



## A systematic evaluation on the performance and mechanism of surfactants as additive of draw solution in forward osmosis



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### ARTICLE INFO

#### Keywords:

Surfactant  
Forward osmosis  
Micelle  
Hydrophile lipophilic balance  
Electrostatic repulsion

### ABSTRACT

In this study, the performances of six different types of surfactants, each with a series of concentrations around its critical micelle concentration, were systematically tested as additives of draw solution (DS) to improve the performance of DS in forward osmosis. The influence mechanism of surfactants on DS characteristics, membrane fouling and characteristics of ideal surfactants were discussed comprehensively. The six surfactants showed big differences in considering their effects on the characteristics of DS and their performances, which could be explained by the combined effects of thin dense layer formed by surfactant molecules adsorbed on the surface of supporting layer (SL), blocked pores of SL by micelles aggregation, increased viscosity of DS, improved hydrophilicity of membrane SL and ionization of the hydrophilic groups. The nonionic surfactants could significantly reduce the reverse solute flux, accompanied by sharply decreased water flux and heavy membrane fouling. Compared with nonionic surfactants, the anionic surfactants with their Hydrophile-Lipophilic Balance around 10 could significantly reduce the reverse solute flux while with no or low decrease of water flux and reversible membrane fouling. In our work, Sodium dodecyl benzene sulfonate, at 5 mmol/L concentration, had the best performance on the improvement of DS.

### 1. Introduction

As an emerging technology for membrane separation, forward osmosis (FO) has attracted increasing attention from researchers and practitioners since it has the merits of low energy consumption, high resistance to membrane fouling and high water recovery [1]. Through decades of development, the mechanism of FO has become more explicit [2]. There even have some valuable pilot application tests in different fields [3–5], but no real large scale application yet. On the one hand, solute reverse osmosis from draw solution (DS) to feed solution (FS) can increase the osmotic pressure of FS, which can decrease the effective transmembrane pressure. On the other hand, solute reverse osmosis can aggravate the contamination of the FS. For example, some inorganic substances with polyvalent ions (such as  $Mg^{2+}$ ,  $Ca^{2+}$ ,  $Cu^{2+}$ , etc.) and organic substances (such as EDTA sodium, 2-methylimidazole-based compounds, etc.) can make the feed solution more complex through solute reverse osmosis [6,7]. Therefore, solute reverse osmosis is one of the major obstacles limiting the applications of FO, especially in the fields of water purification, FO-membrane bioreactor and liquid foods concentration.

To overcome the disadvantage mentioned above, a lot of efforts have been put to exploiting ideal solutes in DS with low reverse

osmosis, such as inorganic substances with polyvalent ions, macromolecular organic compounds, hydrophilic magnetic nanoparticles, etc. [7–9]. Unfortunately, inorganic substances with polyvalent ions ( $Mg^{2+}$ ,  $Ca^{2+}$ ,  $Cu^{2+}$ , etc.), which have lower reverse osmosis than monovalent inorganic substances, may generate insoluble carbonates with  $CO_3^{2-}$  from FS, resulting in aggravated membrane fouling [10]. Although macromolecular organic matter has low solute reverse osmosis and can even be re-concentrated by ultrafiltration, their osmotic pressure is generally low, accompanied with heavy organic membrane fouling [11]. For new hydrophilic magnetic nanoparticles, they can be re-concentrated easily through magnetic field and have low solute reverse osmosis, but the magnetic nanoparticles tend to aggregate in DS and reduce their function [12]. Overall, there are still various shortcomings need to be made up in the application of multifarious DS.

In recent years, different types of surfactants as solutes or additives of DS, come into researchers' vision and have attracted much attention [13–16]. Compared with the classic DS NaCl (reverse solute flux is on the order of  $10^{-5} \text{ mol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ), the reverse solute fluxes of many surfactants are quite low, which are on the order of  $10^{-7} \text{ mol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  [13]. Nguyen and Chen [14] reported that the surfactant Triton X100 coupling  $Na_3PO_4$  or EDTA sodium salt used as DS could decrease solute reverse osmosis effectively. Besides, some surfactants could easily be re-

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concentrated by nanofiltration system, even by ultrafiltration system. With the deepening of studies, more publications have focused on the effect mechanism of surfactants on the performance of DS. Nguyen and Chen thought that the main mechanism was that the interaction between hydrophobic tails of surfactants and hydrophobic membrane created a layer of surfactants on the membrane surface, which constricted the membrane pores.

Nevertheless, the appropriate addition of surfactants could not only change the physical and chemical properties of DS (such as pH, viscosity, etc.), but also change the membrane characteristics, such as membrane hydrophilicity, fouling tendency, etc. [17]. Therefore, with the addition of different surfactants, the process of FO would become more complex and be affected in different ways and with different mechanism. To our best knowledge, although it has been reported that ideal surfactant for DS should have characteristics of low critical micelle concentration (CMC) and long chain [13], there is still lack of comprehensive understanding on the mechanism and clear clues for the selection of ideal surfactants for DS. Besides, there is rare research related to the membrane fouling of different surfactants in FO.

Accordingly, in the present work, we chose six different surfactants according to the presence/absence of charged groups in the hydrophilic head as additives of DS to study the mechanism of different surfactants on the performance of DS in FO. In addition, the membrane fouling and characteristics of ideal surfactants were also discussed comprehensively.

