



Nanoparticle charge affects water and reverse salt fluxes in forward osmosis process

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ABSTRACT

In this study, the mechanism was studied of membrane fouling that occurred when charged nanoparticles (NPs) are present in the feed solution of the forward osmosis (FO) process. Three NPs were used that were positively, neutrally, and negatively charged, respectively, at pH 6.0: zinc oxide (ZnO), titanium dioxide (TiO₂) and silicon dioxide (SiO₂). The FO membrane used was a negatively charged polyamide-based flat sheet. The fouling behavior arising in the presence of each NP type varied according to electrostatic attraction and repulsion between the NPs and the membrane; this affected the water flux and reverse salt flux (RSF). Positive NPs accelerated membrane fouling close to the membrane surface by their electrostatic attraction with the membrane, leading to the formation of a thick fouling layer that acted as a path for salt ion transfer, thereby increasing RSF. On the other hand, negatively charged NPs were electrostatically repelled by the membrane surface and thus, prevented the NPs from attaching easily to the membrane surface, thereby reducing fouling and leading to increased water flux and decreased RSF. To rectify this mechanism, the positively charged NPs was altered to negative by changing the pH; this enhanced water flux and reduced RSF.

1. Introduction

Forward osmosis (FO) is a low-energy membrane-based separation process in which the driving force for separation is the difference in chemical potential between a concentrated draw solution (DS) and a broad range of aqueous solutions, including contaminated wastewater. The FO process produces water using the osmotic pressure difference between the feed solution (FS) and an induction solution (or DS) across a semipermeable membrane [1]. FO can be combined with other membrane processes and has been investigated for a wide range of applications, including treatment of industrial and domestic wastewater, concentration of liquids in food industry and agriculture, and seawater desalination [2]. A previous study has stated that the FO process can be strategically used as a substitute of applications using the pressurized membrane processes [3]. FO-reverse osmosis (RO) hybrid system has been identified as an alternative desalination process in which FO complements the weak points of RO. Firstly, seawater as a DS is diluted by wastewater, which is FS in the FO process. Then the resulting diluted seawater is sent to RO, which can reduce the total energy of the desalination process by decreasing the osmotic pressure during RO. In addition this hybrid process allows both seawater desalination (by RO), and wastewater reuse (by FO) [4].

The FO process is comparable with conventional wastewater

treatment processes such as treatment or desalination of complex industrial streams, especially in the treatment of wastewaters from the exploration and production of oil and gas [5,6]. Other studies have aimed at developing understanding of water transport phenomena through FO membranes, for instance regarding the influences of internal concentration polarization, membrane structure and material, and membrane orientation upon membrane flux [7,8]. The FO process is not free from membrane fouling, especially when applied for wastewater treatment. There are many different organic and inorganic substances in wastewater, which reduces the process efficiency [9]. Moreover, the raw water used in the process of making low-concentration RO feed water for the FO membrane is mainly composed of domestic wastewater; this can accelerate fouling and thereby significantly reduce the efficiency of the process [4].

Despite the benefits of the FO process, its application to real-scale water purification has been hindered by three challenging problems [10]. First, the DS must create an osmotic pressure as a driving force, and then be recovered or removed efficiently in further steps; finding appropriate draw solutes and effective recovery techniques is a topic of wide investigation. Second, more appropriate FO membranes needs to be developed to have high water flux with low reverse salt flux (RSF). Third, although a great deal of research has been conducted on pressure-driven membrane processes, a systematic mechanistic

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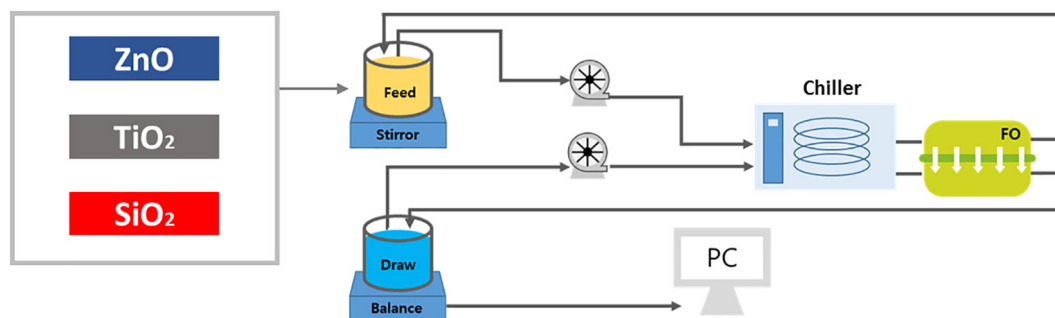


Fig. 1. Experimental setup of a laboratory FO system.

understanding is lacking regarding the fouling and cleaning behaviors of FO processes [11].

