



The role of nanofiltration membrane surface charge on the scale-prone ions concentration polarization for low or medium saline water softening

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ABSTRACT

Based on the fact that when inlet bulk solution with low or medium concentration was involved, scaling was apt to appear on nanofiltration (NF) membrane surface in retentate side. Therefore, study on its surface charge performance under threshold concentration condition is necessary. Two polyamide composite NF membranes (DL and DK) were characterized with streaming potential (E_s), zeta (ξ) potential and other measurements encountering with a wide ionic strength range ($1.5\text{--}108\text{ mol}\cdot\text{m}^{-3}$). Then, the interactional relations between membrane surface charge and concentration polarization (CP) degree, along with the relevant constituting factors were investigated. The results showed that even when the sparingly soluble salt concentration for CaSO_4 exceeded its threshold point, both NF membranes at pressure drop of 180 kPa still exhibited low and medium-intensity surface electrokinetic phenomena ($-5.94 \leq E_s \leq -3.41\text{ mV}$; $-38.82 \leq \xi\text{-potential} \leq -36.34\text{ mV}$). It turned out that solvent flux decreased almost linearly, but measured concentrations of the scalant ions in NF retentate streams increased slightly at first and then rapidly with the enlargement of ξ -potential, respectively. Meanwhile, mass transfer coefficient and boundary layer thickness had a negligible change in the scope of testing ξ -potential. Therefore, $CP_{\text{SO}_4^{2-}}$ and $CP_{\text{Ca}^{2+}}$ decreased apparently with ξ -potential becoming less negative for the adopted NF membranes.

1. Introduction

With rapid development of membrane technology, nanofiltration (NF) membrane shows interesting selectivity towards charged solutes, allowing the separation of plentiful ions in accordance with their electrical charge and size. Thus, NF technology is widely employed in partial demineralization or softening of brackish water, seawater and other brine waters [1, 2]. Nevertheless, surface fouling is still a widespread issue that might impede successful applications of NF membranes [3]. Owing to more extensive pretreatment systems including conventional and membrane pretreatment technologies are adopted, much of the undissolved suspended particulate matters, dissolved macromolecular contaminants and biogenic substance are rejected efficiently. So sparing soluble inorganic scales and precipitation are usually regarded as one of the most problematic fouling and bottlenecks encountered during NF softening processes [4]. The phenomena of the limiting or threshold concentration level below which there is no risk of membrane surface precipitation with a given membrane and treated water are much understood [5]. Despite all this, in order to come out with an effective scaling control strategy, the underlying mechanism

and relevant factors that contribute to NF membrane scaling phenomena should be clarified.

Generally speaking, one of the key factors with respect to membrane surface scaling during NF softening process must be considered is concentration polarization (CP) effect. Because CP dramatically decreases the mass transfer coefficient (k_i) of rejected ions, and hence causes the accumulation of them at NF membrane surface [6, 7]. So the dissolved salts at NF membrane surface easily become supersaturated, even if the bulk concentration still remains unsaturated, as a result of pressure driven membrane processes. After considering the ions concentration profiles at the membrane surface, the film theory thought CP effect was the key element for research on scaling potential prediction and precipitation in NF membrane systems [8]. Previously, reports from Bader showed that for oil-fields water injection experiment was operated at a throughout ratio of 75%, due to CP effect, the concentration of rejected SO_4^{2-} ion in bulk solution (C_b) with $25.63\text{ mol}\cdot\text{m}^{-3}$ concentrated to $100.63\text{ mol}\cdot\text{m}^{-3}$ in retentate stream (C_r) and accumulated to $181.83\text{ mol}\cdot\text{m}^{-3}$ at the NF270 membrane surface (C_m), respectively [9]. Meanwhile, based on the fact that high permeate recovery and remarkable rejection rates for scale-prone ions are often synergistically

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Nomenclature			
$c_{b,i}$	Concentration of solute i in the bulk solution, $\text{mol}\cdot\text{m}^{-3}$	k_i	Mass transfer coefficient, $\text{m}\cdot\text{s}^{-1}$
$c_{m,i}$	Concentration of solute i at the membrane surface, $\text{mol}\cdot\text{m}^{-3}$	l, L	The channel length, m
$c_{p,i}$	Concentration of solute i in the permeate, $\text{mol}\cdot\text{m}^{-3}$	N_A	Avogadro constant, mol^{-1}
CP_i	Concentration polarization module of solute i , –	T	Thermodynamic temperature, K
d_h	Hydraulic diameter, m	V	Cross-flow velocity of feed, $\text{m}\cdot\text{s}^{-1}$
D_i	Diffusion coefficient of solute i , $\text{m}^2\cdot\text{s}^{-1}$	w	The channel width, m
e	Elementary charge, C	z_i	The valence of ion i , $\text{eq}\cdot\text{mol}^{-1}$
E_s	Streaming potential, V	δ	Boundary layer thickness, m
G_t	Total conductance of the system, Ω^{-1}	ΔP	Flow pressure drops, MPa
h	The channel height, m	ϵ_0	The vacuum permittivity, $\text{F}\cdot\text{m}^{-1}$
J	Permeate flux, $\text{m}\cdot\text{s}^{-1}$	ϵ_r	Relative dielectric constant of the solvent, –
k_B	Boltzmann constant, $\text{J}\cdot\text{K}^{-1}$	ξ	Zeta potential, V
		η	Liquid viscosity, $\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$
		ν	Kinematic viscosity of feed, $\text{m}^2\cdot\text{s}^{-1}$
		σ	Membrane surface charge density, $\text{C}\cdot\text{m}^{-2}$

occurred, concentration factors for these ions are generally between 1 and 10 during low- or medium-salinity water softening processes by NF membranes [10–12]. In conclusion, due to the coupling effects of CP and usually associated concentration factor, even though in low- or medium-salinity water ($\leq 100 \text{ mol}\cdot\text{m}^{-3}$) softening processes, scaling was apt to appeared on NF membrane surface in retentate side.

