Desalination 386 (2016) 48–57

Contents lists available at ScienceDirect

Desalination

journal homepage: www.elsevier.com/locate/desal

Ultrasonic irradiation control of silica fouling during membrane distillation process

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HIGHLIGHTS

- Ultrasonic irradiation was introduced into membrane distillation process.
- Influence of ultrasonic irradiation on silica fouling control was investigated.
- Calcium ion would destroy silica colloid stability and aggravate membrane fouling.
- Ultrasonic irradiation can mitigate silica fouling and maintain permeate flux.

ARTICLE INFO

Article history:
Received 19 November 2015
Received in revised form 18 February 2016
Accepted 25 February 2016
Available online 3 March 2016

Keywords:
Membrane distillation
Membrane fouling
Ultrasonic irradiation
Silica
Permeate flux

ABSTRACT

Ultrasonic irradiation was introduced into membrane distillation process and the influence of ultrasonic irradiation on silica fouling control was investigated. A gradual decline of permeate flux during the concentration process of silica solution was observed due to the formation and deposition of colloidal polysilicic acid on the membrane surface. The PTFE hollow fiber membrane could maintain its mechanical properties and initial pore size distribution in the presence of ultrasonic irradiation. Ultrasonic wave brought significant mechanical and thermal effects, and generated powerful shock wave and microstreaming with high speed. The microstreaming, shock wave and acoustic vortex streaming stimulated the liquid–membrane interface continuously, therefore the membrane surface could be effectively kept clean and the permeate flux was hardly affected by the increasing of concentration factor. Due to the charge neutralization and compression of double charged layer induced by calcium ions, the silica colloid stability was eliminated and the colloids aggregated. A large amount of silica scaling was deposited on the membrane surface even at the beginning of concentration process. With ultrasonic irradiation, the permeate flux maintained stable and was enhanced by about 43%; the ultrasonic irradiation could effectively control silica fouling during membrane distillation.

1. Introduction

Membrane distillation (MD) is a thermal separation process using a hydrophobic membrane as separation media. Being different from conventional evaporation, MD can operate at relatively low temperatures and is thus able to tap into the vast amount of low-grade waste heat [1]. MD is also advantageous over conventional pressure-driven membrane processes, such as nanofiltration (NF) or reverse osmosis (RO), as its low operating pressure reduces the capital cost due to the absence of expensive components, such as high pressure pumps and vessels, as well as pressure exchangers [2]. According to the condensation method adopted, the MD systems can be classified into four different categories: direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD) and vacuum membrane distillation (VMD) [3]. Among these four MD configurations, DCMD is the most studied and simplest in design and application [4], in which condensation steps carried out inside the membrane module, leading to a simple operation mode without the need of external condensers like those in SGMD and VMD.

Although there have been extensive studies on the application of MD for desalination, removal of organic matters in drinking water, treatment of wastewater and recovery of valuable components [5–9], the industrial implementation of MD is not yet feasible and significant advancements

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http://dx.doi.org/10.1016/j.desal.2016.02.032

are still needed to reach the theoretical cost competitiveness and develop market share growth [10]. Membrane scaling in MD process is of particular importance, as scaling can alter membrane surface properties, change membrane pore structure, potentially lead to the wetting of membrane pores and ultimately cause a decline in membrane permeability [11]. Scaling of sparingly soluble salts such as $CaCO_3$, $CaSO_4$, and silicates has been identified as a cause of flux decline when recovering water from natural streams, including brines from desalination processes [12–15]. In recent years, a number of studies have investigated the effect of membrane scaling on the overall MD process utilizing different types of membranes such as flat-sheet and hollow fibers, as well as using different modules [4]. However, the majority of studies focused on the negative effects and mitigation strategies of inorganic membrane scaling such as calcium carbonate or calcium sulfate [16–18]. As far as we know, there are limited studies dedicated to silica scaling in the MD process.

Silica is generally found in water supplies in three different forms: reactive, colloidal and suspended particles [19]. Silica scaling has become a common problem in membrane separation processes due to its low solubility. The prevailing forms of silica are meta silicic acids as $(H_2SiO_3)_n$ with low n numbers. Since silicic acid is a weak acid, it is mostly in the undissociated form at or below a neutral pH. Supersaturated silicic acid can further polymerize to form insoluble colloidal silica or silica gel, which can cause membrane scaling [20]. Madaeni et al. [21] studied the silica fouling problem in the application of membranes for water treatment. The chemical cleaning of silica fouled membrane using acid, alkaline, chelating agent, surfactant and detergent solutions was investigated. It was found that cleaning efficiency depended on the type of cleaning agent and its concentration. The results showed that the efficiency increases with the increasing of chemical cleaning concentration while the mix cleaning solution of EDTA, SDS, and NaOH (0.05 wt.%) was the most effective solution for foulant removal. Although polymerized silica scales can be removed by a high pH cleaning solution (pH of 10–11), it will take many hours for sodium hydroxide solution to remove silica scale at the maximum pH allowed by membrane manufacturer [22]. Neofotistou and Demadis [23] applied antiscalant to mitigate silica fouling in desalination systems. The results showed that the NH_2-terminated dendrimer polyaminoamide (PAMAM) was effective for SiO_2 inhibitors with the optimum dosage. However, the dendrimers would form SiO_2-PAMAM composite precipitates after a long period. Silica inhibitors can retard the polymerization of silica; Butt et al. [24] proposed that phosphonate-based chemicals acted as scale inhibitors in membrane systems. In addition, scale inhibitors such as high molecular weight polyacrylates were also used to increase the solubility of silica and to prevent silica scaling formation [25].

