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Control of protein (BSA) fouling by ultrasonic irradiation during membrane distillation process

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ABSTRACT

Ultrasonic irradiation was introduced into membrane distillation process and the influence of ultrasonic irradiation on protein (bovine serum albumin, BSA) fouling control was investigated. Although the initial BSA concentration did not affect permeate flux in experimental ranges, the feed concentration increasing caused permeate BSA raise due to partial wetting of the hydrophobic membrane. The ultrasonic irradiation could enhance permeate flux about 20% without modification of the hydrophobicity/hydrophilicity of BSA in feed. The higher the concentration factor was, the larger the ultrasonic enhancement of permeate flux could be. Severe permeate flux decline can be found when the salt $CaCl_2$ was added into the BSA solution. The presence of Ca^{2+} would aggravate membrane fouling because the BSA molecules interacted with each other via salt bridging and formed BSA-Ca complex. The BSA aggregates scattered on membrane surface and resulted in a dense fouling layer. With ultrasonic irradiation, ultrasonic wave refreshed liquid-membrane interface continuously and alleviated the deposition of BSA aggregates. Therefore, although there were still some small foulant aggregates scattered on membrane surface, most of the membrane pores kept open and clean, the relative permeate flux can maintain about 98% and was hardly affected by concentration factor increasing.

1. Introduction

Membrane distillation (MD) is an emerging thermally separation process using a microporous hydrophobic membrane as separation media [1]. Compared with other desalination processes such as nanofiltration (NF), reverse osmosis (RO) and conventional thermal evaporation, the potential advantages of MD are as follows: (1) lower operating temperature required than conventional distillation, (2) lower operating pressure than RO, (3) 100% (theoretical) rejection of non-volatile solute, (4) unlimited by high osmotic pressure, and (5) lower energy consumption than multistage vacuum evaporation [2–5].

Although there have been extensive studies on the application of MD for water desalination, removal of organic matters from water, treatment of wastewater and recovery of valuable components [6–9], the industrial implementation of MD is not yet feasible because of the following four major factors [10,11]: (1) low permeate flux of the hydrophobic membrane, (2) membrane fouling and membrane pore wetting, (3) long term performance instability, and (4) optimization and development of effective MD process system. Among these considerations, membrane fouling is of particular importance, as fouling can alter membrane surface properties, change membrane pore structure, potentially lead to wetting of membrane pores and ultimately cause a decline in membrane permeability [12].

In recent years, a number of studies have investigated the effect of fouling on MD process, however, the majority of works focus on the negative effects and mitigation strategies of inorganic scaling such as $CaSO_4$, $CaCO_3$ and SiO_2 [13–15]. As far as we know, there are limited studies dedicated to organic fouling in MD process. Protein-like substances have been identified as one of the major membrane foulants in wastewater treatment, seawater desalination and reclamation applications [16]. With regards to MD process, it is deemed necessary to pay more attention on protein fouling because the hydrophobic membrane surfaces show an especially high tendency to get fouled by proteins due to the strong foulant-membrane affinity [17].

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Severe protein fouling was observed during MD process for aqueous solutions containing organic compounds at representative concentrations, for example, Naidu et al. [18] investigated the membrane fouling development in direct contact membrane distillation (DCMD) using synthetic model solutions of bovine serum albumin (BSA), they found significant fouling and the BSA feed solution showed more significant deposits on membrane surface with less significant pore penetration. Gryta [19] performed concentration of NaCl solution containing natural organic matters (NOMs) by MD, it was found that the presence of NOMs in feed caused the fouling formation and led to rapid flux decline and the major component of the fouling layer was composed mainly of protein and sodium chloride. Hausmann et al. [20] found some minerals and proteinaceous material would penetrate into the membrane for DCMD of whey while skim milk caseins seemed to form a protective layer on membrane surface, they judged calcium playing a stronger role on adhesion in the presence of whey proteins. Thygesen et al. [21] used polypropylene (PP) and polytetrafluoroethylene (PTFE) membranes to remove ammonia from model manure via MD, it was also revealed that the protein was one of the main components in the fouling layer deposited on both PP and PTFE membranes.

