



Effect of microwave irradiation on typical inorganic salts crystallization in membrane distillation process



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ABSTRACT

This study is a continuation of our previous work on microwave-assisted membrane distillation. The crystal morphology of sodium chloride and calcium carbonate in the process of membrane distillation and microwave membrane distillation was investigated in this work. The effect of salt concentration on the ability of absorbing microwave of solution and the mass transfer of microwave membrane distillation were also analyzed. The results show that the ionic conduction is the main between two factors (ionic conduction and dipole rotation) which have effect on absorbing microwave. When the mass concentration of sodium chloride solution is up to 5%, the microwave energy absorbed by solution is almost five times as much as pure water. Moreover, the increase in salt concentration has no significant extra influence on the mass transfer of microwave membrane distillation. The crystallization experiments confirm that microwave irradiation do not affect membrane fouling or worsen it. The different effect of microwave irradiation on crystallization of two typical inorganic salts is observed in this study. For sodium chloride, the microwave heating leads to the decline of the total particle number, while the total deposition keeps constant and the size distribution of crystal particles becomes more uniform, compared with conventional heating. As for calcium carbonate, microwave irradiation not only increases the total deposition and total particle number, but also induces the crystals growth. Especially, it is found that microwave irradiation induces the crystals to grow toward aragonite. The ratios of the aragonite number to the total particle number increase from 28.0% and 56.5% to 81.1% and 85.8% at the two higher feed mass concentrations, respectively.

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1. Introduction

Membrane distillation is considered to be a promising technology for desalination of high concentration salty water. However, there are still some obstacles, such as inadequate permeate flux for amplified reactor and membrane fouling, which impede the large-scale application of membrane distillation [1,2]. For solving these problems, many efforts have been done on setting turbulent-inducing components in flow channel of membrane module [3–5] and coupling ultrasonic technique with membrane distillation [6]. Specially, our research team introduced microwave technique into membrane distillation process for investigating the effect of microwave irradiation on the mass transfer of vacuum membrane distillation (VMD) [7]. It was found that the mass transfer was improved significantly under microwave irradiation. But the effect of microwave irradiation on crystallization was still confusing.

It is generally believed that microwave irradiation produces two effects: thermal effect and non-thermal effect (special effect)

[8]. The special effect is an interesting and often confusing phenomenon in many researches, such as microwave-assisted synthesis or preparation of organic compounds [9–14], microwave-enhanced molecules diffusion in polymeric materials [15,16] and microwave-assisted extraction or removal [17–21]. It is considered by many scholars that the light quantum (the quantum of electromagnetic radiation) of microwave has some special effects on reducing the Gibbs free energy of activation of reactions. The effects may be shown in two respects: (1) microwave energy is absorbed and stored in the internal molecule; (2) the arrangement of molecules is changed [8]. Moreover, in a liquid reaction system, the polar molecules irradiated by microwave change directions quickly to form a “micro-agitation” effect, which can also be considered as a microwave special effect.

For ionic solutions placed in a microwave field, the ions will migrate toward corresponding electric field direction and change migration direction continuously with the alternation of the electric field. Based on this principle, it has been confirmed that the rate of ion exchange can be enhanced by using microwave as heat source for ion exchange reactions [22]. This mode of ion migration and changing direction has a special effect on crystallization process.

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Microwave irradiation has been applied directly in salt crystallization from ionic solutions. Rodriguez-Clemente and Gomez-Morales [23] found in precipitation of CaCO_3 from homogeneous solutions that a higher rate of nucleation and greater particle size uniformity can be obtained under microwave heating. Yang et al. [24] did research on CaSO_4 crystallization process, the results of which showed that there was significant difference on the crystal morphology between two operation conditions of microwave heating and conventional heating.

Inorganic salt crystallization is a conventional phenomenon in desalination by membrane distillation. It was found occasionally in our research on desalination by microwave vacuum membrane distillation that microwave irradiation might aggravate membrane fouling [7]. So, in this work, the effect of microwave irradiation on inorganic salt crystallization in membrane distillation is studied systematically. This work mainly focuses on the behavior of two typical inorganic salts in the process of microwave membrane distillation (MWMD).

