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Research on High Flux Direct Contact Membrane Distillation Technology

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Abstract: The effects of membrane materials, membrane module structure and membrane distillation operation parameters on the performance of direct contact membrane distillation were investigated by using strontium chloride solution with Sr^{2+} concentration of 1g/L as test solution. The results showed that the flux of PTFE is the largest, PVDF is the second, PP is the smallest, and the rejection rate of PTFE decreases rapidly with the increase of running time, which indicated that PTFE is easy to be fouled, while PVDF is a better membrane material; When the packaging fraction of membrane module increases, the membrane flux decreases, but the water yield per unit time increases. Considering the economy and treatment efficiency, it is recommended to select the membrane module with packaging fraction of 11.42%; the increase of feed flow rate and temperature is beneficial to the increase of flux in DCMD process. The rejection rate is almost unaffected by the membrane module structure and operating conditions, and remains above 99.9%. This study is helpful to provide research direction for improving membrane distillation flux, and provide theoretical reference for membrane distillation technology to treat practical radioactive wastewater.

1. Introduction

Membrane distillation technology is a fast-developing membrane separation technology in recent years, which has a promising future in radionuclide wastewater treatment. It has the advantages of less secondary waste, simple operation, wide range of application, good purification effect, and mild operating conditions. Compared with ultrafiltration, microfiltration, reverse osmosis and other wastewater membrane methods, one of the most prominent merits is that membrane distillation can deeply treat high salinity wastewater. There are already a host of researches in the fields of the treatment of oily wastewater, radioactive wastewater, printing and dyeing wastewater.

According to the condensing mode of the cold side of membrane, membrane distillation is divided into direct contact membrane distillation (DCMD), vacuum membrane distillation (VMD), air gap membrane distillation (AGMD) and air swept membrane distillation (SGMD). Among them, the DCMD device is the most portable, with mild operating conditions and no risk of radioactive elements diffusing into the environment in the form of aerosols. Therefore, no additional protective equipment is required during operation [1]. It is currently the most widely used membrane distillation method.

At present, the distillation devices are still at laboratory stage at home and abroad. The main reason is the flux of membrane distillation is too little to reach the standard of engineering practice. To solve the problem, current researches focus on optimize membrane material, module and distillation devices [2]. In this research, the direct contact membrane distillation device was selected to treat simulated radioactive waste containing Sr2+, and the effects of membrane materials, membrane module structure, membrane distillation operating conditions and other factors on the membrane distillation property were investigated. Under the premise of economy and efficiency, the high flux will be obtained as much as possible, to provide a reference for the practical application of the direct contact in the treatment of radionuclide waste.

2. Experiments and Results

2.1. Materials and Methods

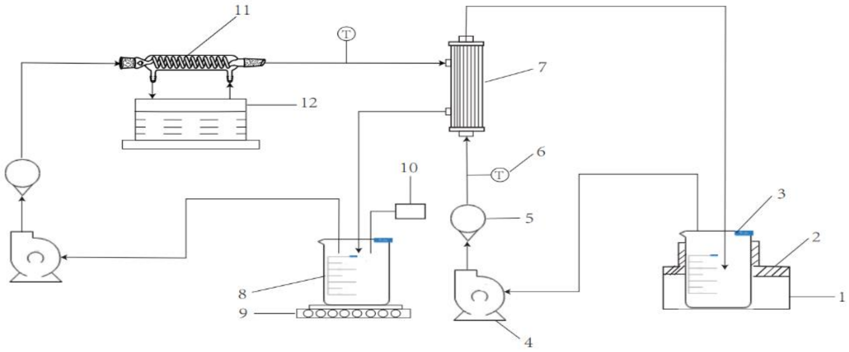
2.1.1. Reagents and instruments

$SrCl_2$ $6H_2O$,AR,99%,Shanghai MACKLIN Biochemical Technology Co., Ltd.; deionized water, conductivity less than 0.7μS/cm, self-made; PP hollow fiber membrane, inner diameter 0.4 mm, PTFE hollow fiber membrane, inner diameter 0.65mm, provided by Tianjin Institute of Seawater Desalination and Comprehensive Utilization, Ministry of Natural Resources; PVDF hollow fiber membrane, inner diameter 0.8 mm, provided by Tiangong University.

