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Treatment of radioactive wastewater using direct contact membrane distillation

Haiyang Liu[a], Jianlong Wang[a,b,*]

[a] Laboratory of Environmental Technology, Institute of Nuclear and New Energy Technology (INET), Tsinghua University, Beijing 100084, PR China
[b] Key Laboratory of Advanced Reactor Engineering and Safety, Ministry of Education, Tsinghua University, Beijing 100084, PR China

HIGHLIGHTS

- DCMD process can separate almost all Cs^+, Sr^{2+}, Co^{2+} from liquid wastes.
- The permeate flux decreased linearly when $NaNO_3$concentration increased.
- DGM could be used to estimate the mass transfer.
- DCMD is a promising separation process for LLRW treatment.

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ABSTRACT

Direct contact membrane distillation (DCMD) was used to treat low level radioactive wastewater (LLRW). The dusty gas model (DGM) was used to analyze the mass transfer mechanism and calculate the permeate flux. The operating parameters such as feed temperature, feed velocity and feed concentration were studied. The experimental results showed that DCMD process can separate almost all Cs^+, Sr^{2+} and Co^{2+} from wastewater. The permeate flux decreased linearly when $NaNO_3$ concentration increased from 1.0 to 200 g/L. The permeate flux remained about 60% of its initial flux even when $NaNO_3$ concentration in feed solution was as high as 200 g/L. The dusty gas model can be successfully applied to estimate the mass transfer, and the experimental permeate flux values fitted well with that calculated by DGM. DCMD is a promising separation process for low level radioactive wastewater treatment.

1. Introduction

With the rapid development of nuclear energy industry in China, a large amount of low level radioactive wastewater (LLRW) will be produced from the nuclear industry, especially from the nuclear power plants [1]. The radioactive wastewater must be treated properly to protect the ecological environment and human health. Furthermore, after Fukushima accident in Japan, the treatment of radioactive wastewater has received increasing attention all over the world to support the sustainable development of nuclear energy industry. The techniques applied for radioactive wastewater treatment include traditional filtration, chemical precipitation, ion exchange, thermal evaporation, biological methods, membrane processes and so on [2–7].

In recent years, with the development of membrane technology, membrane separation technologies have been successfully applied for radioactive wastewater treatment [8–14], such as micro-filtration (MF), ultra-filtration (UF) and reverse osmosis (RO). For example, ceramic membranes [15] and metallic membranes [6] were used for treating radioactive wastewater.

Membrane distillation (MD) is a thermally-driven separation process [16]. This separation technology separates volatile molecular by a hydrophobic membrane, the driving force is pressure difference across the membrane resulting from temperature difference. According to the operating methods of the cool side of the membrane, membrane distillation systems could be classified into four categories according to their configurations [17]: direct contact membrane distillation (DCMD), vacuum membrane distillation (VMD), air gap membrane distillation (AGMD) and sweeping gas membrane distillation (SGMD). Among them DCMD is the most studied MD configuration [18].

Membrane distillation has many advantages comparing with membrane processes mentioned above, such as complete rejection of nonvolatile components, small vapor space, lower operating temperature and pressures, easy to combine with other treatment methods [19–21]. In addition, the osmotic pressure and concentration polarization have little influence on the permeate flux, or

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* Corresponding author. Tel.: +86 10 62784843; fax: +86 10 62771150.
E-mail addresses: wangjl@tsinghua.edu.cn, wangjl@mail.tsinghua.edu.cn (J. Wang).

