



Comparison of membrane distillation and freeze crystallizer as alternatives for reverse osmosis concentrate treatment



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ABSTRACT

Membrane distillation (MD) and freeze crystallizer (FC) were evaluated as alternative reverse osmosis concentrate (ROC) treatment options. A direct contact MD (DCMD) was capable of obtaining 60% water recovery with chemically pretreated ROC. Nevertheless, in repeated cycles, DCMD displayed a trend of reduced water recovery and declining permeate quality. At elevated concentrations, ROC caused scaling and membrane hydrophobicity reduction, indicating reduced membrane life span. On the other hand, FC in three-stage freeze/thaw approach was able to concentrate ROC by 2.3 time, achieving a 57% water recovery with no scaling issues. The fresh ice water quality (total dissolved solids) obtained from FC was within the range of 0.08–0.37 g/L. A brief techno-economic evaluation highlighted advantages and limitations of both options. The efficiency of DCMD as a compact, low thermal process for ROC treatment was compromised by membrane scaling, indicating the necessity for a scaling mitigation pretreatment. This invariably incurs an additional cost. FC was advantageous as a scaling and chemical free process. The high freezing requirement of FC could be met by coupling with refrigerant coolant from liquefied natural gas. Nevertheless, the practical industrial application of FC is inherently restricted due to complex scaling up issues.

1. Introduction

Presently reverse osmosis (RO) technology has gained worldwide acceptance for desalination and wastewater treatment. Globally, RO dominates 60% of total desalination plants [1] due to its techno-economic benefits and high quality water production capacity. RO is a pressure-driven process in which a semi-permeable membrane rejects dissolved constituents present in the feed water [2]. The mechanisms of rejection are due to size exclusion, charge exclusion and physical–chemical interactions between solute, solvent and membrane [2,3].

In Australia, RO technology has taken a prominent role for saline water treatment to meet the rapidly growing demands of irrigation, domestic water supply, mining, coal seam gas (CSG), and power station industry. Presently, Australia has around 10–15 major plants and > 300 small plants using RO technology (Table S1) [4–7].

Although highly reliable, the major limitation of RO is its relatively low recovery at around 35–45%, resulting in large volume of concentrate especially for highly saline feed solutions such as seawater [2]. For coastal plants, RO concentrate (ROC) is discharged back into the ocean. Disposal of ROC containing high salt contents and other chemical compounds could lead to significant environmental issues [3,8]. Mitigation approaches such as dilution are used to reduce ROC salinity

prior to ocean discharge as well as dedicated infrastructures such as long and large pipeline for ocean outfall. This incurs an additional operation cost to the RO plant [5,9,10]. For instance, Perth seawater RO plant, one of Australia's largest coastal RO plant [7], adheres to stringent ocean discharge regulations. This includes 45 times of ROC dilution, post treatment removal of ferric oxyhydroxide flocs from ROC (via a centrifuge and subsequent disposal of the concentrate to landfill) to prevent staining of the white beaches of Cockburn Sound, long pipeline infrastructure as well as constant monitoring of the ocean's dissolved oxygen level at the discharge vicinity [10,11].

For inland RO plants, ROC management is a major challenge as the option of ocean discharge is not available [5,9,11]. The major established inland ROC treatment methods used in Australia are evaporation ponds, deep well injection to aquifer, and brine crystallizer [5,11]. Although low in cost, these technologies are limited for handling high volume flow of ROC, requires large land area and are susceptible to leakage and groundwater pollution [5,9,11]. In the 1980s, only low capacity inland RO plants were applied in Australian mining, and the relatively small ROC volume generated was manageable using evaporation ponds. More recently, the boom in CSG and mining industries resulted in the installation of large treatment plants using RO. For instance, in Queensland, the development of CSG resources in the Surat

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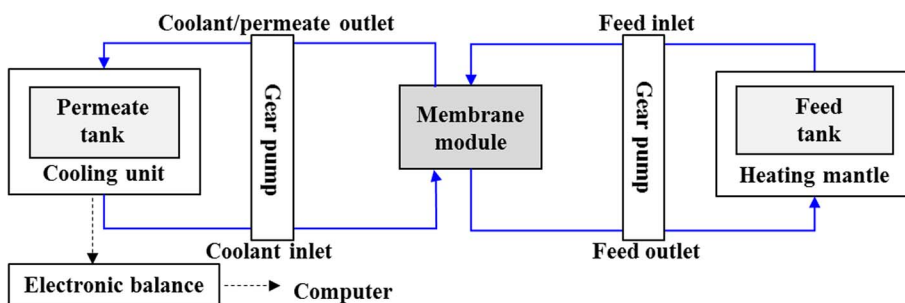


Fig. 1. Experimental setup of DCMD.

and Bowen Basins has seen major growth with the state's annual CSG production, surging from 4 PJ in 1998/1999 to 285 PJ in 2013/2014 [12,13]. CSG extraction generates highly saline wastewater [12,13]. In Australia, RO is predominantly adopted to treat saline CSG wastewater, invariably resulting in substantially large ROC volume. This coincided with greater focus on ROC discharge regulations in Australia.