DDSJ-308F conductivity meter, Shanghai Yidian Scientific Instrument Co., Ltd; PX2202ZH/E electronic scale, precision 0.01 g, Ohaus Instruments Co., Ltd.; ME55/02 delicate scale, precison10-5 g, Mettler Toledo Instruments Co., Ltd.

2.1.2. Direct contact membrane distillation experiment device

The self-constructed direct contact membrane distillation device is mainly composed by the heat side, the cold side and the hollow fiber membrane module. The heat side includes thermostatic water bath, magnetic circulating pump, rotameter and thermometer; the cold side includes water collection beaker, conductivity meter, electronic scales, graham condenser and low temperature cooling circulating pump.



1—thermostatic water bath, 2—thermal cotton sheet, 3—beaker with feed solution, 4—magnetic circulating pump, 5—rotameter, 6—thermometer, 7—hollow fiber membrane module, 8—water collection beaker with deionized water, 9—electronic scales, 10—conductivity meter, 11—graham condenser, 12—low temperature cooling circulating pump

Figure 1: The direct contact membrane distillation device

The process of direct contact membrane distillation is as Figure 1. Before operation, add the prepared $SrCl_2$ solution into the beaker and heat it in the thermostatic water bath to keep the temperature of the feed liquid at about 60 ℃. The hot feed liquid is pumped into the tube side (inner surface of the membrane filament) of the membrane module by the magnetic pump, and the cooling water is pumped into the shell side (outer surface of the membrane filament) of the membrane module. Due to the temperature gap on both sides of the membrane, the feed liquid evaporates in the membrane filament to form vapor, which is driven by the pressure difference on both sides of the membrane to reach the outer surface of the membrane filament through the membrane hole, and is condensed by the circulating cooling water and taken out of the membrane module. It is collected in the collection beaker containing deionized water. The mass change is recorded by the electronic scale in real time, and the water quality is recorded every 1h and its conductivity is measured.

2.1.3. Major investigation index

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Water production rate of the distillation device is measured by membrane flux (J), It refers to the quality of water production per unit time and membrane area, as formula (1)[3]:

$$J=W/(A\cdot t) \tag{1}$$

In the formula: J—membrane distillation flux, kg/ (m2 ħ); W——the quality of water production in certain time, kg; t—W (kg)the time of water collection, h; A——membrane effective area, m2。

There are a lot of factors that influence the membrane flux, like the length of membrane filament, pore radius, temperature gap, feed rate and so on.

The efficiency of membrane distillation device is evaluated by the reject rate(R) as formula (2)

$$R=(C_F-C_P)/C_F \times 100\% \tag{2}$$

In the formula: R—the reject rate of membrane distillation; C_F——mass concentration of strontium in feed liquid, g/L; C_P——strontium mass concentration in the water collection beaker, g/L.

2.2. Experiment Results and Discussion

2.2.1. The influence of membrane material on membrane distillation property

The currently used membrane distillation material includes PP, PTFE, PVDF, which are all good in hydrophobicity, thermal stability and chemical stability, can meet the requirements of membrane distillation experiment. However, the pore radius, mechanical property and hydrophobicity of each material varies, which may lead to differences in membrane flux, the reject rate for pollutants and service life. Therefore, the membrane material should be well-selected.

Produce membrane module using three types of filaments. It is required that the color and quality of the filaments shall be uniform, the surface shall be flat and free of scratches and holes. The effective length of membrane material is 22cm, with an equal packing density(total effective area) at 176cm2(the amount of corresponding membrane filament PVDF50 pieces, PP89 pieces, PTFE67pieces).To conduct direct contact membrane distillation experiment and compare the flux and reject rate of each group respectively. The experiment results are shown in Figure 2 and Figure 3.