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Notation

A	effective membrane area (m^2)
C_p	specific heat (J/kg K)
C_{pv}	specific heat of vapor (J/kg K)
D^0	ordinary diffusion coefficient (m^2/s)
d_h	hydrodynamic diameter (m)
D^k	Knudsen diffusion coefficient (m^2/s)
d_p	mean membrane pore size (μm)
h	heat transfer coefficient in boundary layer (W/m^2 K)
H	total heat transfer coefficient (W/m^2 K)
h_f	heat transfer coefficient of the heat transfer boundary layer in feed side (W/m^2 K)
h_m	heat transfer coefficient of membrane (W/m^2 K)
h_p	heat transfer coefficient of the heat transfer boundary layer in permeate side (W/m^2 K)
H_v	vapor enthalpy (J/kg)
J	permeate flux (kg/m^2 s)
$J_{i,ex}$	experimental permeate flux (kg/m^2 s)
$J_{i,m}$	experimental modeled flux (kg/m^2 s)
J^D	diffusive flux (mol/m^2 s)
J^v	viscous flux (mol/m^2 s)
k	thermal conductivity (W/m K)
K	total mass transfer coefficient (s/m)
k_B	Boltzmann constant (J/K)
k_g	thermal conductivity of gas in membrane pores (W/m K)
k_m	membrane thermal conductivity (W/m K)
k_s	thermal conductivity of polymeric membrane materials (W/m K)
k_{water}	thermal conductivity of water (W/m K)
L_{wet}	length of wetted perimeter of the flow channel (m)
M	the molecular weight (g/mol)
n	the number of date points
Nu	Nusselt number
P	total gas pressure in membrane pores (Pa)
P^0	pure liquid saturation pressure above a flat liquid surface (Pa)
P_c	pure liquid saturation pressure above a convex liquid surface (Pa)
Pr	Prandtl number
Q	total heat flux transferred across the membrane (W/m^2)
Q_f	heat flux through the boundary layer in the feed side (W/m^2)
Q_m	heat flux through membrane (W/m^2)
Q_p	heat flux through the boundary layer in the permeate side (W/m^2)
R	ideal gas constant (J/mol K)
r	membrane pore radius (μm)
Re	Reynolds number
r_h	hydraulic radius (m)
S_{cross}	cross sectional area of the flow channel (m^2)
T	absolute temperature (K)
$T_{f,m}$	temperature on the membrane surface of feed side (K)
$T_{p,m}$	temperature on the membrane surface of permeate side (K)
v	flow velocity (m/s)
x_i	liquid mole fraction of compound i
ΔP	pressure difference across the membrane (Pa)
Δt	sampling time (s)
Δm	mass increase of permeate (kg)
ΔT_m	temperature gradient across the membrane (K)
α_i	activity of the compound i
γ_L	liquid surface tension (N/m)
ε	membrane porosity
ζ_i	activity coefficient of compound i
λ_i	mean free path (μm)
μ	viscosity (Pa s)
ρ	solution density (kg/m^3)
ρ_m	liquid molar density (mol/m^3)
σ_i	collision diameter (Å)
τ	pore tortuosity

Subscript

i	refer to the compound i

membrane distillation can operate at higher salt concentrations comparing with RO. MD process has been researched and used for many different applications [21,22], for example, for ammonia removal [23], for the concentration of olive mill wastewater for polyphenols recovery [24], for petrochemical wastewater treatment and reuse [25] and for coke-plant wastewater treatment [26]. However, the research on the application of DCMD process for radioactive wastewater treatment is limited [27,28]. As MD is a thermally-driven process, so it may be suitable for radioactive wastewater treatment because there are large amount of waste heat in nuclear power plant, which could be utilized for MD process [29–32].

The objective of this study was to investigate the feasibility of the application of DCMD for radioactive wastewater treatment. The operating parameters, including feed temperature, feed velocity and feed concentration, were investigated and discussed in detail, the dusty gas model was used for simulating and analyzing the experimental results.

2. Theoretical analysis

Fig. 1 presents the mass transfer process in a typical DCMD operation, in which both the hot feed and cold permeate are in direct contact with the hydrophobic membrane. In DCMD process the mass transfer occurs due to vapor pressure difference caused by temperature gradient between the two sides of membrane surfaces. As shown in Fig. 1, the vapor pressure on the membrane surface of the feed side is higher than the permeate side due to the temperature difference, therefore the vapor transfer through the membrane pores was driven by the vapor pressure gradient. Then the vapor arrives and condensates at the vapor–liquid interface of permeate side. At last, the condensate (distillate water) transfer through the boundary layer and mixed with cold permeate. Because the membrane used in MD was hydrophobic, only the volatile substances could transfer through the membrane, and the radionuclide ions

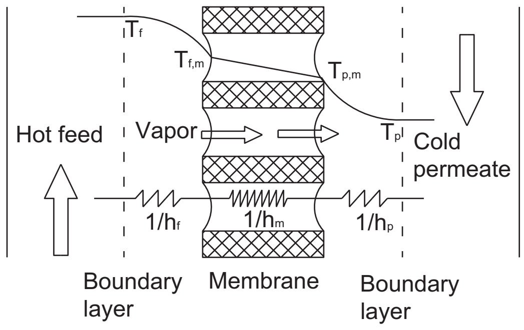

Fig. 1. Schematic diagram of heat and mass transfer in DCMD process.