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Analysis of nanofiltration membrane performance during softening process of simulated brackish groundwater



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HIGHLIGHTS

- NF membrane performance was analyzed during brackish water softening process.
- Operating conditions effect on NF softening efficiency were investigated.
- pH variations in different streams and the underlying causes were discussed.
- High softening efficiency for the specific NF membrane was verified.

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ABSTRACT

In this paper, one brackish water quality, which typically represented the water quality in the Huanghuai region of China, was simulated and used as feedwater to run an ultrafiltration (UF)–nanofiltration (NF) integrated membrane system. Laboratory-scale experiments were carried out to evaluate NF softening performance. The influences of transmembrane pressure (0.6–2.2 MPa), inlet tangential flow velocity (0.087–0.384 m·s⁻¹) and feedwater temperature (7–35 °C) on the softening efficiencies of two NF membranes denoted as DK (termed as NF1) and DL (termed as NF2) were investigated. The experimental results revealed that the calculated total hardness and the bivalent ion (SO $_4^2$ –, CO $_3^2$ –, Mg $_2^2$ + and Ca $_2^2$ +) concentrations in NF2 product water on the same operation were all slightly lower than those in NF1 permeate in two schemes. Additionally, pH value of NF permeate decreased prominently than those in feedwater with increasing of transmembrane pressure, inlet tangential flow velocity or decreasing of feedwater temperature within the testing scope.

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1. Introduction

Traditional freshwater resources are gradually dwindling, and groundwater resources are becoming increasingly saline and not suitable for drinking directly [1,2]. In many countries and regions in the world, most of natural brackish water resources are intrinsically untapped water resources [3]. Desalination of brackish water has become the choice of a key water treatment technology. It is reported that treated brackish water presently accounts for 22%, seawater for 58% and wastewater for 5% of the water treated by desalination technologies [4].

Brackish water usually has a relatively lower TDS value, but the proportions of sulfate/TDS, calcium/TDS and carbonate/TDS are much larger in brackish water than in seawater [5]. Overall, brackish water

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sources are often groundwaters, these groundwaters can be naturally filtrated through porous medium (stone, clay, sand, etc.), and most of the pollutants mentioned earlier representatively exist in surface waters are rejected. It leads to brackish water reverse osmosis (BWRO) plants usually might be fouled by sparingly soluble inorganic scale and precipitation [6].

Owing to its own intrinsic advantages, e.g., higher permeate fluxes and rejection rates for scale-prone bivalent ions, lower operating pressure and investment, nanofiltration (NF) membranes are increasingly applied in the water treatment field such as brackish water and seawater softening processes, wastewater reuse and sewage treatment [7]. NF technology has shown great potential to reduce RO membrane scaling as well as the running costs of BWRO plant due to its remarkable ability to remove sale-forming ions in the water [8]. NF was also proved to be able to cut down the total hardness in the influent solution and thus the following RO feed osmotic pressure could be decreased, which allowed the system product recovery to be increased [9].

During the past decades, many studies have been fulfilled on the utilization of NF membrane for brackish water softening. Sarkar et al. [10] applied NF (SRNF) membrane to treat a brackish water of 8260 $\rm mg \cdot L^{-1}$ TDS with 60% product recovery rate and found that hydraulic permeability of NF membrane reached up to 9.18 $\rm L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$. AlTaee [11] adopt ROSA software to check out the feasibility of the NF-BWRO integrated membrane desalination process. The experimental data showed that under feedwater temperature of 25 °C and feed TDS of 4611 $\rm mg \cdot L^{-1}$, when recovery rate increased from 50% to 80%, permeate concentration for NF90 membrane tended to increase from 586 $\rm mg \cdot L^{-1}$ to 712 $\rm mg \cdot L^{-1}$. The researches mentioned above have proved that permeate water after NF softening treatment served as brackish water desalination pretreatment technology and has high quality for extensive and profound applications.

In this research, the alternative treatment processes were involved two integrated membrane systems with different NF membranes and a tight UF membrane (with molecular weight cut-off (MWCO) of 20 kDa). Performance for brackish water softening was evaluated under different operating conditions (including transmembrane pressure, inlet tangential flow velocity and feedwater temperature). Also, the variations of stream pH before and after NF membrane separation within the testing scope and the underlying causes were investigated. The experimental results gained in this study in order to find the optimal operating conditions that are particular feedwater and membrane. This study is beneficial to enrich related fundamental information, and further understand a series of reliable prediction membrane fouling in RWRO.

2. Material and methods

2.1. Model solution and membranes

Feedwater solution in separating performance experiments was comprised of reagent grade salts dissolved in laboratory deionized water. The specific electrolyte composition used in this study was designed to represent the major ions (>1 ppm) of the underground brackish water, which is mainly distributed in Huanghuai region along the junction of the north China piedmont alluvial plain and central alluvial plain. In addition, humic acid (HA) was used as a representative NOM in this study. The main parameters of the simulated brackish water are presented in Table 1.

