	-	Positron annihilation spectroscopy
PC	-	Phthloyl chloride
PDA	-	Phenylenediamine
PEG	-	Polyethylene Glycol
PEI	-	Polyethyleneimine
PES	-	Polyether sulfone
PI	-	Polyimide
PIP	-	Piparazine
PPDA	-	p-Phenylenediamine

PRO	-	Pressure retarded osmosis
PSf	-	Polysulfone
PS	-	Polystyrene
PSS	-	Poly(sodium 4-styrene-sulfonate)
PVDF	-	Thickness (m)
PVP	-	Polyvinylpyrolidone
PEG	-	Polyethylene glycol
PES	-	Polyethersulfone
PSf	-	Polysulfone
PRO	-	Pressure-retarded osmosis
RO	-	Reverse Osmosis
Re	-	Reynolds number
SEM	-	Scanning Electron Microscope
SDS	-	Sodium dodecyl sulfate
sPEEK	-	Sulfonated poly(ether ether ketone)
sPSf	-	Sulfonated polyethersulfone
TFC	-	Thin-film composite
TEA	-	Triethylamide
TMC	-	Trimesoyl chloride
TPC	-	Terephthaloyl chloride
UF	-	Ultrafiltration

LIST OF SYMBOLS

A	-	Water permeability coefficient (m^3/m^2.s.Pa)
B	-	Solute permeability coefficient (m/s)
C	-	Concentration of salt (mol/l)
$C_{F,b}$, $C_{D,b}$	-	Salt concentration of the bulk feed and draw solution
$C_{F,i}$, $C_{D,i}$	-	Concentration of feed and draw solution near membrane surface inside porous supports
$C_{F,m}$, $C_{D,m}$	-	Concentration of feed and draw solution near membrane surface
C_p, C_f	-	Salt concentration in permeate and feed solutions dh Hydraulic diameter (m)
D	-	Solute diffusion coefficient (m^2 s^{-1})
Do	-	The diffusion coefficient at an infinite dilution (m^2s^{-1})
De	-	Effective diffusion coefficient (m^2s^{-1})
d_p	-	pore diameter, nm
d_s	-	Solute diameter, nm
D_s	-	Diffusion coefficient of salt in the membrane substrate ($m^2.s^{-1}$)
IP	-	Interfacial polymerization
Js	-	Reverse salt flux (g m^{-2} h^{-1})
Jw	-	Water flux (m^3m^{-2} s^{-1})
K	-	Water transport coefficient ($m.s^{-1}$)
K_b	-	Mass-transfer coefficient ($m.s^{-1}$)
K_d	-	Mass-transfer coefficient (m s^{-1})
Kc	-	Mean mass transfer coefficient ($m.s^{-1}$)
K*	-	Solute resistance coefficient independent of diffusivity
Km	-	Solute diffusion resistivity within the porous layer (s m^{-1})
L/t	-	Thickness, m

m	-	Membrane weight, g
M	-	Molality
MWCO	-	Molecular weight cut-off (kDa)
P	-	Pressure (bar)
q	-	Material density ($g.cm^{-3}$)
R	-	Solute rejection
S	-	Membrane structural parameter (m)
T	-	Membrane thickness (m)
Vw	-	Partial molar volume of water (m^3s^{-1})
W	-	Power ($W\ m^2$) *Greek letters*
π	-	Osmotic pressure (Pa)
$\pi_{F,b}$, $\pi_{D,b}$	-	Osmotic pressure of the bulk feed and draw solution (Pa)
$\pi_{F,m}$, $\pi_{D,m}$	-	Osmotic pressure of feed and draw solution near membrane surface (Pa)
$\pi_{F,i}$, $\pi_{D,I}$	-	Osmotic pressure of feed and draw solution near membrane surface inside porous supports (Pa)
ΔP	-	Osmotic pressure (bar)
$\Delta\pi, \Delta\pi_{eff}$	-	Osmotic pressure difference and effective osmotic pressure difference (Pa)
ε	-	Membrane porosity
σ	-	Reflection coefficient
τ	-	Pore tortuosity
η_w, η_s	-	Viscosity of water and solution ($kg\ m^{-1}s^{-1}$)
μ	-	Chemical potential
γ	-	Activity coefficient

