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Fabrication and in-situ fouling mitigation of a supported carbon nanotube/γ-alumina ultrafiltration membrane

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ABSTRACT

A novel ultrafiltration (UF) membrane with built-in defouling capability was made, characterized and tested for its water purification performance. The asymmetric ultrafiltration membrane with pore size of ~ 8 nm is obtained by coating a carbon nanotube (CNT)/γ-alumina composite layer on a porous $PbZr_{0.52}Ti_{0.48}O_3$ (PZT) piezoelectric support. The PZT support not only serves to provide mechanical strength to the membrane, but can also be used to generate ultrasound by application of an alternating voltage (AV). This ultrasound, in turn, avoids and/or removes any fouling during filtration. The conducting composite layer serves both as a size-selective membrane and an electrode. The optimum membrane composition was a 1:1 CNT to alumina weight ratio at a sintering temperature of 600 °C. PZT-supported membrane structures were poled with a 3×10^3 kV/m electric field. Filtration of poled and unpoled membranes was carried out with a 2.5 g/L dextran solution to test anti-fouling performance. It was found that a poled membrane with application of a 20 V AV had a stable permeance of 55.6 $\mathrm{L\,m^{-2}\,h^{-1}\,bar^{-1}}$. The permeance of an unpoled membrane without application of a voltage was 31.0 $\mathrm{L\,m^{-2}\,h^{-1}\,bar^{-1}}$. This 79% permeance increase is ascribed to the mitigation of fouling during the filtration of the dextran solution.

1. Introduction

Membranes are widely used in water treatment and purification due to their high cost-efficiency and minimal environmental impact [1]. However, water purification membranes tend to foul. They are covered by or impregnated with retained particles, colloids, macromolecules and precipitates [2]. This results in significant decrease in flux, and consequently increase of operational costs and the need for off-line cleaning and premature failure of membranes. Fouling is the foremost technical challenge in membrane filtration [3].

Many research efforts have been made to develop methods for limiting or reducing membrane fouling. Examples include application of vibration [4], gas sparging [5], electrical fields [6] and ultrasound [7]. After the application of ultrasound (US) was reported for the first time, in 1980 [8], several more studies of membrane cleaning [9,10] and fouling control [11–13] followed. It was confirmed that, when intense US waves propagate through a liquid, gas bubbles form in the negative pressure waves when the liquid's local tensile strength is exceeded. These bubbles rapidly grow and subsequently collapse in the positive waves resulting in a strong localized energy release. This process is known as cavitation, often utilized in the cleaning of surfaces [14,15]. Kobayashi et al. [16,17] investigated the effects of ultrasonic conditions during cross-flow filtration with flat sheet membranes. They found that ultrasound frequency, power density and the irradiation direction had a significant effect on flux recovery after treatment. Gondrexon et al. [18,19] investigated the effect of ultrasound on membrane fouling during ultrafiltration. They found that this application of ultrasound significantly increased the water permeance for ultrafiltration of nano-particles, natural clay and skim milk. A permeance enhancement factor of 1.6–13.5 was found.

Recently, the in-situ generation of ultrasound by a piezoelectric layer inside the membrane was introduced to sidestep the requirements of external ultrasound generation and/or off-line cleaning [20–23]. We believe that this use of a built-in structure will result in a better efficiency by a sophisticated optimization of continuous, in-line fouling mitigation. In addition, we anticipate that the ultrasound, emanating from the membrane structure will disrupt the formation of laminar boundary layers. These boundary layers adversely affect salt rejection and are thought to facilitate the settling of fouling particulates. To have the piezoelectric layer emit ultrasound, AVs are applied between both sides of the PZT supported CNT/γ-alumina composite membrane. In-situ sound generation has also been reported [20–23] for membranes

