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Removal of strontium ions from simulated radioactive wastewater by vacuum membrane distillation

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ABSTRACT

The removal of Sr^{2+} ions from simulated radioactive wastewater by vacuum membrane distillation (VMD) with hollow fiber membrane module was investigated. The removal efficiency of Sr^{2+} could maintain over 99.60%, and the membrane flux could maintain at 6.71 $L{\cdot}m^{-2}{\cdot}h^{-1}$ when Sr^{2+} in the feed solution was about 10 mg/L. The effect of operating parameters on the membrane flux was examined, including feed temperature (30–70 °C), permeate side vacuum (0.10–0.98 atm) and feed flow velocity (10.5–41.8 L/h). Pressure buildup effect was observed during the process, which could interfere with the membrane flux. Pressure buildup effect was more serious when permeate side vacuum degree was over 0.9 atm. Considering energy consumption efficiency, the permeate side vacuum degree should be maintained at the turning point (0.9 atm) of the membrane flux. Dusty gas model could simulate the mass transfer of VMD process well with ARE (average relative error) of 5.31%. VMD is potential for the removal of Sr^{2+} ions from aqueous solution.

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1. Introduction

After the tragedy of Fukushima Daiichi, the treatment of radioactive wastewater has received increasing attention. As one of main fission products of nuclear reactor, strontium has been regarded as a hazardous radionuclide with half-life of about 30 years (El-Kamash, 2008). A series of methods have been investigated to remove Sr^{2+} from aqueous solution, including adsorption (Chen and Wang, 2010, 2012; Park et al., 2010; Chen et al., 2014; Chen et al., 2016), ion exchange (El-Kamash, 2008) and membrane technology (Liu and Wang, 2013; Ding et al., 2015, 2016), etc. Compared with other methods, membrane technology could achieve both high salt removal efficiency and high concentration factor (Lawson and Lloyd, 1997). Among all membrane technologies, membrane distillation (MD) has some features in terms of its unique purification mechanism. Unlike series of pressure-driven membrane methods, MD employs the temperature difference across the membrane as the mass transfer driving force (Lawson and Lloyd, 1997). The membrane of MD system is made from hydrophobic materials which only allow volatile matters to pass through the membrane during the separation process (Lawson and Lloyd, 1997). Non-volatile matters like Sr^{2+} will be remained in the original wastewater. In previous research, we confirmed that MD could achieve high nuclide removal efficiency (Liu and Wang, 2013). Compared with pressure-driving membrane method, MD has several advantages including milder and safer operation condition, capability in processing high salinity wastewater, potential to use low quality heat sources like waste heat in nuclear power plant as mass transfer driving force (Lawson and Lloyd, 1997; Zuo et al., 2016).

Classified by the membrane configuration, MD process could be categorized into four types: direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD) and vacuum membrane distillation (VMD). DCMD is the original MD type with the simplest structure. However, as the permeate solution contacts with the membrane directly, the permeate will be polluted directly if there is a damage on the membrane or membrane wetting. The rest of three MD types decrease the risk of permeate pollution by separate membrane and permeate. For VMD method, it creates a vacuum atmosphere in the shell side of the membrane column to strip the permeate vapor from the shell. Besides decreasing the influence of membrane wetting, the application of vacuum also increase the mass transfer driving force of the system, which could increase the membrane flux of VMD process. There are three types of membrane column for VMD method: flat sheet membrane, spiral wound membrane and hollow fiber membrane (El-Bourawi et al., 2006).

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Nomenclature

A	total effective membrane area (m^2)
AGMD	air gap membrane distillation
DCMD	direct contact membrane distillation
DGM	dusty gas model
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 ($L{\cdot}m^{-2}{\cdot}h^{-1}$)
MD	membrane distillation
P	mean pressure in membrane pores
P^{sat}	pure water vapor saturated pressure
PP	polypropylene
ρ	density of permeate solution in the present study ($kg{\cdot}L^{-1}$)
r	average pore radius (m)
R	universal gas constant ($8.314\ J{\cdot}mol^{-1}{\cdot}K^{-1}$)
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
VMD	vacuum membrane distillation

Among these three types, hollow fiber type could achieve the highest surface density area. This results in the more compact system of the membrane module, which increase the real application potential of hollow fiber VMD method.