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CHAPTER 1

INTRODUCTION

1.1 Research background

Merchant ships such as small feeder ships, car carrier ships, giant container vessels, oil tankers, warships, and cruise ships have given many advantages to human life via huge carrying capacity and low cost shipping compared to other vehicles (McCARTHY and Khambaty, 1994; Huq *et al.*, 1996; Tsolaki and Diamadopoulos, 2010; Moslehyani *et al.*, 2015a). However, their oily water discharge is known to have a negative impact on the marine ecosystem. In particular, one of the merchant ship crew practice is using their clean ballast water to wash out ship oil or fuel tanks, which result in oil emulsion in ballast water (Oemcke and Leeuwen, 2004; Hewitt *et al.*, 2009). Afterward, that oily ballast water will be pumped offshore without proper treatment. This practice results in the spillage of oily ballast offshore, causing extensive ecological and economic damage to the aquatic ecosystems around the world, along with serious human health issues including death due to the discharge of toxic waste to the environment. This negative impact of merchant ship on marine and human life has become a subject for green global organizations since the late 1980s. Accordingly, the International Maritime Organization (IMO) adopted the Ballast Water Management (BWM) convention in February 2004, and its ratification is under way for having green environment. Prior to BWM, marine pollution

(MARPOL) was made by England in 1978 to prevent pollution from all types of ships (Fakhru'l-Razi *et al.*, 2009). The marine pollution 1973-1978 (MARPOL 73-78) conventions have regulated that merchant ships install a water purification system for their wastewater in order to purify water on board such that their oily wastewater discharge contains no more than 15 mg of oil per liter. In particular, merchant ships should store their retentate of treated oily ballast water or untreated oily ballast water in their segregated ballast tanks (SBT) until the vessel reaches a reception facility in the port (Yazu *et al.*, 2001). Regarding these regulations, the merchant ship industry has been looking forward to have green and advanced on board oily ballast water treatment. Hence, companies specializing in water and wastewater treatment systems are providing ballast water treatment systems and processes for merchant ships (Fakhru'l-Razi *et al.*, 2009). Currently, the potable ultraviolet (UV) irradiation application as the greenest and most advanced process is of interest to the merchant ship industries due to their promising performance (Moslehyani *et al.*, 2015b). Schematic diagram of a 20,000 deadweight (dwt) merchant ship elements are shown in Figure 1.1.

