SYNTHESIS AND CHARACTERIZATION OF PVDF/NaCl MEMBRANE FOR AMMONIA REMOVAL FROM AQUACULTURE

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ABSTRACT

SYNTHESIS AND CHARACTERIZATION OF PVDF/NaCl MEMBRANE FOR AMMONIA REMOVAL FROM AQUACULTURE

In this study, polyvinylidene fluoride (PVDF) flat membrane was fabricated to remove the total ammonia from saline water by direct contact membrane distillation (DCMD) at 50°C. The effects of initial NH_3 concentration from 0.03 to 0.47M, temperature from 30 to 50°C and salinity from 0 to 35ppt on ammonia-ammonium equilibrium, however, were initially investigated by using Law of Ostwald's Dissociation. The PVDF membranes were then fabricated from dimethylacetamide (DMAc) solvent and sodium chloride (NaCl) from concentration 0 to 6M as the dope additive by phase inversion in a distilled water coagulation bath. The membranes were dried and then characterized as well as applied in DCMD. Cocurrent configuration of DCMD was applied to study the mass transfer, water vapor flux and ammonia removal efficiency of the membranes. The experimental results showed the NH_3 concentration in the water increases as the temperature and pH increased. The NH_3 concentration, however, is relatively higher in pure water than that in saline water from temperature 30 to 50°C. The drying process has improved the hydrophobicity of the membranes. The contact angles of the membranes with NaCl addition, however, were reduced from 165 to 156°. Graphically, the average ranges of membrane thickness and pore size were 71.5 to 111.5μm and 43.5 to 118.5nm, respectively. The addition of NaCl has reduced the average pore size from 81 to 66nm. Statistically, average ranges of the membrane thickness and pore size were 83 to 99μm and 82 to 103.3nm, respectively. The average membrane porosity with additives were increased from 78% to >82% compared to that of without the additives. The mass transfer coefficient of ammonia and the water vapor flux increase as the membrane porosity increased. The membrane porosity of 84% and effective membrane area of 56.40cm^2 achieved the highest ammonia removal efficiency about 24% within 2 hours of DCMD.

ABSTRAK

Dalam kajian ini, membran polyvinylidene fluoride (PVDF) berkepingan telah disintesiskan untuk menyahkan ammonia dari air separa garam pada suhu 50°C menggunakan proses sentuhan terus penyulingan membran. Walau bagaimanapun, kesan daripada kepekatan NH_3 awal dari 0.03 hingga 0.47M, suhu dari 30 hingga 50°C dan kepekatan garam dari 0 hingga 35ppt terhadap keseimbangan ammonia-ammonium, telah dikaji dengan menggunakan Hukum Penceraian Ostwald. Membran PVDF kemudian disintesiskan daripada pelarut dimetilacetamide (DMAc) dan sodium klorida (NaCl), sebagai bahan tambah dari kepekatan 0 hingga 6M. Membran terbentuk melalui proses fasa kesongsangan dalam takungan air suling. Membran tersebut dikering dan dicirikan serta diaplikasikan dalam DCMD. Konfigurasi kokuren DCMD telah digunakan untuk mengkaji pemindahan jisim ammonia, flux wap air dan kecekapan pemisahan ammonia oleh membran-membran tersebut. Hasil kajian menunjukkan bahawa kepekatan ammonia dalam air akan tambah apabila suhu dan pH bertambah. Secara relatif, kepekatan ammonia dalam air tulin adalah lebih tinggi berbanding di dalam air garam, dari suhu 30 hingga 50°C. Proses pengeringan juga didapati meningkatkan kehidrofobikan membran-membran tersebut. Sudut sentuh bagi membran yang mengandungi NaCl berkurang dari 165 ke 156°, berbanding tanpa NaCl. Ketebalan dan saiz liang membran ditentukan secara graf masing-masing adalah 71.5 hingga 111.5µm dan 43.5 hingga 118.5µm. Pertambahan NaCl telah mengurangkan purata saiz liang dari 81 kepada 66nm. Secara statistik, purata ketebalan membran dan saiz liang masing-masing adalah 83 hingga 99µm dan 82 hingga 103.3nm. Keliangan bagi membran yang mengandungi NaCl meningkat dari 78% kepada >82%. Pemalar pemindahan jisim ammonia dan flux wap air juga bertambah apabila keliangan membran bertambah. Keliangan sebanyak 84% dan keluasan membran sebanyak 56.40cm² telah mencapai kecekapan pemisahan ammonia tertinggi iaitu sebanyak 24% dalam tempoh 2 jam DCMD beroperasi.

