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Analysis of an osmotically-enhanced dewatering process for the treatment of highly saline (waste)waters

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ABSTRACT

The dewatering of highly saline (waste)waters by typical osmotic membranes, such as reverse osmosis (RO) or forward osmosis (FO), was significantly improved by a novel process in which an osmotic pressure gradient across the membrane is eliminated or reduced by increasing osmotic pressure in the permeate side. In this work, the concept of an osmotically enhanced dewatering (OED) process was fundamentally analyzed via conceptual modeling and verified experimentally under various hydraulic and osmotic pressure conditions. No or less osmotic gradient across the membrane resulted in higher water recovery than RO. Larger water flux was also produced than FO because the loss of osmotic driving force by internal concentration polarization (ICP) was greatly reduced. For instance, a series of experiments demonstrated that water flux of 1.2 LMH was obtained at low hydraulic pressure of 15 bar when a feed of 2.4 M NaCl was dewatered by the OED process. In addition, membrane characteristics (A, B, S) were optimized by modeling, and further examined experimentally using typical NF and FO membranes. Lastly, less reverse solute diffusion ensured a product of high quality after dewatering, suggesting that this process can be applied to not only highly saline shale gas produced water treatment, but also protein and pharmaceutical enrichment.

1. Introduction

As the demand for water rises with population growth, industrialization, and increasing stress on existing water sources, there is an impetus to purify impaired water sources such as effluent from plants, municipals or mines [1,2]. Impaired water is a multicomponent mixture that can contain high concentrations of dissolved solids, toxic heavy metals, and valuable resources. Thus, if the impaired waters can be treated, the reliability of freshwater sources into which the effluent is discharged can be improved and the treated wastewater itself can act as an independent water source – especially for inland sites [3–5].

The most common practice for the dewatering and reuse of wastewater is the use of nanofiltration (NF) and reverse osmosis (RO) membranes, because of their ability to extract fresh water from wastewater. These membranes have been widely used in desalination [6], water softening [7,8], the removal of heavy metals [9], and wastewater treatment [10]. In the RO and NF processes, however, considerable hydraulic pressure is required to act as the driving force for the water flux to overcome the osmotic pressure of the feed solution. As a result, water recovery is limited by the feed concentration and the burst pressure of the membrane, which is a major drawback for the

application of this process, particularly to the wastewater characterized by high salinity.

On the other hand, previous studies have reported that forward osmosis (FO) can efficiently treat large volumes of wastewater [11–13]. FO uses differences in osmotic pressure to drive the transport of water across the membrane, leading to high water recovery through the use of a high saline draw solution. Although FO systems that utilize thermolytic draw solutions such as ammonium bicarbonate may be more energy efficient than thermal-driven processes, thermolytic draw solutes must also be vaporized and recovered, a process which requires high thermal energy for the solution to change phases [14]. In addition, using a high concentration draw solution elevates the rate of reverse solute diffusion. Solutes that diffuse from the draw solution can contaminate the feed solution and lower the purity of the product [15].

In this paper, we newly propose an osmotically enhanced dewatering (OED) process that can be used to effectively concentrate a feed solution with high salinity. In this OED process, instead of using only hydraulic pressure to create the driving force for water transport across a semi-permeable membrane, an osmotic pressure equalizer (OPE) is utilized in the permeate side simultaneously to offset the osmotic pressure of the feed solution as schematically illustrated in Fig. 1.

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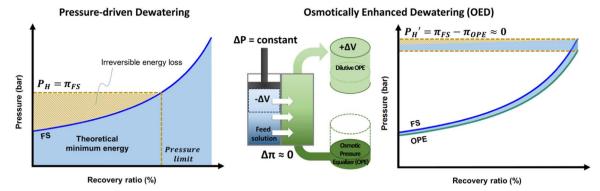


Fig. 1. The conceptual illustration of the OED process with the energy use of pressure-driven and osmotically enhanced dewatering. The theoretical minimum energy required for dewatering is represented by the blue area under the osmotic pressure curve. For dewatering to occur, the hydraulic pressure applied to the feed solution must be higher than the osmotic pressure difference; this required energy is represented by the area of the rectangle under the dashed line. The difference between the two areas, indicated by the brownish hatched area, represents irreversible energy loss. The OED process can operate at lower applied pressures (P_H ' = π_{FS} - π_{OPE}) because the OPE solution reduces the difference in osmotic pressure, resulting in much less irreversible energy loss. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.).