It has been demonstrated that boiling and filtration of saline wastewater can reduce the occurrence of protein fouling [22], the wastewater was boiled for 30 min and then was filtered out using a filter paper after cooling to room temperature, this method effectively prevented a rapid flux decline in DCMD. Feed pretreatment with microfiltration (MF) and ultrafiltration (UF) can remove partial protein containing in model manure solution, which was beneficial to permeate flux maintenance, Zarebska et al. [23] also found that the Novadan cleaning agents were the most successful in removing proteins compared with deionized water and NaOH/citric acid. So far, the investigations of protein fouling mitigation and membrane cleaning specially for MD process are still scarce. Nevertheless, it is worth highlighting that the protein fouling control and membrane cleaning methods used in pressure-driven membrane processes may be full of important reference value. Cui and Wright [24] created a gas–liquid two-phase flow across the UF membrane surface by injecting air into the feed stream, suppressed the BSA polarization layer and enhanced the permeate flux dramatically. In addition, the addition of a polyelectrolyte [25] and antiscalant polyaspartic acid [26] to the BSA solution can significantly reduce protein fouling for the PVDF MF membrane and BW30 RO membrane, respectively. Membrane cleaning is a key step to restore the initial flux and to continue the regular separation, both surfactant solutions [27] and saline solutions [28] are common and effective agents using for cleaning of UF membrane fouled by protein. It was also reported that the oxidative cleaning with NaClO would increase the membrane hydrophilicity and improve the initial permeate flux [29].

In recent years, application of ultrasonic technique has been tentatively introduced into membrane filtration processes. Ultrasonic wave is referred to the acoustic wave with the frequency between 20 kHz and 10 MHz [30] and the ultrasonic is used mainly in membrane fouling monitoring, membrane cleaning and membrane flux enhancement [31–35]. Li and Mairal et al. [36,37] applied ultrasonic technique as a non-destructive, real-time, in situ measuring technique for direct monitoring of membrane fouling and cleaning during UF and RO, and found that the ultrasonic technique was a useful technique for the investigation of fouling and cleaning in membrane applications. Xu et al. [38] introduced ultrasonic technique to create novel anti-fouling membrane processes for membrane water treatments, it was reported that ultrasonic irradiation during membrane filtration was very effective in removing foulants from membranes. Massive studies showed that the ultrasonic effect was useful for the fouled membrane cleaning, as an alternative tool, ultrasonic cleaning was more efficient compared with other typical cleaning methods using physical and chemical methods [39,40].

Although ultrasonic irradiation has been successfully applied to enhance the performance of different pressure-driven membrane separation process, relatively few studies have been carried out with the use of ultrasonic to mitigate protein fouling in MD process. The objective of this paper is to introduce ultrasonic irradiation into DCMD process and to investigate the influence of ultrasonic irradiation on protein fouling mitigation. Compared with traditional physical and chemical methods for membrane fouling control, ultrasonic irradiation is expected to be a real-time, in situ membrane fouling mitigation technique and can also ensure MD system continuous operation without chemicals addition and membrane drying.

2. Experimental

2.1. Materials and membrane module

The PTFE hydrophobic hollow fibers with a mean pore diameter of 0.26 μm, supplied by DD Water Group Co., Ltd. (China), were chosen to fabricate membrane modules. The SEM images of the PTFE membrane are shown in Fig. 1. Forty pieces of hollow fibers were assembled into a polyester tube (diameter d_{in}/d_{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 characteristics of the membrane and membrane modules are presented in Table 1.

2.2. Membrane distillation setup

The MD 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 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 was 0.25 m/s, while the cold side being 1.0 m/s. The feed temperature 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 kept at 20 °C by a spiral glass heat exchanger immersed in the constant temperature trough of the cooler (SDC-6, Nanjing Xinchen 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 protein fouling mitigation, the membrane module was immersed vertically in a water bath ($15 \times 15 \times 42$ cm^3), 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).