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## Evaluation of the transport parameters and physiochemical properties of forward osmosis membranes after treatment of produced water



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#### ABSTRACT

The application of semipermeable membranes for dewatering of complex oil and gas wastewaters continues to be a topic of increasing interest. Several studies have explored the fouling propensity and contaminant rejection of osmotically driven membranes during forward osmosis (FO) treatment of produced waters; however, none have investigated changes in membrane transport and physiochemical properties after exposure to these feed streams. In this study we discuss the impacts of produced water exposure on the transport and active layer surface properties of cellulose triacetate (CTA) and polyamide thin-film composite (TFC) FO membranes. While produced water exposure yields some, albeit minor changes to the membrane performance and surface characteristics of the CTA and the traditional TFC membranes, close to 50% reduction in reverse salt flux and contaminant transport was observed for a surface-modified TFC FO membrane; only minimal changes in water permeability were recorded. Results of this study demonstrate the chemical and physical robustness of FO membranes for treatment of oil and gas wastewaters, and they highlight a knowledge gap that exists in membrane polymer selection and contaminant interactions with the membrane polymer matrix that should be further addressed in future membrane fouling studies.

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#### 1. Introduction

1.1. Separation of complex wastewaters by semipermeable polymeric membranes

The application of semipermeable polymeric membranes for dewatering of complex feed streams, and especially those laden with a variety of organic compounds and hydrocarbons, has increased in recent years [1–8]. This is especially true in the oil and gas industry, where rapid oil field exploration has spurred the development of tight-barrier membrane processes for treatment of wastewaters like hydraulic fracturing flowback and produced waters [3–6,8–15]. While exceedingly higher total dissolved solids (TDS) concentrations in produced water has been a traditional hurdle for a variety of water treatment technologies, the prevalence of dissolved organic and aromatic compounds in oil field wastewaters has gained significant attention [16–21]. The organic compounds present in feed streams like produced water exhibit a

http://dx.doi.org/10.1016/j.memsci.2015.09.031 0376-7388/© 2015 Elsevier B.V. All rights reserved. wide range of physiochemical properties and concentrations; however, their potential effects on membrane performance and sustainability is not yet clear. The effects of produced water and organic compound exposure on the properties of pressure driven membranes (e.g., water permeability, contaminant rejection, and membrane surface characteristics) has only been briefly investigated [7,22]. Polyamide thin-film composite (TFC) membranes exhibited notable changes in membrane performance and physiochemical properties, while no significant changes were reported for a traditional cellulose triacetate (CTA) RO membrane. Several studies have explored the application of osmotically driven membranes (i.e., forward osmosis (FO)) for similar feed waters [13,23–26]; however, none have specifically investigated the changes in membrane performance and active layer surface properties.

## 1.2. Impacts of produced water treatment on FO membrane performance

The impacts of FO membrane selection and system operating conditions on fouling propensity and overall membrane

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**Fig. 1.** Percent change in SRSF ( $\Delta J_S/J_W$ ) of CTA, TFC1, and TFC2 membrane coupons exposed to produced water (48 h) and after (a) osmotic backwashing (30 min) and (b) chemical cleaning (30 min). The percent change in SRSF is relative to virgin membrane performance before exposure to produced water. Data adopted from [9].

performance were recently investigated during our produced water treatment study [9]. Water flux, contaminant rejection, and chemical cleaning were evaluated for a CTA membrane and two TFC membranes. One TFC membrane has a traditional polyamide surface chemistry, while the other was one of the first surfacemodified polyamide FO membranes to be reported. Coday et al. did not focus specifically on identifying changes in membrane surface properties or permeability after exposure to produced water; however, membrane surface characterization after each experiment did suggest a shift in each membrane's physiochemical properties, especially the polyamide TFC membranes. Changes in membrane surface properties were supported by membrane integrity tests, during which variations in specific reverse salt flux (SRSF)—the ratio of RSF  $(I_s)$  of draw solution (DS) solutes to water flux  $(J_w)$  from the feed to the DS-were monitored (Fig. 1). The SRSF of the CTA membrane only minimally changed after treatment of produced water, while significant and equally unique trends were observed for both polyamide TFC membranes (Fig. 1a). SRSF increased on average by 74% for TFC1 and decreased by 39% for TFC2. While irreversible fouling and cake enhanced concentration polarization (CECP) might have biased these changes in membrane performance (as suggested by the relatively large standard deviation shown for TFC1), integrity tests conducted after chemical cleaning (Fig. 1b) revealed that changes in membrane performance could possibly be a direct result of chemically or physically induced changes to the membrane. No direct correlation could be established between system operating conditions and the observed changes in membrane performance after exposure to produced water.

