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Concentrating brine from seawater desalination process by nanofiltration–electrodialysis integrated membrane technology

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HIGHLIGHTS

- A NF–ED integrated membrane technology for utilization of brine from seawater desalination process was proposed and investigated.
- The rejection of Ca^{2+}, Mg^{2+} and SO_4^{2-} in NF process could be 40%, 87% and 100% respectively.
- The NaCl concentration in ED could be concentrated to more than 160 g/L.

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ABSTRACT

Extraction and concentrating of NaCl from brine were promising technologies to solve the disposition problem of seawater desalination. A novel nanofiltration–electrodialysis (NF–ED) integrated membrane technology was proposed and investigated. The separation of monovalent & bivalent ions and concentrating of solute were realized in NF and ED process, respectively. A kind of NF membrane with high SO_4^{2-} rejection and common homogeneous ion exchange membrane were investigated. The results showed that the operating pressure and feeding solution concentration have obvious effect on the water permeate flux and ions rejection ratios. Due to the special properties of NF membrane, almost all of the SO_4^{2-} in the brine was rejected. The rejection of Ca^{2+} and Mg^{2+} was 40% and 87%. The concentrations of Ca^{2+} in permeate were 392 mg/L in a brine-recycle experiment, which induce lower scaling potential when used as the raw water of ED. The highest NaCl concentration in the concentrating cell could be as high as 160 g/L and the NaCl recovery was about 70% after 5 h under 15 V, while total concentration of "impure ions" (K^+, Ca^{2+} and Mg^{2+}) was about 5 g/L. A concept process was also proposed attempting to realize comprehensive utilization of brine.

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1. Introduction

The shortage of healthy freshwater supply has become more and more serious due to global population and environment problem. Currently, already 1.6 billion people live in regions with absolute water scarcity, which will be increased to two-third of the world's population by 2025 [1]. Among those different solutions, seawater desalination has become important and perspective option in the near-sea regions. Till 2014, the global cumulative commissioned desalination capacity has reached 80.9 billion m^3/d, 59% of which was contributed by seawater desalination [2]. However, a large amount of brine was also effluent out as byproduct. Taking a conventional seawater reverse osmosis (RO) plants of 50% recovery as example, the quality of discharged brine was the same as the fresh water. Disposal of such huge volume of brines may have large environmental impact on the ocean, especially in the closed or semi-closed sea area [3]. Meanwhile, nearly all of the compounds in the seawater (especially NaCl) were rejected in the brine. NaCl is an important industry material widely used in soda and chlor-alkali industry. So no matter for ecology or economy, utilization of the salt resource reserved in the brine is a perspective technology and have attracted more and more researchers.

Because the seawater desalination brine is a mixture of salts and the concentration is not high enough to be economically utilized in industry, separation and concentrating are essential. Several researches on valuable technologies have been carried out [4,5]. Aliza Ravizky et al. [6] reported a dual purpose plant in Eilat, in which an evaporation pond was used to produce salt economically. However, the large area and relatively low efficiency are the most important problems. Membrane distillation (MD), RO and electrodialysis (ED) are the typical membrane technologies used for concentration process. MD, combining membrane and evaporation technology, has high thermal efficient. The researches on the seawater desalination brine have been reported, most of which focus on desalinating, but not concentrating [7–11]. The

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membrane fouling & scaling, and wetting of hydrophobic membrane are key factors of industry application, and energy source & energy consumption optimization are also important [9,10,12–14]. RO is commercially used for a long time compared to MD. A two-stage RO process has been proposed and even been industry operated, which enables to increase water recovery and reduce concentrate quantity [15–17]. As a kind of pressure-driven membrane process, the operation pressure of RO is decided directly by the osmosis pressure. For example, the operation pressure would be as high as 9.0 MPa when the concentrated seawater reached 8% due to its high TDS [16]. The higher TDS of concentrated brine is expected, the higher operation pressure and higher operation & capacity cost will be required. Besides osmosis pressure, scaling potential is another that restricts to concentrate the brine from SWRO. Different from RO, ED is an electro field driven membrane process with lower membrane fouling and higher brine concentration. Some researches on seawater desalination brine have also been carried out. A Zero Discharge Desalination (ZDD) based on ED was proposed, which was used to concentrate NaCl due to its energy-saving feature [18]. It was also pointed out that the scaling ions (Ca^{2+}, Mg^{2+} and SO_4^{2-}) were the most troublesome factor. Similar to above process, an electrodialysis and evaporation combined salt manufacturing process was reported [19]. The discharged brine form a SWRO plant is introduced to the electrodialyzer, the concentrated solution of which was supplied to multiple-effect vacuum evaporator. The results showed that the energy consumption was 80% of the one using seawater as the feed water. High concentration NaCl solution (more than 180 g/L) was also obtained by S. Casas [20,21] and Yoshinobu Tanaka [22]. Meanwhile, bipolar electrodialysis was also evaluated and combined with nanofiltration to produce both rich Mg^{2+} and Ca^{2+} brines for phosphate recovery and HCl and NaOH for desalination treatments [23].