Owing to the dissociation of ionisable groups at the membrane surface or adsorption of charge solute, NF membranes in contact with electrolyte solution are typically charged. These charged behaviors will influence the spatial distributions and concentrations of dissolved solutes at the membrane-solution interface, resulting in the variation of separating performance, the rate of back diffusion and accumulation near the membrane surface, therefore CP phenomenon occurred in the vicinity of NF membrane surface is affected [13, 14]. During the past decades, in a similar low- or medium-concentration of inlet bulk electrolyte solution, many studies have been fulfilled on the research of membrane surface charge properties for saline water softening, mainly focusing on the charge properties measurements, formation mechanism and the role of surface charge on NF membrane separating performance [15–17]. Ma et al. [18, 19] adopted NTR-7450 membrane to explore the mechanism of charge formation in the case of KCl , K_2SO_4 and K_3PO_4 electrolyte solution, and found that under the condition of low ionic strength ($0.1\text{--}10 \text{ mol}\cdot\text{m}^{-3}$), the membrane volume charge density could be correlated with the feed concentration in terms of a Freundlich adsorption isotherm. Whereas in medium or high electrolyte concentration ($10\text{--}200 \text{ mol}\cdot\text{m}^{-3}$), with the increase of salinity, the absolute value of streaming potential (E_s) increased gradually while both zeta (ξ) potential and membrane surface charge density (σ) first increased and then decreased as well as finally tended to be stable, and peaked at $100 \text{ mol}\cdot\text{m}^{-3}$. Du et al. [20, 21] demonstrated that in $1.0\text{--}10 \text{ mol}\cdot\text{m}^{-3}$ electrolyte solutions, as the electrolyte concentration increased, the concentrations of countra-ions far away from NF membrane surface would begin approaching that of adsorbed ions at the membrane surface, resulting in a decrease in Donnan potential difference. Adsorbed countra-ions caused the driving force degradation, which led to the decrease of the absolute values of E_s and ξ -potential [22]. In the process of σ declining, the repulsive interaction between NF membrane and scale-prone ions also decreased, which benefited the aggravation of membrane surface scaling potential. Nevertheless, until recently, very few investigations dedicated to the influence of surface charge

performance on CP behavior occurring in the vicinity of NF membrane skin-layer.

Based on the fact that in this process when inlet bulk solution with low or medium concentration was involved, scaling was apt to appear on NF membrane surface in brine side. In this context, it seems necessary to illuminate the interaction mechanism between NF membrane surface charge performance and the scale-prone ions' concentration polarization extent. First, setting the representative non-alkaline sparingly soluble salt (CaSO_4) as the potential pollutant, a series of NF membrane charge performance under threshold concentration condition was evaluated. Then, membrane surface charge impacts on the solvent flux, the scale-prone ions concentrations in NF retentate and mass transfer coefficient as well as boundary layer thickness were investigated. In the end, the relationships between surface charge behavior and CP for the candidate NF modules were analyzed comprehensively. The results will provide supplementary theoretical basis from a charge performance standpoint for NF membrane surface scaling potential prediction and taking effective anti-scaling strategies in the saline water softening process.

2. Material and methods

2.1. Electrolyte solution and membranes

In this research, a series of two-component ternary electrolyte solutions ($\text{K}_2\text{SO}_4 + \text{CaCl}_2$) with the molar ratio of 1:1 and a representative non-alkaline sparingly soluble salt precipitation (CaSO_4) were involved as the study carrier and subject, respectively. The related electrolyte solution samples were comprised of analytical grade salts dissolved in deionized water ($5 \mu\text{s}\cdot\text{cm}^{-1}$). Two commercial available NF membranes with excellent selectivity named DL (termed as NF1) and DK (termed as NF2), were adopted in this study [23]. Characteristics of the selected virgin NF membranes are displayed in Table 1. It could be found that the molecular weight cut-off (MWCO), the effective pore radius and contact angle for the aforementioned membranes are 250–400 and 150–300 Da, 0.70 and 0.65 nm, 51° and 45.1° , respectively [18, 23, 24].

Table 1
Characteristics of the selected virgin NF membranes.

Type of membrane	MWCO (Da)	RMS (nm)	Pore size (nm) ^a	Contact angle ($^\circ$)	Pure water flux ($\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{MPa}^{-1}$)	Ion selectivity (–)
DK	150–300	10.9	0.65	45.1	37.51	2.10–3.28
DL	250–400	14.6	0.70	51	39.98	4.63–5.04

^a Data provided by manufacturer.

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