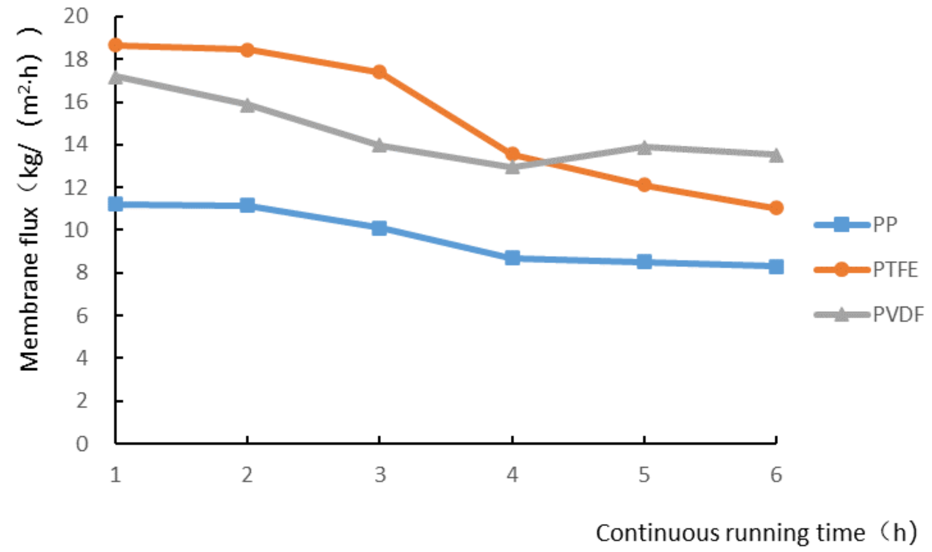

Figure 2: The flux of three types of membrane material varies with time

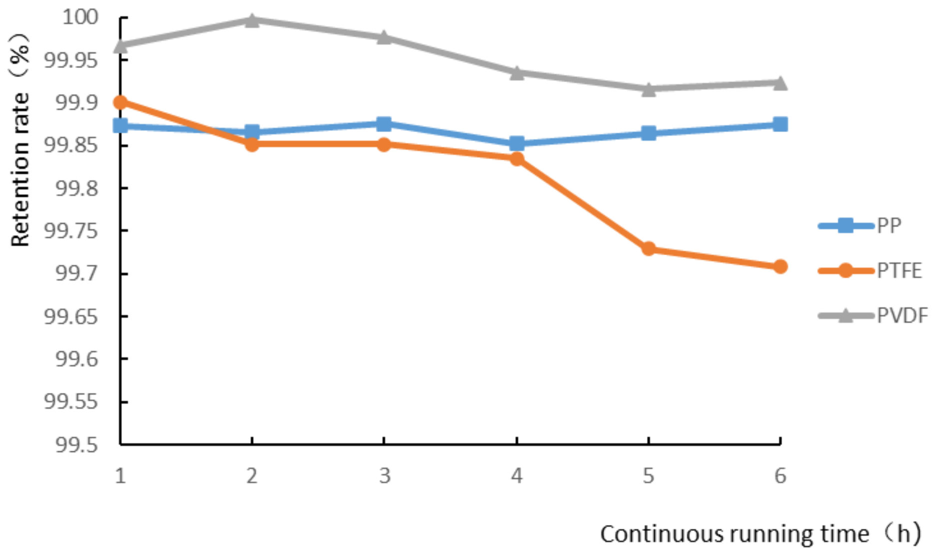

Figure 3: The reject rate of three types of membrane material varies with time

Figure 2 indicated that, at the start of the experiment, the flux of PTFE is the maximum of the three, followed by PVDF and PP; as the distillation continues, the flux of PTFE continues to decrease from 18.652kg/(m2 ћ) to 11.029kg/(m2 ћ). Comparingly, the flux of PVDF decreased slightly, and can maintain a high-level of flux.

Figure 3 indicated that, at the beginning of the experiment, the reject rates of strontium in the feed solution of PP, PTFE and PVDF were similar, 99.873%, 99.901% and 99.967% respectively. With the increase of operation time, the salt content in the feed solution increases, and the reject rate of PTFE decreases significantly, while the other two materials show no obvious changes. It implies that PTFE is easily soaked and polluted because of its low water permeability pressure, which cannot meet the requirements of long-term continuous operation.

2.2.2. The influence of packing density on membrane distillation property

Adopted PVDF membrane filament at the length of 22cm, with the feed temperature at 60℃, feed rate at 0.66m/s, temperature gap between the heat and the cold side no less than 30℃, adjusted the packing density at 5.71%, 11.42%, 17.14% and 22.85%(corresponding membrane filament 25, 50, 75, 100). To test the influence of packing density on direct contact membrane distillation property. The experiment results are shown in Figure 4 and Chart 1.