Ultrasonic wave is referred to the acoustic wave with the frequency between 20 kHz and 10 MHz. Several concomitant effects, such as mechanics, thermotics and cavitation effect, during the propagation of ultrasonic wave in various media, have been recognized to be beneficial to many physical and chemical processes [26]. For membrane separation processes, the ultrasonic technique is used mainly in membrane fouling monitoring, membrane cleaning and membrane flux enhancement [27–31]. Li et al. [32–34] applied ultrasonic technique as a non-destructive, real-time, in situ measuring technique for direct monitoring of membrane fouling and cleaning during ultrafiltration (UF) and RO, and found that the ultrasonic technique was a useful technique for the non-destructive investigation of fouling and cleaning in membrane applications. Kobayashi et al. [35–39] introduced ultrasonic technique to create novel anti-fouling membrane processes for membrane water treatment; it was reported that ultrasonic irradiation during membrane filtration was very effective in removing foulants from membranes. Massive evidences exist that the ultrasonic effect is useful for water cleaning of fouled membrane; the ultrasonic cleaning offers advantages and is an effective method compared with other typical cleaning methods using physical and chemical methods [40–42].

The objective of this paper is to introduce ultrasonic irradiation into DCMD process to develop a novel ultrasonic assisted direct contact membrane distillation hybrid process and to investigate the influence of ultrasonic irradiation on silica scaling mitigation. Compared with traditional physical and chemical methods for scaling control, ultrasonic irradiation is expected to be a real-time, in situ technique and can also ensure MD system continuous operation without chemical addition and membrane drying.

2. Experimental

2.1. Materials and membrane module

The polytetrafluoroethylene (PTFE) hydrophobic hollow fiber membrane with a mean pore diameter of 0.26 μm, supplied by DD Water Group Co., Ltd. (China), was chosen to fabricate membrane modules. The SEM images of the PTFE membrane are shown in Fig. 1. The hollow fibers in the number of 40 pieces were assembled into a polyester tube (diameter $d_{\text{in}}/d_{\text{out}} = 15/20$ mm/mm) with two UPVC T-tubes and two ends of the bundle of fibers were sealed with solidified epoxy resin to compose a membrane module. The effective membrane length was 100 mm for each membrane module. The characteristics of the membrane and membrane modules are presented in Table 1.

2.2. Ultrasonic assisted membrane distillation hybrid process

The ultrasonic assisted membrane distillation experimental setup is schematically shown in Fig. 2. The hot feed stirred continually by a magnetic stirrer flowed through the shell side of the fibers, and the cold distillate with conductivity in the range of 22 to 25 μS/cm flowed through the lumen side. The initial volumes of the feed and the distillate were 4.0 L and 0.25 L, respectively. Both solutions were circulated in the membrane module with the help of two magnetic pumps (MP-15RN, Shanghai Seisun Pumps, China). The feed and the distillate flowed co-currently through the module, and the circulation feed rate (V_f) was 0.25 m/s, while the cold side (V_p) being 1.0 m/s. The feed temperature ($T_{f\text{-}inlet}$) was fixed at 53 °C by a Pt-100 sensor and a heater connected to an external thermostat (XMTD-2202, Yongshang Instruments, China). The distillate temperature ($T_{p\text{-}inlet}$) kept at 20 °C by a spiral glass heat exchanger immersed in the constant temperature trough of the cooler (SDC-6, Nanjing Xincheng Biotechnology, China). The temperature of both fluids was monitored at the inlet and outlet of the membrane module using four Pt-100 thermoresistances connected to a digital meter (Digit RTD, model XMT-808, Yuyao Changjiang Temperature Meter Instruments, China) with an accuracy of ± 0.1 °C. An electric conductivity monitor (CM-230A, Shijiazhuang Create Instrumentation Technologies, China) was used to monitor the distillate water quality.

In order to investigate the influence of ultrasonic irradiation on silica scaling mitigation, the membrane module was immersed vertically in a water bath ($15 \times 15 \times 42$ cm^3) and transducers were adhered to the four outside surfaces of the water bath stainless steel shell. The ultrasonic bath was capable of generating ultrasonic with a frequency of 20 kHz and an acoustic power of 260 W. The ultrasonic irradiation device was supplied by Quanyi Electronic Equipment Co., Ltd. (Baoding, China).

2.3. Experimental

Certified analytical reagent grade Na_2SiO_3 and $CaCl_2$ were supplied by Sinopharm Chemical Reagent Co., Ltd. (China). Silicate stock solution was prepared from sodium metasilicate and stored in a polyethylene bottle. The feed silica solution can be obtained by adding known volumes of silicate stock solution and water in the polyethylene container.

The feed silica solution concentration of 150 mg/L was expressed as SiO_2 in this study, and its pH was adjusted to 7.0 using dilute hydrochloric acid before MD experiment. During MD process, no make-up water was added into the feed tank, indicating that the feed was gradually