2. Experimental

2.1. Materials and membrane module

One PVDF flat membrane (FluoroTrans[®]) with a nominal pore size of 0.2 μm and a thickness of 127 μm , which was purchased from Pall Co., was chosen to fabricate membrane modules. The flat membrane was clipped by two PTFE plates with parallel grooves. And these components were fixed using PTFE screws through the edges of the two plates. The reason for using the PTFE plates and screws is that the microwave dielectric loss of the PTFE materials is very small. Therefore, the membrane module can be placed in microwave cavity for a long time. The effective membrane area of the module is about 0.0029 m^2 . The characteristics of the membrane material and the module are presented in Table 1.

2.2. MWMD setup

The MWMD experimental setup is schematically shown in Fig. 1. The salt solution, which was stirred continually by a magnetic stirrer, was heated to one certain temperature by a constant temperature water bath, and then circulated through the hot side of the module using a cycle water pump. The steam flowed through membrane pores into the cold side of the module and was captured by coolant, the pure water. Then the condensate was carried into coolant bottle. The overflow entered into measuring cylinder and was measured by electronic balance. In the process of circulation, the membrane module was radiated continually by microwave (2.45 GHz, 700 W), which was provided by household microwave oven. The temperature and velocity of the solution in the module inlet and outlet were monitored by temperature meter and rotameter, respectively. The conductivity meter was used to monitor the conductivity of the liquid in the coolant bottle.

Table 1
The characteristics of the membrane material and the module.

Membrane and module	Properties
Membrane material	PVDF
Nominal pore size (μm)	0.2
Thickness (μm)	127
Porosity (%)	75
Effective membrane area (m^2)	0.0029

Sodium chloride and calcium carbonate as the typical inorganic salts in water were chosen as research objects. And a series of experiments with MD and MWMD were conducted to investigate the effect of microwave irradiation on crystallization process of sodium chloride and calcium carbonate.

2.3. Crystal morphology analysis

For investigating the morphology of the crystals deposited on the membrane surface, scanning electron microscopy (SEM) and two image analysis software were adopted. The precise information of the crystal morphology could be acquired through certain treatment to the SEM images by software.

First, the images of the crystals on membrane surface were obtained by a scanning electron microscopy (HITACHI, S-3000N).

Then, two image analysis softwares including Nano Mesurer (1.2) and Imagej (1.44i) were adopted to analyze the crystal morphology. For the image which has a relatively obvious boundary in color between crystals and the background of the membrane surface, Imagej (1.44i) can automatically recognize and extract the crystal objects through adjusting the gray scale of the image. After special treatment, this software can provide much information about the crystal particles in the image, such as particle size and count, total area and area fraction, etc. If the image has not a relatively significant boundary in color between crystals and background, the other software named Nano Mesurer (1.2) will be introduced to the treatment of the image. But the precondition is that the crystals can be identified by the naked eyes. Each recognizable crystal is numbered manually, then the crystal size is measured by software.

3. Results and discussions

3.1. Effect of sodium chloride concentration on the MWMD process

Two phenomena, dipole rotation and ionic conduction, will occur in inorganic salt solution, if the solution is exposed to microwave irradiation. Therefore, the ion concentration may have a great impact on absorbing microwave of the solution and the mass transfer of the MWMD process. For clarifying this point, sodium chloride solution was chosen to implement a series of experiments at eight concentration levels with conditions of feed velocity of 1.1 m/s and feed average temperature of 54 °C. In addition, the inlet temperature and the velocity of the coolant (pure water) were set to 10 °C and 0.5 m/s, respectively.

First, the effect of sodium chloride concentration on the ability of absorbing microwave of the solution was investigated. On the condition that the average temperature was set to a constant value of 54 °C and no coolant fluid was added in the cold side of the module, the inlet temperatures of the solution under conventional heating and microwave heating were both measured. Normally, when the feed solution flows through the module, natural heat loss will appear. Microwave irradiation can effectively compensate the heat loss. The temperature increment caused by absorbing microwave can be obtained from Eq. (1)

$$\Delta T = (T_{wo} - T_{wi}) - (T_{co} - T_{ci}) \quad (1)$$

where ΔT is the temperature increment caused by absorbing microwave. T_{wo} and T_{wi} are the outlet and inlet temperatures of the feed solution in module channel under microwave heating, respectively. For the same reason, T_{co} and T_{ci} are the outlet and inlet temperatures of the feed solution under conventional heating, respectively.

The effect of sodium chloride concentration on absorbing microwave of the solution is presented in Fig. 2. It can be seen

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