Commercial polyether sulfone hollow fibers UF membrane is supplied by Lanlv Co. Ltd. (MWCO 20 kDa, inner/outer diameter = 0.9/1.3 mm). Based on the excellent membrane selectivity of bivalent ion over monovalent ion according to our prior work [7], commercially available polyamide composite NF membranes denoted as DK (termed as NF1) and DL (termed as NF2) are used in this study. The specifications of UF and NF membranes are summarized in Table 2 [12,13].

2.2. Laboratory-scale experimental set-up

The flow chart of this laboratory scale UF–NF integrated membrane system was shown in Fig. 1. Model brackish water was firstly pumped to UF system by diving pressure, and the UF product water was

Table 1
Water quality analysis of simulated brackish groundwater in Huanghuai region.

Parameters	Values	Parameters	Values
Conductivity (µS·cm ⁻¹) pH TDS (mg·L ⁻¹) Total hardness (mg·L ⁻¹) TOtal alkalinity (mg·L ⁻¹) TOC (mg·L ⁻¹) HCO ₃ (mg·L ⁻¹)	$1819-2290$ 7.55 ± 0.08 $907-1144$ $281-327$ $201-279$ $2.6-3.5$ $147-172$	K ⁺ (mg·L ⁻¹) Na ⁺ (mg·L ⁻¹) Ca ²⁺ (mg·L ⁻¹) Mg ²⁺ (mg·L ⁻¹) Cl ⁻ (mg·L ⁻¹) SO ₄ ²⁻ (mg·L ⁻¹) NO ₃ (mg·L ⁻¹)	38.2-44.6 251-289 61.8-71.6 26.9-31.8 394-455 126-145 61.6-69.8

Note: The total hardness and total alkalinity according to CaCO₃ terms.

Table 2Characteristics of the selected membrane modules.

Items	UF	NF1 [12]	NF2 [13]
Manufacturer	Lanlv (China)	GE	GE
Module type	HF-1500	DK	DL
Module configuration	Hollow fiber	Flat-sheet	Flat-sheet
Membrane materials	Polysulfone	Polyamide	Polyamide
Molecular weight cut-off (Da)	20,000	150-300	250-400
Membrane effective area (m ²)	0.533	1.13×10^{-2}	1.13×10^{-2}
Pore size (nm)	10	1.0	1.0
Salt rejection (%)	≈ 0	>96%ª	>96% ^a
Range of feed temperature (°C)	0-50	0-45	0-45
Range of feed pH	2-11	2-11.5	1-11
Range of operating pressure (MPa)	0-0.2	0-4.0	0-4.0

 $[^]a$ Testing parameters: 0.048 MPa, 25 °C, 2000 mg \cdot L $^{-1}$ MgSO $_4$ solution, recovery rate 15%

continued to pump into NF unit to gain NF softening permeate water. All brackish water NF softening experiments were carried out using a custom fabricated laboratory-scale tangential flow NF membrane evaluating device. Four flat-sheet circular membrane vessels were designed with single membrane area of 28.26 cm² (with an inner diameter of 3 cm) each, and a channel height of 2.5 mm.

2.3. Experiment procedures

In the UF–NF process, UF was operated at a roughly constant transmembrane pressure of 0.07–0.09 MPa and a fixed filtrate flux of $20 \text{ L} \cdot \text{h}^{-1}$ with product water recovery of 95%.

The NF experiments were performed in a closed-loop mode. The orthogonal experiments were fulfilled based on single-factor tests, which were employed in three tasks to investigate the influencing of transmembrane pressure, inlet tangential flow velocity and feedwater temperature on NF1 and NF2 softening performance with UF pretreatment, respectively. The surveying ranges of transmembrane pressure, inlet tangential flow velocity and feedwater temperature were 0.6–2.2 MPa, 0.087–0.384 m \cdot s $^{-1}$ and 7–35 °C. Each group of filtration experiment lasted 2 h in order to achieve the equilibrium state.

2.4. Analytical methods

Conductivity and TDS of the water samples were measured using a digital conductivity meter (DDS-11A, Shanghai Leici Instrument Company, China); concentration of the main cations (Ca^{2+} , Mg^{2+} , Na^{+} and K^{+}) was analyzed using an inductively coupled plasma (ICP) spectrometer (ELAN DRC-e, America); concentration of the anions (CI^{-} , SO_4^{2-} and NO_3^{-}) was measured using an ion chromatography (DIONEX-90, America); concentration of HCO_3^{-} was obtained by the charge balance in the electrolyte solution, and concentration of CO_3^{2-} was calculated by the dissociation equilibrium between CO_3^{2-} and HCO_3^{-} , which has been described everywhere [14]. pH was measured in real time after each sampling by a pH meter (PHS-3C, China); TOC was analyzed using a TOC-5000A analyzer (Shimadzu, Japan).

3. Results and discussion

3.1. Laboratory test of UF pretreatment

The average TOC in the simulated brackish groundwater samples and UF filtrate samples was 2.75 \pm 0.13 and 1.12 \pm 0.07 $mg\cdot L^{-1}$ and the calculated average removal rate of the UF module was about 58.5%. It indicated that the candidate UF membrane could produce filtration with good and stable quality based on the molecular-sieving mechanism.

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