Fouling is one of the most inevitable phenomena in membrane filtration operations [12]. Remedial actions such as membrane cleaning or feed water pretreatment are often used to reduce membrane fouling or to increase the available operation time between cleanings [13]. Membrane fouling is affected by operating conditions such as cross flow or pressure, but the underlying cause depends on the fouling potential of the feed water. Therefore, quantifying the fouling potential of the feed water is useful for membrane fouling control and mitigation. Particulates are foulants in many membrane processes and stacked on the membrane surface, block the pores of the membrane and degrade process performance, adding energy and chemical wastewater due to frequent cleaning [14]. Similarly, FO membranes have been found to be vulnerable to accumulated by the particles. Previous research was conducted regarding an FO process including pretreatment by filtering through a 0.45 μm cartridge filter [15]; this pretreatment led to increased flux but more than a fifth of the foulants that remained after the pretreatment were particulates 3 μm or smaller. It is shown that, understanding the membrane fouling mechanism is necessary to enhance economy and efficiency of the process [16].

Usually membranes have negative surface charge [17,18] and foulants present in wastewater are also negatively charged [9,19]. Electrostatic, and to a lesser degree, chemical interactions between the membrane and foulants are the main factors affecting membrane fouling [20]. Particulate substances also have charge polarities [21], similar to organic substances, and these polarities may also affect membrane performance. Various studies on this subject have been conducted in pressure-driven membrane processes [22,23]. However, studies investigating the mechanisms of fouling in FO are rather scarce, and information on particulate fouling of FO membranes is limited in the literature. To date, there has no work investigating the effect of particle charge upon FO membrane surface and fouling behaviors in runs performed under identical physicochemical conditions [3].

Most researches related to FO have been focused on the finding new DSs that greatly reduce the RSF and allow easy reuse of the DS, and the developing new membrane materials that will raise the relatively low fluxes of the FO process compared to those offered by pressurized membrane-based processes. However, the identification of the mechanisms of FO membrane operation and the finding a suitable operation strategy for the process application through the characterization of the developed membrane are also essential. The purpose of the present research is therefore to investigate the effect of particles' charge upon FO performance, namely water flux and RSF. Differently charged nanoparticles (NPs) were added into feed solution to understand their roles in the FO process.

2. Materials and methods

2.1. Materials

2.1.1. Nanoparticles (NPs)

Zinc oxide (ZnO), titanium dioxide (TiO_2) and silicon dioxide (SiO_2) were positive, neutral, and negative charged NPs, respectively, at pH 6.0. All the NPs purchased from Sigma-Aldrich. All NPs had the same size distribution of 20–40 nm. SiO_2 and TiO_2 were stored in powder form and ZnO was dispersed in deionized (DI) water. Zeta potential values of these materials are given in Section 3.1. NP surfaces have different chemical compositions. They have oxygen (O-) atoms are involved mostly have negative charges and that the surface charges are strongly dependent on the pH value of the liquid phase. To control the charge of the NPs, and to simulate a pH range (about 5.5–7.0) similar to that of actual sewage treatment plants [4], optimal pH range was investigated and it was found to pH 6.0 [24,25]. Adjustment of pH was carried out using 0.01 M HCl and NaOH.

2.1.2. Membrane

FO membranes purchased from Porifera Inc. (California, USA) were used in this study. The active layer of the membrane is known to be made of polyamide (PA), supported by polysulfone and polyester materials [4]. Prior to use, the membranes were rinsed several times with DI water, then soaked in DI water and stored in a refrigerator at 4 $^{\circ}\text{C}$ [26].

2.2. FO test

2.2.1. Operation conditions

Fig. 1 schematically illustrates the experimental setup and Table 1 lists the experimental conditions. The flat frame FO membrane cell used consisted of two symmetric channels (L 77 mm, W 26 mm, D 3 mm); the effective area of the membrane was 20 cm^2 . The FS and DS were supplied at the cross flow velocity of 8.55 cm s^{-1} into the FO membrane by a gear pump (Longer Pump WT3000-1FA, China). A stirrer was used to

Table 1
FO operational conditions.

Name	Condition
Cross flow velocity	8.55 cm s^{-1}
Feed solution (FS)	DI water + NPs (1 L)
Draw solution (DS)	0.5 M NaCl (1 L)
NP concentration	6 mg L^{-1}
pH	6.0
Temperature	25 \pm 0.5 $^{\circ}\text{C}$
Operational time	3 h ^a

^a In Test 1, the 3 h of monitored operational time to observe the initial accumulation of NPs on the membrane. In Test 2, the 3 h of monitored operational time after a 12 h unmonitored period of operation to allow a fouling layer to form on the membrane.

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