There are a series of researches on the treatment of radioactive wastewater with MD method (Zakrzewska-Trznadel et al., 1999; Khayet, 2013; Liu and Wang, 2013, 2016; Wen et al., 2016). However, there were few researches focusing on Sr removal by VMD method. As discussed above, VMD has a potential to be an effective method to remove Sr^{2+} ions from solution.

The objective of this study was to investigate the treatment of Sr^{2+}-containing radioactive wastewater by hollow fiber VMD method. The effect of operating parameters on the performance of VMD process, including feed temperature, shell side vacuum degree and feed flow rate was studied. The mathematical simulation of VMD mass transfer process was discussed to predict the membrane flux.

2. Materials and methods

2.1. Chemicals and membrane modules

$SrCl_2$ (Sinopharm Chemical, China; analytical pure) solution of about 10 mg/L was employed as feed in this experiment. Hollow fiber membrane made from polypropylene (PP) (Wochi, WHPP96-21, China) was applied in the VMD process. Property of the membrane is listed in detail in Table 1.

2.2. Experimental set-up

The VMD system was made up of feed container, feed heating system, MD module, condenser system, vacuum pump, peristaltic pumps, precision balance, flowmeter and thermometers (Fig. 1).

Table 1
Performance of the membrane module.

Parameters	Units	Value
Hollow fiber number	–	140
Effective membrane length	mm	140
Total effective inner area	m^2	0.062
Packing density	–	0.35
Porosity	–	60%
Mean pore diameter	μm	0.18
Inner radius of the fiber	mm	0.50
Outer radius of the fiber	mm	1.36

During VMD process, the solution would be heated to the preset temperature at first and then be pumped into the membrane module. At the shell side of the module, the vacuum pump would create a vacuum atmosphere and the permeate vapor would be stripped away from the shell and be condensed into liquid in the condenser which was filled with chilled water. Then the rest of solution in the lumen side would be recycled into the container to supplement the heat loss.

2.3. Analytical methods

Sr ions concentration was analyzed by flame AAS (Hitachi, ZA3000, Japan). Membrane flux of MD was obtained by detecting water mass collected in the condenser with precision balance.

The membrane flux was deduced as follows:

$$J = \frac{\Delta m}{\rho A \Delta t} \tag{1}$$

where J is the membrane flux ($L{\cdot}m^{-2}{\cdot}h^{-1}$); Δm is the water mass collected in the condense (g); ρ is water in the present study ($g{\cdot}m^{-3}$); A is the membrane area (m^2); Δt is the sampling interval (h).

3. Results and discussion

3.1. Effect of feed temperature

In this section, feed temperature was varied from 30 to 70 °C, with 0.98 atm of permeate vacuum degree and 41.8 L/h of feed flow velocity. Fig. 2 shows the variation of permeate at different feed temperatures. It could be seen that at all temperatures, the permeate mass increased linearly with time, indicating that the VMD system reached the steady state for producing water. Therefore the sampling time was determined to be 10 min.

Fig. 3 shows the variation of the membrane flux with feed temperature. It could be seen that the membrane flux increased exponentially ($R^2 = 0.9762$) with the feed temperature, which could be elucidated by the variation of saturated vapor pressure in the lumen side. During the VMD process, the pressure difference across the membrane played as the mass transfer driving force. The saturated vapor pressure of lumen side P^{sat} (Pa) at temperature of T(K) could be calculated by Antoine Equation:

$$\log P^{sat}(T) = \exp\left(23.1964 + \frac{3816.44}{46.13 - T}\right) \tag{2}$$

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