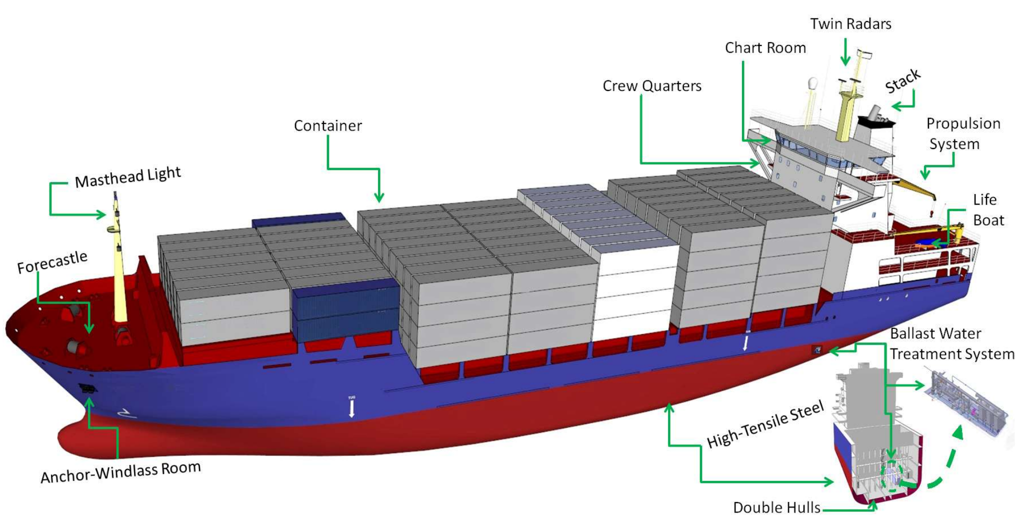

Figure 1.1: Schematic diagram of merchant ship (Moslehyani *et al.,* 2016)

Membrane separation processes such as reverse osmosis (RO) (Kwak *et al.*, 2001), nanofitration (NF) (Bowen *et al.*, 1997), ultrafiltration (UF) (Yan *et al.*, 2006) and microfiltration (MF) (Ellis *et al.*, 2000) has been widely used for hydrocarbon filtration. Among all membrane technologies, ultrafiltration (UF) is an efficient membrane process for ship oily water treatment (Cho *et al.*, 2000; Lee *et al.*, 2004). Nanocomposite membrane is one of the promising types of UF membrane which shown a great effect for wastewater treatment. Nanocomposite membrane usually fabricated via incorporating high efficient nanoparticles or nanotubes (fillers) into the polymer membrane matrix (Guo *et al.*, 2007). Organic and inorganic fillers have been interest due to their uniform dispersing, high mechanical and economical property for different industrial applications. Zeolite Socony Mobil–5 (ZSM-5) is one of the nanocomposite filler, which is a type of aluminosilicate zeolite belonging to the pentasil family of zeolites. Its chemical formula is $Na_nAl_nSi_{96-n}O_{192}\cdot 16H_2O$, ($0<n<27$), which was patented by Mobil oil company and it is widely used in the petroleum industry as a heterogeneous filler (Liu *et al.*, 2006). In comparison, ZSM-5 with single walled carbon nanotubes (SWCNTs) (Zhou *et al.*, 2012), multiwall carbon nanotubes (MWCNTs) (Fang *et al.*, 2010) and others, has showed high efficiency fillers into the morphological structure of membrane. For example, the modified ZSM-5 possess antifouling behavior which was one of the key factor for UF nanocomposite membrane in addition to low cost making if beneficial to be use as membrane filler (Zhang *et al.*, 2008). However, major drawback of these nanoparticles are they have low hydrophilicity and high agglomeration, which need to be minimized using some techniques such as functionalizing by silane group. However, in the long term process UF membrane is facing different defects such as non-optimized membrane composition and non-optimized applied condition which caused low efficiency in the whole membrane process. Thus, UF membrane process need to be designed with any Design of Experiment (DOE) computing techniques to optimize the controllable parameters in order to make the membrane process robust (Mohammadi *et al.*, 2004).

DOE computing enables designers to determine simultaneously the individual and interactive effects of many factors that could affect the output results in any design (Lendrem *et al.*, 2001). DOE also provides a full insight of interaction between design elements, therefore, helps turn any standard design into a robust one (Giunta *et al.*, 2003). Simply put, DOE helps to pin point the sensitive parts and sensitive areas in designs that cause problems in yield. Designers are then able to fix these problems and produce robust and higher yield designs prior going into production. Taguchi design method is one of DOE computing technique that lets designer to choose a product or process that functions more consistently in the operating environment (Giunta *et al.*, 2003). Taguchi designs try to identify controllable factors (control factors) in process that minimized the effect of the noise factors (Tsui, 1992). During Taguchi computing, designer manipulate noise factors to force variability to occur and then determine optimal control factor settings that make the process or product robust, or resistant to variation from the noise factors (Park *et al.*, 2006). A process designed with this goal will produce more consistent output. A product designed with this goal will deliver more consistent performance regardless of the environment in which it is used (Lakshminarayanan and Balasubramanian, 2008). Hence, membrane filtration parameters such as membrane composition, contaminant concentration, applied pressure and applied temperature are known to be significant factors that affect the performance of the process, which may be optimized using the DOE method to achieve higher performance process (Wu *et al.*, 2009).