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LIST OF SYMBOLS

A	Surface area (m^2)
B	Permeance (mol/(m^2.s.Pa))
B_c	Pore size morphology constant (-)
c_{fb}	Concentration of solute in the feed bulk (mol/L)
c_{fm}	Concentration of solute at membrane surface in feed side (mol/L)
$c_{protein}$	Protein concentration in the feed (-)
c_{solute}	Solute concentration on the membrane (mol/L)
C_b	Solute concentration in the bulk (mol/L)
C_{gel}	Solute concentration in the gel layer (mol/L)
C_l	Membrane distillation coefficient (kg/m^2.s.Pa)
C_m	Solute concentration at the membrane surface (mol/L)
$C_{NH_3,0}$	Equilibrium concentration of ammonia at initial time (mol/L)
$C_{NH_3,t}$	Equilibrium concentration of ammonia at time t (mol/L)
C_p	Solute concentration in the permeate (mol/L)
C_P	Heat capacity of the feed (in heat transfer case) (J/kg.K)
d_h	Hydraulic diameter (m)
d_i	Inside diameter of hollow fiber (m)
d_p	Mean pore diameter (m)
D	Solute diffusion coefficient (m^2/s)
$D_{eff,\infty}$	Bulk diffusivity (m^2/s)
D_m	Diffusivity coefficient of ammonia gas in the membrane pores (m^2/s)
D_{eq}	Equivalent sand particle diameter (m)
D_{50}	Mean size, sieve size which will pass 50% of the granular media (m)
F	Feed mass flow rate (kg/day)
g	Acceleration of gravity (m^2/s)
h_f	Thermal boundary layer coefficient, feed side (W/m^2.K)
h_p	Thermal boundary layer coefficient, permeate solution (W/m^2.K)
H	Henry's constant (-)
H_{filter}	Head loss in the filter (m)

$\Delta H_{v,i}$	Heat transfer coefficient ($W/m^2.K$)
I	Ionic strength (mol/L)
J	Flux of ammonia ($kg/m^2.h$)
J_i	Independent permeate flux ($kg/m^2.h$)
k	Mass transfer coefficient of solute (m/s)
k_g^T	Thermal conductivity of the gas present in the pores (W/m.K)
k_m^T	Average membrane thermal conductivity (W/m.K)
k_s^T	Thermal conductivity of the membrane matrix (W/m.K)
K	Overall mass transfer coefficient (m/s)
K_a	Dissociation constant of acid (mol/L)
K_b	Dissociation constant of base (mol/L)
K_c	Equilibrium constant of the pure water (mol/L)
K_f	Mass transfer coefficients in feed solution (m/s)
K_m	Mass transfer coefficients across the membrane (m/s)
K_s	Mass transfer coefficients in permeate solution (m/s)
K_w	Ionic product constant (mol^2/L^2)
L	Length (m)
L_c	Effective lengths of channel (m)
L_m	Effective lengths of membrane (m)
L_p	Effective pore length (m)
m	Mass (kg)
m_{gs}	Average mass of one grain of sand (kg)
m_m	Dry weight of membrane (kg)
m_{mpi}	Weight of the pyknometer with IPA and membrane (kg)
m_{mpw}	Weight of the pyknometer with water and membrane (kg)
m_{pi}	Weight of the pyknometer with IPA (kg)
m_{pw}	Weight of the pyknometer with water (kg)
N_i	Molar flux ($mol/m^2.s$)
N_L	Liquid flux ($kg/m^2.s$)
p_i	Partial pressure of component *i* (Pa)
$P_{liquid} - P_{vapor}$	Transmembrane hydrostatic pressure (Pa)

