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Pilot study of seawater nanofiltration softening technology based on integrated membrane system☆

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HIGHLIGHTS

- A 100 $m^3 \cdot d^{-1}$ pilot-scale UF–NF integrated membrane system was established.
- Operating factors have little effect on overall NF permeate quality.
- The dual-stage NF system has excellent DOC and ion rejection during the long run.
- The dual-stage NF system had excellent anti-fouling property during the long run.
- Energy consumption of the NF process is about 1.8 $kWh \cdot m^{-3}$ at 25 °C and 3.5 MPa.

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ABSTRACT

A pilot-scale ultrafiltration (UF)–nanofiltration (NF) integrated membrane system (IMS) which included a self-cleaning crossflow UF filtration process and a dual-stage NF process with a capacity of 100 $m^3 \cdot d^{-1}$ NF permeation water was established for seawater softening investigation. The separation performance of the dual-stage NF process under different conditions, such as operating pressure, recovery rate, and inlet flowrate, was extensively investigated; long-term performance and energy consumption of the dual-stage NF process were analyzed as well. The results showed that during the long-term operation, UF could provide qualified filtrate for NF. The dual-stage NF process achieved high separation performance with good permeate quality, especially high rejection of Ca^{2+} and Mg^{2+} (>90%). In addition, the dual-stage NF process showed good anti-fouling characteristics, with the normalized NF permeate flux maintaining at an essential constant value of about 14 $L \cdot m^{-2} \cdot h^{-1}$ at 3.5 MPa, and DOC rejection around 90% during the long-term experiment. Energy consumption of the dual-stage NF process increased with the increase of the operating pressure and the decrease of the raw seawater temperature, which was about 1.8 $kWh \cdot m^{-3}$ at 3.5 MPa and 25 °C.

1. Introduction

Fresh water shortage problem has become a limiting factor of local economy development in many countries and regions for a long time, especially in drought and coastal areas. Therefore, it is an inevitable trend to desalinate seawater to provide fresh water for the domestic and industrial usage. So far, there are two main processes dominating the desalination market, thermal process and membrane-based process. Thermal desalination process does not require complicated pretreatment, but has high-energy consumption. Recently, there has been a growing emphasis on membrane-based seawater desalination technology, especially integrated membrane system (IMS) [1–3] that combines multiple classes of membranes with different characteristics. IMS can lower membrane fouling rates, energy consumption, and operating cost, resulting in a more stable operation [4]. Commonly, IMS uses microfiltration (MF) [5,6] or ultrafiltration (UF) [7–11] as a pretreatment technique to provide high quality filtrate for the following nanofiltration (NF) or seawater reverse osmosis (SWRO) process.

A suitable seawater pretreatment system should stably provide feed water with low turbidity and SDI (Silt Density Index) for NF or SWRO process. UF membranes could consistently remove particles, bacteria, virus, as well as colloidal matter, and they are more reliable in producing low fouling potential RO feed water than conventional pretreatment techniques even during harmful algal blooms event [12]. Nearly 100% of the UF filtrate turbidity was below 0.01 NTU and 95% of the filtrate SDI_{15} was below 3.0, which could satisfy the requirements of SWRO feed water completely. In addition, UF membranes showed excellent

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removal efficiency for high molecular weight organic matter, which could produce filtrate with content of dissolved organic matter (DOM) less than 2 mg·L^{-1} [7,13], thus it could increase the lifespan of the RO membrane by 20–30% [14]. Meanwhile, UF has larger capacity compared with conventional pretreatment techniques, and could save about 50% of the footprint [15]. UF pretreatment has been successfully implemented in several large (>100,000 $m^3·d^{-1}$) SWRO plants [12].

NF is an emerging pretreatment technique in the seawater desalination field since the late 1990s. NF membrane has unique characteristic of selectively separating divalent ions as well as multivalent ions from monovalent ions, making it capable of highly rejecting sulfate from seawater while retaining a moderate portion of monovalent ions therein, thus it can provide nearly sulfate-free seawater for SWRO desalination [16–18] and some other industrial applications such as offshore flooding. Meanwhile, with a molecular weight cut-off (MWCO) of 200–1000 Dalton (Da), NF membrane could typically reject organic matter with molecular weight greater than 200 Da [18].

During the past decades, NF had been used as a pretreatment process of SWRO and formed a novel IMS [1,17,19–24]. Saline Water Conversion Corporation (SWCC) is the pioneer in developing applications and operation of NF pretreatment for seawater desalination. Researchers in SWCC have done much efforts in the development of NF–SWRO IMS and hybrid system of NF–SWRO–MSF as well as NF–MSF [17,20,25–29]. They have carried out long-term operation of NF–SWRO system since 2000, and demonstrated that it is possible to operate NF at 65% recovery at pH = 6 utilizing only low feed pressure of <25 bar. This led to increase in SWRO production by 42%, without any chemical cleaning or membrane replacement required for SWRO membranes [30], and reduce the desalination cost by about 30% [31]. Drioli and coworkers demonstrated that the IMS of NF–RO with membrane distillation/crystallization units could even increase the water recovery rate up to 92.8% without significant increase of the costs [32].

