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Ultrasonic defouling of reverse osmosis membranes used to treat wastewater effluents

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Abstract

On-line ultrasonic cleaning was used to remove fouling from a commercially important polyamide based reverse osmosis membrane during cross-flow filtration of $CaSO_4$, Fe^{3+} and carboxyl cellulose solutions. In each case, the permeate flux of the membrane increased significantly, with virtually no decrease in rejection in the presence of ultrasonication. Membrane surface characterization via scanning electron microscopy (SEM) confirmed the beneficial effect of ultrasonication on the membrane permeate flux. These studies suggest that ultrasonic defouling may be a very useful approach for the future development of reverse membranes, especially as far as fouling with organic materials is concerned.

Keywords: Defouling; Reverse osmosis membrane; Flux enhancement; Ultrasound

1. Introduction

Membrane fouling is one of the main problems encountered in the operation of membrane systems used in the treatment of wastewater effluents. Fouling occurs through the irreversible deposition of retained particles, colloids, macromolecules and salts at the membrane surface and/or inside the membrane. This can lead to a significant decline in the permeate flux [1]. Many techniques have been developed to overcome fouling. Backwashing and backflushing are typically used to clean membranes, while more recalcitrant foulants can be removed by use of detergents and acids or alkalies [1–4]. However, these chemicals sometimes damage the membrane materials and cause secondary pollution [5]. As a consequence, other physical cleaning methods not requiring the use of chemicals have been developed, e.g. periodic reversals in the flow direction and periodic reduction in the feed pressure along with continuous flow [4], using pulsating and reverse flow [6], magnetic and centrifugal fields [7], as well as the sparging of gas bubbles into the feed [8–13]. More recently, applications of electric field [14] and electrokinetics [15] have shown considerable promise, but these methods are generally limited to ionic foulants.

At the same time, the application of ultrasound for enhancing permeation in membrane processes has been extensively studied [16–23]. The basic physical phenomenon behind the effect of ultrasound is cavitation, i.e. the formation, growth and implosive collapse of bubbles in the liquid. These bubbles or cavities are formed by ultrasound waves passing through the medium in a series of alternate compression and expansion cycles. Hot spots are created in the liquid where the temperature and pressure of the gas in the cavity rises to enormously high values, owing to the expansion and implosive collapse of bubbles at nucleation sites within the liquid. Further, when a cavitating bubble is oscillating near a solid surface, it does so asymmetrically, resulting in the generation of microjets (microstreams) of high velocity. Fluid flowing at these high velocities can decrease the thickness of boundary layers and diffusional resistance and therefore enhance the rates of mass transfer. Most of the studies to date were concerned with the enhancement of electrolyte diffusion through dialysis membranes by ultrasound irradiation. For example, Li et al. [21,22] used ultrasonic equipment to enhance electrolyte dialysis through membranes and found that the amount of solute in the permeate increased with acoustic pressure.

The effect of ultrasound was also examined by using cellophane membranes for salt dialysis, such as sodium, potassium and calcium chlorides. The variables investigated included the intensity of the ultrasound field, the solute concentration and the irradiation time [24]. Moreover, the enhancing effect of specially

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modulated ultrasound signals on a water desalination process in ion-exchange hollow fibre modules was investigated by Band et al. [16]. Depending on the solution concentration and the hydrodynamic conditions of the membrane process, the ultrasound treatment enhanced permeation.

Kost and Langer's patent [18] proposed a method for enhancing or controlling the permeability of low and high molecular weight molecules in a membrane system, which is exposed to ultrasound of selected intensity of 0.05–30 W/cm^2 and frequency between 10 kHz and 20 MHz for most polymeric membranes and an intensity of 0.05–3 W/cm^2 and frequency of 1–3 MHz for biological membranes. A Japanese patent [25] proposes the application of ultrasound to a purification process in which water is passed through an apparatus containing an ion-exchange resin bed. Ultrasound, which is generated constantly during the adsorption and regeneration periods, breaks the boundary layer and improves these processes.

