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Separation of cesium ions from aqueous solution by vacuum membrane distillation process

Fei Jia [a], Jianlong Wang [a, b, *]

[a] Collaborative Innovation Center for Advanced Nuclear Energy Technology, INET, Tsinghua University, Beijing 100084, PR China
[b] Beijing Key Laboratory of Radioactive Waste Treatment, Tsinghua University, Beijing 100084, PR China

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ABSTRACT

The separation of cesium ions (Cs^+) from water solution was studied using vacuum membrane distillation (VMD) process. The removal efficiency of Cs^+ was more than 99.76% and the membrane flux maintained about 6.14 L $m^{-2} \cdot h^{-1}$ during the continuous operation. The dusty gas model was used to analyze the mass transfer of VMD process and it could simulate the mass transfer process well with average relative error of 4.84%. Mass transfer mechanism during cesium ions separation by VMD was investigated. Knudsen diffusion, i.e. collision between water vapor molecules, was proved to be the dominant mass transfer influence mechanism during VMD process. Anionic counterpart of Cs^+ (Cl^- and NO_3^-) had little influence on Cs removal, while high salt loading of the feed solution would decrease Cs removal efficiency of VMD due to salt crystallization on the membrane. Effects of operation factors (feed temperature: 303–343 K, vacuum side pressure: 5.05–90.9 kPa, feed flow velocity: Reynolds number 100–400 and feed salt concentration: NaCl 0–100 g/L) on the permeate flux were investigated. It was found that membrane flux was exponentially proportional to the feed temperature, directly proportional to the feed velocity and inversely proportional to the vacuum side pressure and feed salt concentration. VMD process has showed its promising prospect in cesium ions separation from water solution.

1. Introduction

As the rapid development of the nuclear power plants, the radioactive wastewater treatment is becoming more and more important.

Cesium is one of main fission products released from nuclear reactor. Among Cs isotopes, ^{137}Cs has a relatively long half-life about 30 years and is a beta and gamma radiation source. Such properties of ^{137}Cs make it to be a hazardous isotope to human and ecosystem (El-Kamash, 2008). Various methods have been studied for cesium removal from aqueous solution, including ion exchange, adsorption and membrane technologies (Zakrzewska-Trznadel, 2003; El-Kamash, 2008; Chen and Wang, 2010; Park et al., 2010; Khayet, 2013; Liu and Wang, 2013; Mertz et al., 2013; Chaudhury et al., 2014; Chen et al., 2015; De Haro-Del Rio et al., 2015; Ding et al., 2015; Chen et al., 2016; Kadam et al., 2016; Lu et al., 2016; Wen et al., 2016b). Among these treatment methods, membrane technology could achieve high nuclides removal efficiency. Membrane technologies are usually pressure-driven system, such as ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), which use pressure difference across the membrane as the mass transfer driving force. Among them, RO could achieve the highest salt rejection rate, while it could do little with high salinity solution.

There are two important objectives in the treatment of radioactive wastewater, i.e., achieving radionuclides rejection as high as possible and reducing radioactive waste volume as much as possible (Zakrzewska-Trznadel, 2013). It is necessary to develop new treatment technology to simultaneously achieve these two targets. Due to the unique mass transfer mechanism, membrane distillation could be a potential to meet this requirement. Temperature difference across the membrane is used as the mass transfer driving force for MD process (Lawson and Lloyd, 1997), which can be achieved by the employment of hydrophobic materials to fabricate the membrane. Due to the hydrophobicity of the membrane, only volatile substances like water vapor were allowed to pass through the membrane. Therefore, nonvolatile substances like Cs^+ ions would be rejected in the feed solution (Lawson and Lloyd, 1997). Theoretically, this method could achieve complete

* Corresponding author. Energy Science Building, Tsinghua University, Beijing 100084, PR China.
E-mail address: wangjl@tsinghua.edu.cn (J. Wang).