From above works, it could be found that ED has become a perspective technology in the utilization of seawater desalination brine. Its high concentration production, low operation pressure, and lower fouling potential are the most important advantages compared to other technologies. However, monovalent ion selective ion exchange membrane used in all of these studies is much costly. More important is that the scaling potential caused by high bivalent ions concentration still exists. In order to avoid these two problems, a novel nanofiltration–electrodialysis (NF–ED) integrated membrane technology was proposed and investigated in this paper. NF was used to remove the scaling ions (especially SO_4^{2-}) due to its excellent separation performance [24–26]. The normal homogeneous ion exchange membrane was applied in concentrating of NF permeated. The integration and key factors of this process were investigated and discussed.

2. Experiment

2.1. Experimental procedure

The schematic diagram of NF–ED process was shown in Fig. 1, which was composed of NF softening process and ED concentrating process. The effects of operating conditions in once-through process were investigated. A NF process was carried out to simulate the industry application and obtain high recovery ratio, in which the NF brine was piped back and mixed with raw water. The permeation from the retentate-recycle NF process was used as raw water in desalting and concentrating tank of ED system. Na_2SO_4 solution (4%wt) was used as the electrode solution. The solution in desalting & concentrating tank and electrode tank was circulated under flow velocity of 5 cm/s and 10 cm/s respectively in ED concentrating process. The experiment was completed when the concentration of Cl^- in concentrating tank remains nearly constant.

2.2. Membranes and apparatus

In a NF–ED integrated process, the choice of NF membrane should follow several criterions: a) high permeate flux, b) low Na^+ and Cl^- rejection, c) high SO_4^{2-} rejection, d) low operation pressure. Based on the performance of mainly commercial NF membranes [27], DL2540 (GE Co. Ltd.) was selected in this paper. The homogeneous ion exchange membrane (Zhejiang Qianqiu Group Co. Ltd., China) was used in ED process. The properties of the NF and ion exchange membrane were summarized in Tables 1 and 2.

2.3. Chemistry of raw water

Three kinds of artificial seawater desalination brine were studied as raw water in NF process. Their compositions were shown in Table 3. Both of the pH values were adjusted to 5.7 before the experiment to prevent the scaling of $CaCO_3$.

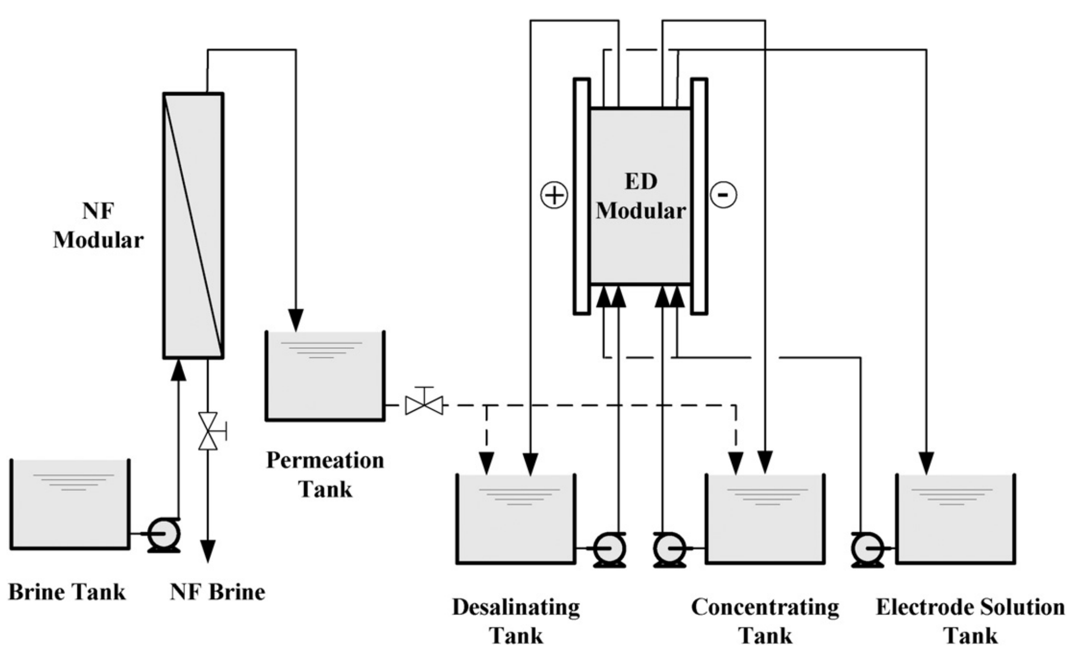

Fig. 1. Schematic diagram of NF–ED hybrid membrane process.

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