Although ultrasound has been used to enhance the performance of membrane separation, relatively few studies have dealt with the use of ultrasonication to eliminate or prevent the fouling of membranes. The patent of Harvey [26] proposed the use of an acoustic liquid whistle or ultrasound transducers to produce cavitation to prevent clogging of membranes and to remove concentration–polarisation in a water desalination process with semi-permeable reverse osmosis membranes. In order to eliminate or reduce polymeric membrane fouling, Chai et al. [20] reported the anti-fouling effect of ultrasound in the ultrafiltration of dextran solutions with polyacrylonitrile membranes. Likewise, Kobayashi et al. [27] attributed an observed increase in permeate flux to the cleaning of membrane fouling by ultrasound irradiation. The ultrasound effect was also studied for cleaning microfiltration and ultrafiltration membranes after peptone filtration [19]. They have shown that ultrasound promoted water washing of membranes. Despite these efforts, relatively little is still known about the use of ultrasonication for the defouling of membranes. Consequently, in the present study ultrasound was used to clean a commercially important polyamide-based reverse osmosis membrane fouled with both organic and inorganic contaminants.

2. Experimental work

A commercial polyamide-based reverse osmosis membrane suitable for low-pressure operation was obtained from Fluid Systems Company. The $CaSO_4$ solution was prepared by the reaction of certain amounts of $Ca(OH)_2$ and H_2SO_4 (analytically pure reagents obtained from Sigma). Desired concentrations of Fe^{3+} were prepared by dissolving certain amounts of $FeCl_3{\cdot}6H_2O$ (analytical grade from Merck) into distilled water, and the pH of the solution was changed to 4.5 by use of HCl (analytical grade from Merck). The sample of carboxymethyl cellulose (CMC) with a molecular weight of 250,000 was obtained from Aldrich. The CMC solutions were prepared by dispersing a known weight of depressant in cold distilled water and then dissolving it with boiling distilled water. The solutions were prepared fresh each day. Distilled water was used in all tests.

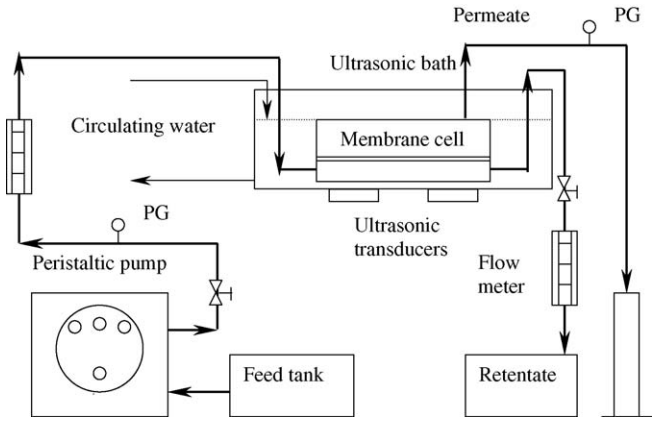

Fig. 1. Schematic diagram of membrane filtration in an ultrasonic bath. PG denotes the pressure gauge.

The experimental set-up is shown in Fig. 1. The membrane was placed in the cross-flow filtration unit, which was immersed in a water bath (30 cm × 24 cm × 20 cm) of a sonicator (Ultrasonic cleaner, Denmark). The ultrasonic bath was capable of generating ultrasound with a frequency of 20 kHz and a power intensity of 2.8 W/cm^2. Two of the four ultrasonic transducers in the ultrasonic bath were used to act on the membrane cell. The effective membrane filtration area was 14 cm × 4 cm. The temperature of the solutions in the feed tank was maintained at 20 °C to within ±1 °C. During the experiments, the flow rate of the feed solutions and operating pressure were maintained at about 5 mL/min and 100 kPa, respectively. The permeate flux (L m^{-2} h^{-1}) of the membrane was determined by measuring the volume of the permeated solution at certain intervals. The calcium and iron concentrations in the feed, retentate and permeate solutions were analysed with an atomic absorption spectrometer (Varian, AA20, Australia). The CMC concentrations in the feed, retentate and permeate solutions were determined by the total organic carbon (TOC) using a TOC analyser (Dhorman). Experiments were replicated at least once and only the mean values are reported.

Fresh sheet of the flat polyamide reverse osmosis membrane was cut into rectangular sections of approximately 18 cm × 6 cm. All membranes were cut from a single sheet of membrane for all the filtration experiments on the same types of solutions. All the membranes were immersed in distilled water for 24 h prior to the filtration experiments. The morphological changes of the membrane with and without ultrasonic treatment were investigated by using a scanning electron microscope (SEM) images.

The rejection values reported in this study are based on the following definition:

$$R = 1 - \frac{C_P}{C_R} \quad (1)$$

where C_P is the concentration of the permeate and C_R is the concentration of the retentate. C_P and C_R thus represents the steady state bulk concentration of Ca, Fe and TOC in the permeate and retentate streams leaving the membrane cell in the reverse