1.2 Problem statement

Every year, two thousand tons of bilge water from bilge is discharged at sea and this huge quantity is a major pollution problem for marine ecosystems and more or less directly for human health. However, because on-board storage of bilge water is not always feasible, remediation is required and thus, different kinds of treatment have been proposed for these, at which physical processes are currently the most commonly-used techniques. The bilge water separators have been equipped in ships for bilge remediation.

However, there are still some engineering problems for this kind of separator which allowed some particles discharge at sea. Other problems are included (1) (the extremely long time required process), (2) (production of high toxic retentate) after the separation process, (3) (high cost engineering to maintain and protect separators). The other filtration techniques which can be used to replace the mechanic separators include chemical demulsification technique, UF membrane separation technique and etcetera. Among all treatment process, the UF membrane separation by using nanocomposite membranes is the most efficient due to use promising modifier like nanoparticles, nanofillers, and so on. On the other hand, some of the nanoparticles including carbon nanotubes, metallic oxides, aluminumsilicates have a drawback of agglomeration, which need to be more studied before using due their disadvantages on membrane performance. Since, some of the nanoparticles should modified using silane agents (e.g. Zeolites and clay) to minimized the agglomeration or replaced with another alternative nanoparticle due to the high agglomerations which cannot be minimized easily (e.g. silver oxide and multiwalled carbon nanotubes (MWCNTs)). In fact, in order to obtain the higher performance in terms of hydrocarbon rejection, lower flux decline and high water permeability, membrane process need to be optimized using one of optimization technique such as DOE computing. Therefore, in this study attempts were made to fabricate novel nanocomposite membrane and employing Taguchi method to optimize the process parameters.

1.3 Objectives of the study

Based on the background of this study, the objectives of this study are:

1) To fabricate and characterize novel nanocomposite PVDF/ modified ZSM-5 using AEAPTMS ultrafiltration membranes for oily water filtration.

2) To investigate the effects of membrane, applied temperature and applied pressure on response of process performance.

3) To optimize the membrane parameters using Taguchi method of DOE computing by computer software for higher process efficiency.

1.4 Scope of the study

To achieve the objectives of the study, the following scopes of the study have been identified:

1) Modifying the ZSM-5 via using silane functionalize group and characterizing the M-ZSM-5.

2) Embedding the M-ZSM-5 on the PVDF polymer matrix for PVDF/M-ZSM-5 flat sheet membrane fabrication.

3) Characterization of PVDF/M-ZSM-5 flat sheet membrane using transmission electron microscopy (TEM), field emission scanning electron microscopy (FESEM) combined with energy dispersive X-rays (EDX), Fourier transform infra-red spectroscopy (FTIR), contact angle and thermal gravimetric analysis (TGA).

4) Designing the experiment using Taguchi method for membrane separation process.

5) Evaluating the efficiency of the membrane separation process in terms of hydrocarbon rejection, flux decline and flux permeation.

1.5 Significance of the study

The significance of this study is by using novel membrane composition (PVDF/M-ZSM-5) and membrane process optimization using Taguchi method of DOE computing for bilge water treatment. Three different nanocomposite membrane has been prepared to be use for bilge water filtration. The applied membrane (A), applied temperature (B), and operating pressure (C) have been selected as three controllable process parameters and experimented randomly in 16 trials and three level by Taguchi method of DOE computing via computer software for higher performance achievement in this study.

3) Effect of PVDF/ZSM5s membrane on performance and fouling resistant of the nanofiltration and reverse osmosis membranes should be studied to achieve higher performance in membrane separation.

4) Effect of unmodified ZSM5s by other polymers (e.g. PES, PEI, PSf, PI and etcetera) for the different wastewater treatment applications.

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