Until now, most of the pilot scale investigations on NF seawater softening were mainly based on the performance of single NF element of 2.5 or 4.0 inch diameter, and there is rare investigations focusing on the performance of full-scale 8-inch diameter commercial NF elements except the SWCC [17]. In our previous pilot scale experiment [33–35], we investigated the performance of UF–NF IMS on softening seawater for SWRO desalination and for offshore water flooding. The pilot scale application including one 4-inch NF module and two UF modules, and the results we obtained were promising. However, it could not provide overall information about the seawater softening process, which is different from full-scale NF modules that could guide industrial production exactly. To our viewpoint, full-scale operation of 8-inch diameter commercial NF elements is much more valuable for the comprehensive insight into the separation performance of the NF modules, which could further foster the extensive application of NF seawater softening technique. Therefore, in this study, we established a full-scale 100 $m^3·d^{-1}$ UF–NF IMS composing ten 8-inch commercial membrane elements in the NF process. We conducted a pilot trial by using coastal seawater to investigate the separation performance and stability of the IMS, assess the membrane fouling tendency of the NF module, and provide operating parameters for the application of the IMS for surface seawater softening.

2. Materials and methods

2.1. Membrane modules

All the hollow fiber UF membrane modules and the 8-inch spiral-wound NF membrane modules used in the pilot test are commercially available. Their characteristics are shown in Table 1 according to the membrane manufactures.

Table 1
Characteristics of UF and NF membranes used in this pilot test.

Items	UF module	NF module
Module type	CREFLUK-PUF-8040	NF-400
Module configuration	Hollow fiber	Spiral wound
Separation layer material	PP	Polyamide
Molecular weight cut-off (Da)	50,000	100–500
Membrane effective area (m^2)	60	37
Pore size (nm)	100	≈1
Rejection rate (%)	≈0	85–95[a]
Highest inlet water temperature (°C)	40	45
pH range	2–13	2–11
Highest operating pressure (MPa)	0.25	4.1

[a] Testing parameters: 0.48 MPa, 25 °C, 500 mg·L^{-1} $CaCl_2$ solution, recovery rate 15%.

2.2. Experimental setup and procedures

The schematic diagram of the pilot-scale UF process was shown in Fig. 1(a). Seawater was firstly transferred by a feed water pump to the seawater tank. Then it was delivered across a self-cleaning filter to the UF modules by a low-pressure pump. The operation cycle of the UF system contained four steps, 30 s flushing with raw seawater, 20 min filtration, 15 s gas washing with pressured air, and 75 s backwashing with UF filtrate, respectively, which was controlled by a PLC system. The UF filtration process used a crossflow circulation manner, during which the retentate stream was completely recycled back to the UF feed water by a recycling pump, and no discharge was performed until the end of the filtration duration. During the test period, SDI_{15} and turbidity of the UF filtrate were measured periodically to evaluate the stability of the UF system.

Fig. 1(b) presented the schematic diagram of the dual-stage NF system. The 1st stage had two parallel pressure vessels, both of which contained four 8-inch commercial NF elements. The 2nd stage had only one pressure vessel which contained two 8-inch commercial NF elements. During the operation, the retentate of the 1st stage served as feed to the 2nd stage. The permeate of both stages was collected and mixed as product water. Performance of the NF membranes was evaluated within the operating pressure range of 2.0–4.0 MPa and the feed water temperature range of 23–17 °C in autumn.

The rejection, R, and the permeate flux, Jv, are calculated using Eqs. (1) and (2):

$$R = \frac{C_F - C_P}{C_F} \times 100\% \quad (1)$$

$$J_V = \frac{Q}{A} \quad (2)$$

where C_p and C_F are the average solute concentrations in the permeate and the feed side, respectively; Q is the flowrate of the permeate; and A is the overall effective membrane area of the membrane modules.

The specific energy consumption of the dual-stage NF process is calculated as a function of operating pressure, feed and permeation flowrate, as well as efficiency of high-pressure pump and energy recovery device (ERD), which is shown in Eq. (3) [36]:

$$Es = \frac{P_f Q_f \left(\varepsilon_{pump}\right)^{-1} - P_b Q_b \varepsilon_{ERD}}{Q_p} \quad (3)$$

where Es is the specific energy consumption of the NF process; P_f and P_b are the pressure of the feed and the retentate stream, respectively; Q_f, Q_b, and Q_p are the feed, the retentate, and the permeate flowrate, respectively, ε_{pump} and ε_{ERD} are the efficiency of the high pressure pump and the ERD, respectively.