#### 1.3. Objectives

The main objective of this study was to investigate the impacts of exposure to produced water on the transport and physiochemical properties of FO membranes. This study was not meant to supplement the fouling performance and data previously presented for produced water treatment [9], but to elucidate the impact of oil and gas wastewater on the short-term chemical stability of FO membranes. Of special interest was the performance and sustainability of a TFC membrane coated with a proprietary hydrogel to enhance its physical robustness and antifouling properties. A set of bench-scale experiments were conducted using raw produced water feed and three FO membranes, each exhibiting unique surface properties and polymeric chemistries. Each membrane was exposed to produced water for an extended period of time and then thoroughly cleaned prior to membrane autopsy. Integrity testing water flux (deionized water feed), RSF, and membrane surface properties were evaluated before and after exposure to produced water to highlight changes in membrane performance and permeability. An FO transport and structural parameter model [20] and containment rejection data collected during membrane exposure tests support the findings of this investigation. The results of this study can help guide future investigations on the performance and sustainability of polymeric membranes for produced water treatment and highlight the need for more rigorous membrane performance testing after fouling studies at the bench-scale.

#### 2. Materials and methods

#### 2.1. FO membranes

One asymmetric CTA membrane and two polyamide TFC membranes were investigated (Hydration Technology Innovations (HTI), Albany, OR). The first TFC membrane (designated TFC1) is a traditional polyamide membrane with no surface modification, while the active layer of the second TFC membrane (designated TFC2) was modified by HTI with a proprietary hydrogel to enhance its antifouling properties. The support layer of both TFC membranes are reported to be made of polysulfone. Membrane coupons were soaked in deionized water for 24 h prior to all experiments and then installed in each test cell with their active layer facing the feed. The water and solute permeability coefficients (A and B, respectively) and the modeled structural parameter (S) of the three membranes were determined using methodologies developed by Tiraferri et al. [20]. The A, B, and S coefficients for each membrane were similar to those reported in our previous study [9], which employed membranes from the same casting. All coefficients of determination  $(R^2)$  and coefficients of variation (CV) supporting the validity of the modeled results are summarized in Table 1.

#### Table 1

Coefficients of determination ( $R^2$ ) and coefficients of variation (CV) for the three FO membranes investigated in this study. CV is the coefficient of variation of  $J_W/J_{S}$ . CV is defined as the standard deviation divided by the arithmetic mean.

Regression ana-	CTA	CTA	TFC1	TFC1	TFC2	TFC2
lysis parameter	virgin	PW	virgin	PW	virgin	PW
$R^2 - J_W$ $R^2 - J_S$ $CV$	0.995	0.991	0.995	0.975	0.975	0.980
	0.989	0.990	0.993	0.979	0.987	0.983
	1.95	3.50	1.92	8.75	10.46	9.54

#### 2.2. Bench-scale FO system

The bench-scale system and custom-made membrane test cell (194 cm<sup>2</sup> active area) used in this investigation are the same as those used in our previous study [9] and have been thoroughly described elsewhere [27]. Three layers of a commercially available tricot spacer were used in the DS flow channel in all experiments to provide mechanical support for the membranes and to minimize physical stress on the membrane at the internal edges of the flow channel. LabView data acquisition software (National Instruments, Austin, TX) coupled with UE-9 Pro DAQ hardware (LabJack, Lakewood, CO) were used to control experimental test conditions and to log experimental data.

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