Typically, the thermodynamic limit of pressure-driven membrane process (i.e., RO/NF) occurs at the point where the hydraulic pressure is equal to the osmotic pressure of the feed solution ($P_H = \pi_{FS}$). However, the pressure required in our OED process ($P_{H'} = \pi_{FS} - \pi_{OPE}$) is significantly lower because the OPE neutralizes the osmotic pressure of the feed solution ($\pi_{FS} \approx \pi_{OPE}$), leading to the production of more water without increasing hydraulic pressure. Under these conditions, the net driving force for water flux depends solely on the applied hydraulic pressure. Therefore, the OED process can be applied beyond the limits of water recovery arising from the maximum allowable hydraulic pressure of the membrane modules and pumps in other pressure-driven processes [16]. Furthermore, the overall irreversible energy loss is significantly minimized, compared to that observed in typical RO process as shown in Fig. 1. A similar process of using the saline permeate solution was introduced as an osmotically assisted RO in a recent modeling work [17]. However, further fundamental study including the experimental validation is greatly needed to explore the potential of such a process for dewatering of highly saline wastewaters.

In this study, the concept of an osmotically enhanced dewatering process was more fundamentally analyzed by theoretical modeling and laboratory experiments under different osmotic/hydraulic pressure ratios. Then, we newly defined its applications compared with other osmotic membrane processes; RO, FO and PAO (pressure-assisted osmosis). Furthermore, the OED process model developed in this work was experimentally validated, including its performance dependence on the concentration of OPE as well as hydraulic pressure applied. This model was further utilized to gain insight into the system performance for dewatering and to explore the impact of membrane properties such as water and solute permeability, and the structure parameter on the performance. Based on the results of these analyses, we discuss the potential use of the OED process in practical applications, particularly for the treatment of highly saline wastewaters.

2. Materials and methods

2.1. Osmotic membranes

Two commercial FO and NF membranes were used for this study: CTA FO from HTI and NF90 from DOW Filmtec. After rinsing with deionized (DI) water several times, all membrane samples were stored in DI water at 4 $^{\circ}\mathrm{C}$ prior to the experiments. The characteristics of these membranes are summarized in Table 1.

2.2. Experimental setup

The OED experiments were performed with a laboratory cross-flow

Table 1Characteristics of the osmotic membranes used in the lab-experiments.

	CTA FO	NF90
MWCO (Da)	_	200 [18]
Bursting pressure (bar)	48 [19]	41
Water permeability, A (L m ⁻² h ⁻¹ bar ⁻¹)	0.49	8.14
Salt permeability, B (L m ⁻² h ⁻¹)	0.68	4.39
Structure parameter, S (μm)	398	6354

system configured in the same manner as described in our previous studies [20,21]. The OED test cell had a rectangular channel on each side of the membrane. The dimensions of the symmetric channels were 77 mm L \times 26 mm W \times 3 mm H. A diamond mesh spacer was inserted into the feed channel and 10 permeate carrier sheets were placed in the permeate channel to support the membrane by preventing the deformation of the active layer under high hydraulic pressure [22,23]. A variable speed gear pump (Cole-Parmer, Vernon Hills, IL) and a high-pressure pump (Hydracell, Minneapolis, MN) were used to recirculate the OPE and the feed, respectively. The cross-flow velocity of both channels was kept constant at 17.1 cm/s. The applied pressure was monitored using digital pressure meters and controlled with a back-pressure regulator at the feed channel outlet. The temperature of both the feed and osmotic pressure equalizer was maintained at $25.0 \pm 1.0\,^{\circ}\text{C}$.

2.3. Feed and osmotic pressure equalizer solutions

Two different solutions were used as feeds in this study to analyze OED performance: one containing sodium chloride (NaCl) and the other with both NaCl and calcium chloride (CaCl₂). An NaCl or magnesium chloride (MgCl₂) solution was used as the osmotic pressure equalizer. All of the chemicals used were reagent grade. The concentration of the OPE was adjusted to yield various osmotic pressures in order to neutralize the osmotic pressure gradient between the feed solution (FS) and the OPE.

2.4. Measurement of water flux and solute rejection

The performance of OED using FO and NF membranes in terms of water flux and solute rejection was determined using the OED unit described above. The water flux (J_w) passing through the membrane was measured based on the weight change of the OPE bulk solution. To measure the rejection of cations from the mixture solution in OED mode, samples of the feed and osmotic pressure equalizer were taken after a complete OED run and the concentration of each solute was

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