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Nomenclatures

A	Total effective membrane area (m^2)
AGMD	Air gap membrane distillation
ARE	Average relative error
σ_i	Molecular collision diameter (m)
DCMD	Direct contact membrane distillation
d	Hydraulic diameter (m)
DGM	Dusty gas model
d_p	Membrane average pore diameter (m)
Δm	Mass increase of the permeate collected in the condense (g)
ΔP	pressure difference across the membrane surface (Pa)
Δt	Sampling time (h)
δ	Membrane thickness (m)
ε	Membrane porosity
J	Permeate flux ($L \cdot m^{-2} \cdot h^{-1}$)
J_{exp}	Actual permeate flux ($L \cdot m^{-2} \cdot h^{-1}$)
J_{DGM}	Simulated permeate flux ($L \cdot m^{-2} \cdot h^{-1}$)
k_B	Boltzmann constant ($1.380 \cdot 10^{-23}$ J/K)
Kn	Knudsen number
λ_i	Gas molecular mean free path (m)
μ	Viscosity of feed flow ($m \cdot s \cdot g^{-1}$)
MD	Membrane distillation
NF	Nanofiltration
P	Mean pressure in membrane pores (Pa)
PP	Polypropylene
P^{sat}	Pure water vapor saturated pressure (Pa)
ρ	Density of permeate solution in the present study ($kg \cdot L^{-1}$)
r	Average pore radius (m)
R	Universal gas constant (8.314 J $mol^{-1} \cdot K^{-1}$)
RO	Reverse osmosis
SGMD	Sweeping gas membrane distillation
T	Mean temperature in membrane pores (K)
T_f	Temperature of feed side (K)
$T_{f,m}$	Temperature of membrane surface in feed side (K)
T_p	Temperature of permeate side (K)
$T_{p,m}$	Temperature of membrane surface in permeate side (K)
τ	Pore tortuosity of membrane
UF	Ultrafiltration
v	Flow velocity ($m \cdot s^{-1}$)
VMD	Vacuum membrane distillation

salt rejection and had ability to process high salinity solution which is difficult for RO method. These merits made MD methods possible to meet the two main objectives in radioactive wastewater treatment.

MD methods could be classified into four types from configuration, i.e., direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD) and vacuum membrane distillation (VMD) (Lawson and Lloyd, 1997). The first one was the original type while suffered from membrane wetting phenomenon obviously. During the operation, due to the existence of pollutants, the property of the membrane could be changed which would attenuate the hydrophobicity of the membrane. If membrane wetting occurred, the solute would pass through the membrane and would pollute the permeate solution. To solve this issue, the other three methods were developed to isolate the membrane from contacting with the permeate solution directly. Compared with AGMD and SGMD, VMD method created a vacuum environment in the permeate side. Under the vacuum circumstance, the permeate vapor would be stripped more quickly which would increase the mass transfer driving force. Three types of VMD membrane column have been developed: flat sheet, spiral wound and hollow fiber membrane (El-Bourawi et al., 2006). Compared with other two types, hollow fiber membrane had the highest surface density area, because hollow fiber membrane requires support materials and this could contribute to increase membrane packing density in the membrane module. Hence hollow fiber membrane module could achieve compact MD installation and higher flux.

The radioactive wastewater treatment by MD method has been investigated previously (Khayet, 2013; Liu and Wang, 2013; Wen et al., 2016a). We also found DCMD could achieve nearly complete nuclides rejection (Liu and Wang, 2013). Although DCMD has been used for the treatment of radioactive wastewater, only a few researches were performed for Cs removal by VMD. For example, Wen et al. (2016a) has investigated Cs removal efficiency by VMD and influence of operation parameters to the membrane flux. It was found that VMD could achieve high Cs removal efficiency. However, the research about mass transfer mechanism during Cs removal by VMD is still limited. Figuring out mass transfer mechanism during Cs removal by VMD can contribute to get a deeper insight of how different parameters influence the mass transfer mechanism during Cs removal by VMD. This can contribute to optimize VMD performance.

Therefore, present study aimed to investigate the mass transfer mechanism during Cs removal by VMD. Mathematical model and experimental data were combined together to analyze mass transfer during the process and estimate the membrane flux. Effects of anionic counterpart of Cs^+ (Cl^- and NO_3^-) and feed salt concentration on Cs removal efficiency were investigated. Effects of operation factors, including feed temperature, vacuum side permeate pressure, feed flow velocity and feed salt concentration, on permeate flux were studied.

2. Materials and methods

2.1. Chemicals and experimental set-up

Due to similar chemical property, non-radioactive isotope of CsCl and $CsNO_3$ (Sinopharm, Beijing, China) was used as surrogate for radioactive isotope of CsCl and $CsNO_3$ in present study. NaCl was purchased from Sinopharm (Beijing, China). Except for anionic counterpart of Cs^+ (Cl^- and NO_3^-) and feed salt concentration study, CsCl solution containing 10 mg/L of Cs ions was used as feed solution in other experiments in the present study. All chemicals used in the present study were of analytical grade.

Fig. 1 shows scheme of experimental set-up. The experimental system was composed of three parts: feed solution circulation system, membrane module and vacuum condensation system. The feed solution circulation system consisted of effluent tank, effluent heating system, peristaltic pump, thermometers and flowmeter. The vacuum condensation system was made up of condensation system with chilled water, vacuum pump, vacuum buffer bottle, precision balance and thermometer. The present research employed commercial polypropylene hollow fiber MD membrane module (Wochi, WHPP96-21, China). Table 1 lists several important parameters of the membrane module.

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