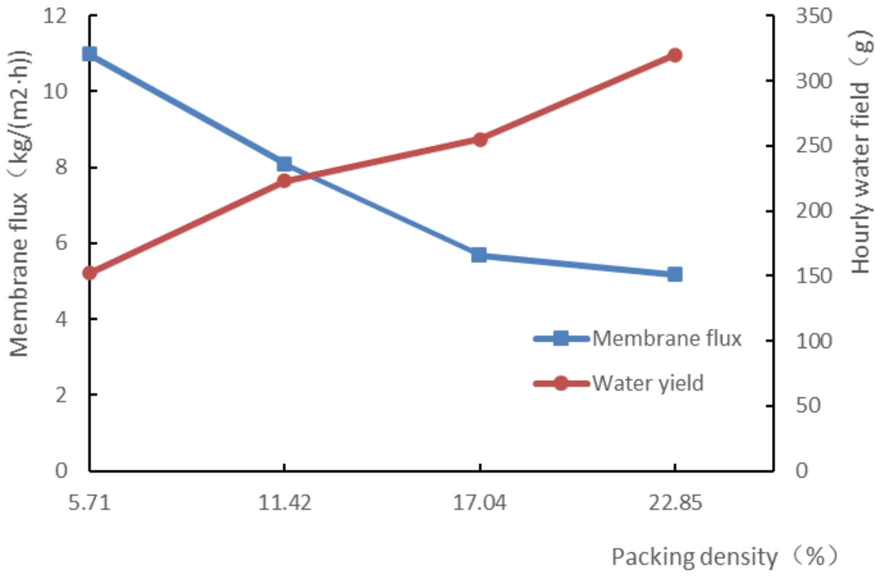

Figure 4: The influence of packing density on direct contact membrane distillation property

Table 1: The influence of different packing density on direct contact membrane distillation property

Membrane Filament Packing Density (%)	Number of Corresponding Membrane Filament	Membrane Flux (kg/(m² h))	Water Yield per hour(g)	Reject Rate (%)
5.71	25	10.99	151.77	99.95
11.42	50	8.17	225.68	99.96
17.14	75	6.02	251.20	99.96
22.85	100	5.64	320.15	99.95

Figure 4 and Table 1 have shown that as the increase of packing density, the membrane distillation flux decreases, while the water yield per hour increases. The reason that caused this phenomenon is that the lower the packing density, the more sufficient will the feed liquid along the radial direction of the shell mix. It is conducive to the consistency of the temperature of the fluid at different positions on the channel, and to a certain extent, the temperature of the local hot and cold fluid will be avoided to be too close, weakening the channeling effect.

The reject rates of membrane modules with different packaging densities for strontium-containing solution are basically the same, ranging from 99.8% to 99.9%. Takeing economy and efficiency into account, membrane module with a packing density at 11.42% is optimal.

2.2.3. The influence of feed rate on membrane distillation property

50 pieces of PVDF hollow fiber membrane module with the length of 22cm were adopted, with the feed temperature at 60℃ and the temperature gap between the heat side and the cold side no less than 30℃, selects feed rate at 0.33, 0.66, 0.99, 1.32m/s(corresponding water flux 30, 60, 90, 120L/h)to conduct the experiment. The experiment results are shown in Figure 5.

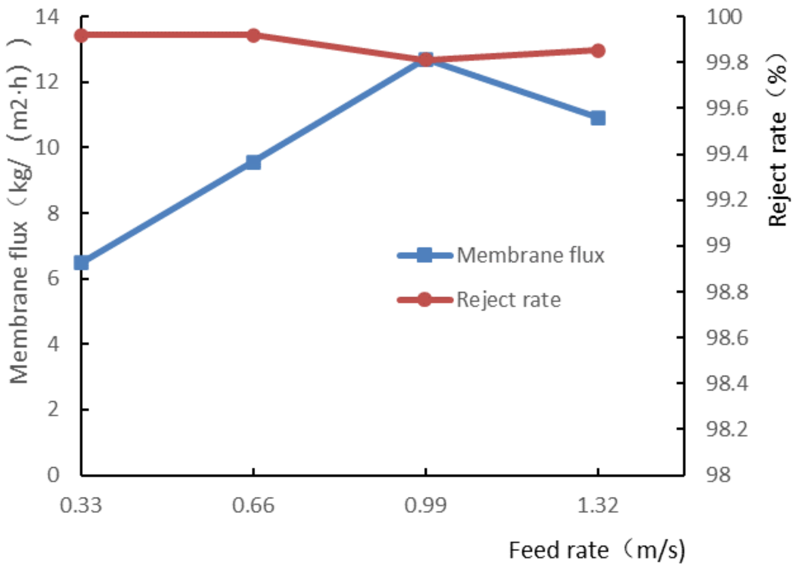

Figure 5: The influence of feed rate on direct contact membrane distillation property