## 2. Materials and experimental set-up

### 2.1. FO membrane

An early generation thin-film-composite (TFC) FO membrane (Hydration Technology Innovations, HTI, Albany, Oregon, USA) was used in the experiment. The characteristics of the membrane were shown in Table 1. The TFC membrane was asymmetric and comprised of an active layer (AL) and a porous support layer (SL) embedded non-woven polyester mesh.

The TFC membrane was pre-wetted through being soaked in a 25 wt % solution of isopropyl alcohol for 10 min at room temperature, followed by washing thoroughly using DI water [18]. Then, the pre-wetted membrane was stored in DI water (with a conductivity of 18.2 MΩ·cm, Millipore Integral) at 4 °C before experiment. Each piece of membrane was tested for its initial water flux and solute reverse osmosis flux before it was put into experiment. In all parallel experiment, three membranes with their initial water flux and reverse solute flux difference less than 5% were chosen.

### 2.2. Characteristics of surfactants, DS and FS

The DS used in the experiment was 0.6 M NaCl solution coupling with different surfactants. NaCl has fairly stable Physico-Chemical property that would not react with the surfactants. Different types of surfactants, each with a series of concentrations were tested as the additives of DS. The concentrations of all surfactants were chosen depending on their CMC (shown in Table 2), but all were at low level, less than 25 mmol/L. Deionized water with neglected osmotic pressure

**Table 1**  
Characteristics of FO membrane.

Parameter	Symbol	Units	Value
HTI-TFC membrane thickness	$\delta$	$\mu\text{m}$	$114 \pm 3$
Contact angle of membrane AL	$\theta_{AL}$	$^\circ$	$48.6 \pm 2.6$
Contact angle of membrane SL	$\theta_{SL}$	$^\circ$	$84.4 \pm 3.6$
Membrane area	$S_m$	$\text{cm}^2$	50
Pure water permeability	$A$	$\text{L m}^{-2} \text{h}^{-1} \text{bar}^{-1}$	$1.75 \pm 0.13$
Salt permeability	$B$	$\text{L m}^{-2} \text{h}^{-1}$	$1.25 \pm 0.30$

**Table 2**  
Characteristics of the tested surfactants.

Surfactant	Type	Abbr.	Mol. Wt.	HLB	CMC (mM)
Oleic acid	Anionics	OA	282.47	1.0	0.004 [19] (17 °C)
Sodium dodecyl benzene sulfonate	Anionics	SDBS	348.48	10.6	2.76 [20] (25 °C)
Potassium oleate	Anionics	PO	320.55	20.0	0.9 [21] (25 °C)
Sodium dodecyl sulfate	Anionics	SDS	288.38	40.0	8.16 [22] (30 °C)
Polyoxyethylene lauryl ether	Nonionics	Brij35	1199.55	9.5	0.06 [23] (25 °C)
Polyethylene glycol <i>tert</i> -octylphenyl ether	Nonionics	Triton X-100	646.85	13.5	0.27 [20] (25 °C)

Note: theoretically, HLB = 10 was the turning point between hydrophilicity and hydrophobicity.

was used as FS in all experiments.

In general, surfactants are classified into four categories according to the presence/absence of charged groups in the hydrophilic head, which are anionics, cationics, amphoteric and nonionics, and are used for different purposes. According to their Hydrophile-Lipophilic Balance (HLB) value, they could be used as defoaming agent with HLB = 1–3, as W/O type emulsifier with HLB = 3–6, as wetting agent with HLB = 7–9, as O/W type emulsifier with HLB = 8–18, as detergent with HLB = 13–15, as solubilizer with HLB = 15–18. In our study, four anionic surfactants with increased HLB were chosen to test the effect of surfactant hydrophilicity on the performance of DS. And two regular nonionic surfactants were tested to figure out their performance compared with the anionics. Cationic surfactants were not chosen as they could damage the membrane through charge neutralization and make the membrane more hydrophobic through electrostatic attraction [24]. Amphoteric surfactants were also not considered in the experiment because they showed the characteristics of the anionics or cationics depending on pH. The characteristics of tested surfactants were shown in Tables 2 and 3.

### 2.3. Lab-scale test unit

A schematic of the laboratory scale FO system was shown in Fig. 1. The membrane was set in the middle of the membrane cell with its active layer faced FS (AL-FS) in all experiments. The dimension of the chamber on each side of the cell was 100 mm × 50 mm × 5 mm. Two variable speed gear pumps were used for the circulation of DS and FS at constant cross-flow velocities of 42.5 cm/s. The temperature of FO system was controlled at  $30 \pm 0.1$  °C by using a thermostat water bath. NaCl solution of 2 mol/L was added into DS intermittently to maintain its constant concentration. The pH of DS was measured by a pH meter (SG3-ELK, Mettler-Toledo, Switzerland). The weight change of FS was measured with a digital balance (PL2002-IC, Mettler-Toledo, Switzerland) connected to a computer data logging system to calculate water flux. The conductivity change of FS was measured by using a conductivity meter (SG3-ELK, Mettler-Toledo, Switzerland) to calculate reverse solute flux. Each test had three parallel samples. For each parallel sample, the data was collected in two different periods after the 30 min of system operation. After each FO run, the membrane was cleaned with DI water (30 °C) for 30 min at a higher cross-flow velocity of 74.3 cm/s.

The water flux ( $J_w$  (LMH)) across the FO membrane was calculated by Eq. (1) [28]:

$$J_w = \frac{\rho_w \Delta m}{S_m \Delta t} \quad (1)$$

where  $\rho_w$  was the density of FS,  $\Delta m$  was the weight change of FS over a predetermined time  $\Delta t$ , and  $S_m$  was the effective area of the membrane.

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