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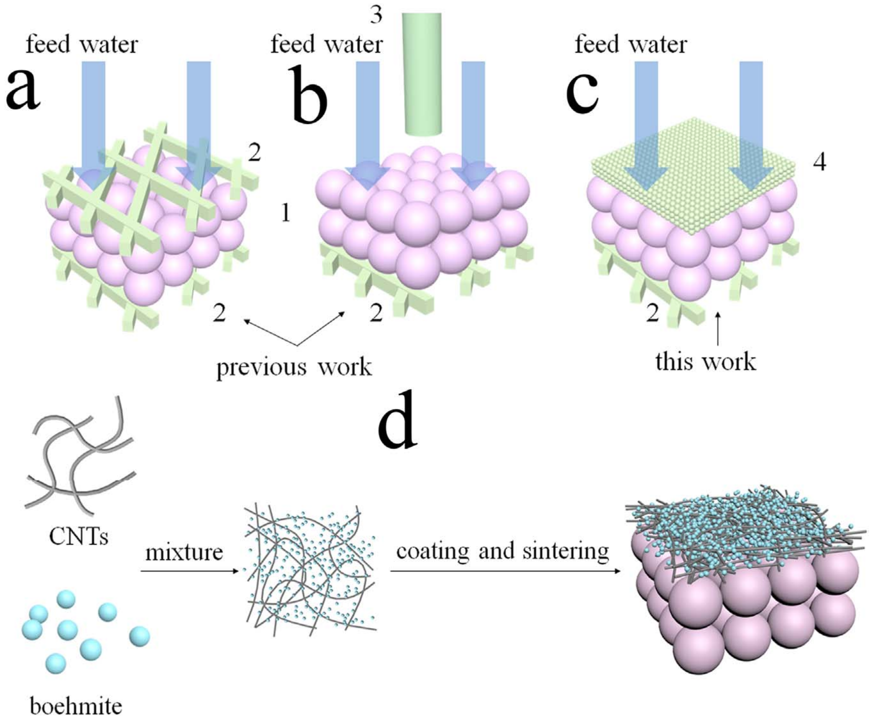
made of piezoelectric polyvinylidene fluoride (PVDF) polymer. PDVF has at least four known crystalline structures (α, β, γ and δ) and is commonly used for micro- and ultra-filtration (MF and UF). The membranes studied had a pore size of 220 nm, and the PVDF membrane, was poled in an electric field of 16.3 × 10^3 kV/m to transform all crystalline structures to the all-trans (β) phase which is mainly responsible for its piezoelectric properties. It was found that application of 0.5–1 kHz AVs to poled PVDF membranes during filtration resulted in an increase of the permeance and, simultaneously, a decrease of the fouling rate. After 30 min filtration, the permeance of a PVDF membrane with no voltage applied was 50 $\mathrm{L\,m^{-2}\,h^{-1}}$, while the permeance of a PVDF membrane with an effective AV of 10 V application was increased to 160 $\mathrm{L\,m^{-2}\,h^{-1}}$. In another study, lead zirconate titanate (PZT), an inorganic piezoelectric material, was used to fabricate a macro-porous membrane with an average pore size at about 300 nm [24]. It was found that application of an AV at 70 kHz, at sonic resonance, to a poled PZT membrane during dead-end filtration of a 1 μm latex dispersion resulted in complete fouling mitigation, the permeance was maintained at its initial value.

The geometries of PVDF and PZT piezoelectric membranes reported in literature are shown in Scheme 1a and b. A porous steel electrode is present at the permeate-side of the membrane, another porous steel electrode is present at or above the feed-side of the membrane. A feed-side electrode at mm's distance away from the membrane is referred to as a “remote electrode”. A remote electrode, the “rod electrode” in Scheme 1b, has the advantage that it does not adversely affect the membrane process at the feed side, for instance by reduction of filtration surface, promotion of localized fouling, and inhomogeneous field distribution. Remote application of a voltage to the piezoelectric layer is possible due to capacitive coupling in combination with minor ionic conductivity in the liquid [24]. However the distance between the remote electrode and the membrane surface may lead to substantial, unwanted electrical energy dissipation.

In this paper, we present fabrication and characterization of a composite membrane with an asymmetric structure as shown in Scheme 1c. Disk-shaped porous PZT supports with 300 nm pore size are made by pressing and sintering and provided with a conductive layer with an average pore size of < 10 nm. In this structure, the functions of feed-side electrode and size-exclusion filtration are combined in a separate membrane that is supported by the PZT layer. The conducting thin film membrane is a composite of carbon nanotube (CNT) and 15 nm γ-alumina particles. We have chosen for a composite structure containing CNTs since CNTs have a superior electrical conductivity and thermo-chemical stability [25,26]. Ultrafiltration membranes consisting of aligned CNT [27,28] and CNT networks [29,30] were demonstrated to have a high water permeance due to their ultrathin and fibrous morphology. Jassby et al. [31] synthesized conducting ultrafiltration membranes based on the CNT and polysulfone. Huang et al. [32] prepared ultrafiltration membrane that had a composite CNT/PVDF structure. Both membrane types adhered well to their polymeric supports (PS and PVDF) and had excellent electrical conductivity. But during our initial explorations and the literature [33], it was found that 100% CNT networks are too much prone to mechanical damage from wear and scratching. Hence it is necessary to enhance the mechanical properties of the CNT networks for practical application. Methods such as CNT modification [34,35] and adding binders [36,37] were found to improve the mechanical properties of CNT networks. In this work, we studied the use of CNT/γ-alumina composite structures for the membrane. The alumina particles form strong necks to reduce their external surface. We anticipated that the presence of hydrophilic alumina between the hydrophobic CNT would improve the adhesion with the hydrophilic PZT ceramic support, and at the same time improve scratch resistance (Scheme 1d). While the effect of CNT reinforcement on the mechanical properties of alumina is well-documented [38–42], no studies have been reported for the effect of alumina particle additions to CNT networks.

In the present work, composite CNT/γ-alumina membrane layers were prepared by dip-coating CNT/γ-alumina suspensions on porous PZT substrates, followed by thermal processing. The effect of thermal processing and composition were studied by SEM, EDS, N_2 sorption and nano scratch tests. The eventual optimized composite membrane was poled in an electric field of 3 × 10^3 kV/m at 120–140 °C to obtain permanent piezoelectricity. Water purification performance was tested with dextran solutions to evaluate the membranes' transport properties and anti-fouling performance.



Scheme 1. a, b, c: Schematic of the cross flow membrane module used for membrane operation with in-situ ultrasound generation: 1) symmetric piezoelectric membrane, 2) permeate side mesh electrode, 3) remote rod electrode, 4) asymmetric structure with a conductive membrane layer and a piezoelectric support. d: Composite membrane fabrication process.

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