Figure 5 have shown that, when the feed rate is 0.33~0.99m/s, the flux of membrane distillation will increase as feed rate increases. This is because the increase of feed rate reduces the thickness of boundary layer, weakens the polarization phenomenon, which is conducive to the mass transfer in membrane distillation. However, when the boundary layer is thin to a certain extent, it will no longer be affected by the feed rate. With the increase of feed rate continues, the membrane flux will no longer increase [4]. For a specific membrane module, the most economical and appropriate feed rate should be selected, which is closely related to the inner diameter of the membrane filament.

Under different feed rate, the reject rate of membrane module for the pollutants in the material liquid changed slightly. They can maintain a high level of above99.9%.

2.2.4. The influence of feed temperature on membrane distillation property

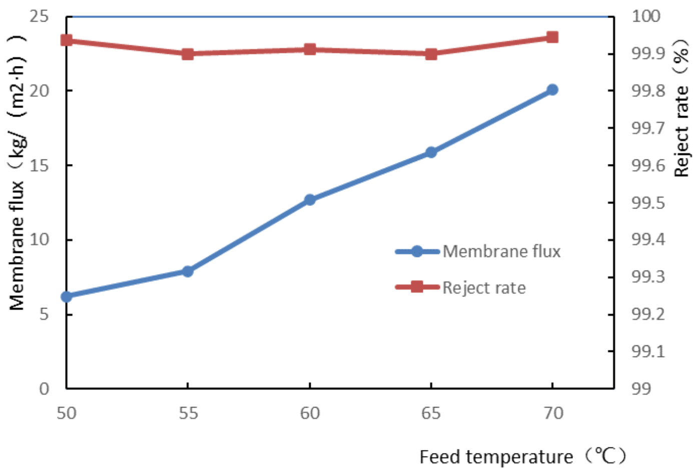

Figure 6: The influence of feed temperature on direct contact membrane distillation property

50 pieces of PVDF hollow fiber membrane module with the length of 22cm were adopted, with the feed rate at 0.99m/s(corresponding flux 90L/h), the temperature gap between the heat side and the cold side greater than or equal to 30℃. Generally speaking, the temperature of the heat side ranges

from 50 to 70℃. But in the experiment, it implied that the heat side of a few set of membrane distillation device in the lab stabilizes around 70℃ during circulation. Therefore, this research selects the feed temperature of 50℃, 55℃, 60℃, 65℃ and 70℃, monitoring the temperature of the heat side in real time to make sure the actual temperature of the heat side fluctuates within 2℃. The experiment results are shown in Figure 6.

Figure 6 showed that the membrane flux increases with the rise of feed temperature, the higher the temperature is, the faster the flux increases. The reject rate is not influenced by the temperature, and can be stable above 99.9%.

3. Conclusions

This research carried out experiments in following three aspects: hollow fiber membrane material, hollow fiber membrane module structure, Membrane Distillation operation parameter. The results are as follows:

(1) Compared the performance of three commonly used materials for the treatment of simulated radioactive wastewater, PTFE membrane flux is the highest, but with the increase of running time, the interception rate decreases quickly, easy to be polluted, cannot meet the requirements of continuous operation, the use of PVDF hollow fiber membrane material is recommended for follow-up experimental research;

(2)When the packing density of membrane module increases, the water yield per unit of time increases accordingly, while the membrane flux decreases. The reject rate was unacted on membrane module structure. Considering the efficiency and economy of membrane distillation, membrane module with a packing density of 11.42% was recommended.

(3)From the perspective of membrane distillation operation parameter, membrane flux increases as feed rate increases, until it reaches a certain level; membrane flux also increases when the feed temperature rises. The reject rate is irrelevant to operation parameters. Considering the economy and operability, it is better with feed rate at 0.99m/s, and control the feed temperature at 70℃.

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