P_m	Mean pressure (Pa)
ΔP	Pressure drop (Pa)
ΔP_{bp}	Bubble point pressure (Pa)
ΔP_{TM}	Transmembrane pressure (Pa)
q_c	Heat transferred by conduction (W/m^2)
q_d	Heat which contributes to the evaporation of the distillate (W/m^2)
q_f	Heat flux in feed solution (W/m^2)
$q_{i,p}$	Total heat input of the process (W/m^2)
q_m	Heat flux in the membrane (W/m^2)
q_p	Heat flux in permeate side (W/m^2)
q_v	Vaporization heat flux of ammonia (W/m^2)
$\sum_i q_{v,i}$	Sum of latent heat to vaporize the component i (W/m^2)
Q	Flow rate (kg/s)
Q_e	Treated effluent flowrate (kg/s)
Q_w	Waste sludge removed (kg/s)
r	Radius (m)
r_{dis}	Pore size distribution (m)
r_{max}	Maximum pore size (m)
r_{mean}	Mean pore size (m)
R_c	Cake resistance (m^2/kg)
R_m	Membrane resistance (m^2/kg)
R_{obs}	Observed retentate (-)
R_{real}	Real retentate (-)
S	Salinity (ppt)
S_0	Inlet $cBOD_5$ (mg/L)
T	Temperature (K)
$T_{f,i}$	Temperature of feed inlet (K)
$T_{p,o}$	Temperature permeate outlet (K)
v	Speed/velocity (m/s)
v_a	Loading rate (m/s)
v_{mf}	Minimum fluidization velocity (m/s)

V	Total volume of the feed solution (m^3)
V_r	MBR system volumetric rate (m^3/day)
X	Concentration of volatile suspended solids (mg/L)
X_e	Concentration of volatile suspended solids, treated effluent (mg/L)
X_w	Concentration of volatile suspended solids, waste sludge (mg/L)
W_d	Weight of dry membrane (kg)
W_w	Weight of wet membrane (kg)

Greek letter

α	Degree of ionization (-)
ε	Porosity/void (-)
ε_m	Membrane surface porosity (-)
δ	Thickness (m)
ρ	Density of water (kg/m^3)
ρ_i	Density of isopropyl alcohol (kg/m^3)
ρ_m	Density of membrane (kg/m^3)
ρ_p	Density of polymer/sand particle (kg/m^3)
ρ_w	Density of water (kg/m^3)
θ	Hydraulic detention time (in MBR) (days)
θ	Liquid-solid contact angle (in membrane characterization) (^{o})
θ_c	Liquid-solid contact angle (in membrane characterization) (^{o})
θ_c	Mean cell residence time (in MBR) (days)
$\Delta\pi$	Osmotic pressure (Pa)
τ	Tortuosity of the membrane (-)
μ	Viscosity (Pa.s)
ω	Kinematics viscosity (m^2/s)
γ_L	Liquid surface tension (N/m)

LIST OF ABBREVIATION

AGMD	Air gap membrane distillation
BOD	Biological oxygen demand
$cBOD_5$	Carbonaceous biological oxygen demand
COD	Carbon oxygen demand
CO_2	Carbon dioxide
CP	Concentration polarization
CPC	Concentration polarization coefficient
DCMD	Direct contact membrane distillation
DMAc	*N,N*-dimethylacetamide
DMF	*N,N*-dimethyformamide
DMSO	Dimethylsulphoxide
DO	Dissolved oxygen
FLR	Feed loading rate
HLR	Hydraulic loading rate
HRT	Hydraulic retention time
IPA	Isopropyl alcohol
LEP_f.	Liquid entry pressure of feed solution
Lpm	Liter per minute
MBR	Membrane biological reactor
MD	Membrane distillation
MLSS	Mixed liquor suspended solid
MP	Mixing parameter
N_2	Nitrogen gas
NH_3	Unionized ammonia
NH_4^+	Ionized ammonia
NMP	*N*-methyl-2-pyrrolidone
PE	Process efficiency
PES	Polyethersulfone
PS	Polysulfone
PVDF	Polyvinylidene fluoride
rpm	Revolution per minute
RAS	Re-circulating aquaculture system
Re	Reynolds number

RT	Retention time
Sc	Schmidt number
SGMD	Sweeping gas membrane distillation
Sh	Sherwood number
SRT	Solid retention time
TA	Total ammonia
TAN	Total ammonia nitrogen
TPC	Temperature polarization coefficient
TPE	Temperature polarization effect
TE	Thermal efficiency
TN	Total nitrogen
TP	Total phosphorus
VMD	Vacuum membrane distillation

LIST OF APPENDIX

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