Ideally, achieving close to zero liquid discharge would be the ideal management solution for ROC. One such technology that exhibit promising concentrate treatment is membrane distillation (MD). MD is a thermal membrane distillation process that operates by transporting water vapor to the distillate (permeate) side through the pores of a hydrophobic membrane [14,15]. As a vapor pressure driven process, MD is not significantly affected by salinity, which accentuates its suitability for treating highly saline ROC. Moreover, the vapor mass transfer mechanism in MD potentially offers complete rejection of ions, enabling the production of additional fresh water from ROC. The low thermal requirement (at 50–80 °C of feed temperature) in MD can be met by solar or heat waste integration [15,16], which is an added advantage over other thermal technologies. However, treating hypersaline ROC which contains high amount of dissolved ions in a thermal based membrane process such as MD, may lead to high inorganic ion precipitation. This phenomenon was highlighted by a number of MD studies [17–20]. For instance, Martinetti et al. [18] demonstrated that the water recovery of vacuum-enhanced direct contact MD operated with ROC was limited by precipitation of inorganic ions such as calcium (Ca) based ion precipitation on the membrane surface. Similarly, Naidu et al. [19] observed the susceptibility of MD towards CaSO₄ precipitation. Specifically, these studies identified Ca as the main scalant in ROC, due to its inverse solubility under thermal condition. Ca then serves as nucleation sites for other species such as Na and Mg. Scaling precipitation could potentially limit MD's long-term performance for ROC treatment. Hence, removing Ca from ROC could potentially reduce scaling issues in MD operation. Scaling mitigation techniques such as inorganic pretreatments could enhance the performance of MD [17,20–22]. A few MD studies have evaluated scaling mitigation techniques such as antiscalant [17], chemical softening [20], membrane cleaning and air back washing [21,22]. However, repeated cycles of operation are essential in determining the feasibility of MD for ROC treatment and the effectiveness of pretreatment as well as the implication of additional pretreatment cost.

Given the susceptibility of scaling in membrane processes, it is worth evaluating the potential of a non-membrane based technology for ROC treatment. In this regard, freeze crystallization (FC) is a non-membrane based technology that show promise for concentrate treatment. The principle of FC is based on liquid–solid phase in which, fresh water ice forms during freezing, while highly soluble dissolved salts concentrate remains in the liquid phase [23,24]. This leads to the separation of ice and concentrate [24–29]. Nevertheless, a major challenge of FC is the co-adsorption of salts with ice crystals. A large number of small sized ice crystals (ice slurry) are generated in FC, resulting in high specific surface area. These surfaces are usually covered by a slew of salts due to the strong adhesive force between ice and salt concentrate [27–29]. This entails washing to remove the salts. For

instance, Chang et al. [28] highlighted that under optimal FC operating condition for seawater, the optional amount of washing water to clean raw ice was about 50% of the raw ice produced. Minimizing washing would enhance FC performance as well as avoid the dilution of concentrate. A potential way of minimizing washing is to reduce the surface area of ice formed. The prospect of treating ROC using FC with minimal washing is explored in this study.

This study aims to evaluate the performance of MD and FC as alternative ROC treatment technologies. The potential of pretreated ROC for improving the performance of MD was considered. Repeated cycles of MD operation were carried out to determine the membrane life span and effectiveness of pre-treatment. The approach of FC with minimal washing requirement was explored. Water recovery ratio, fresh water quality, tendency of scaling development as well as membrane life span were the main factors used in comparing the performance of MD and FC for ROC treatment.

2. Materials and methods

2.1. Direct contact membrane distillation (DCMD)

In this study, MD performance capacity for ROC treatment was evaluated with a bench scale DCMD (Figs. 1 and S1). In this set-up, an acrylic membrane cell with depth, width, and length of 0.2 cm, 5.0 cm and 8.0 cm (40 cm² effective membrane area) was used. A commercial hydrophobic polytetrafluoroethylene (PTFE) flat sheet membrane (General Electric, US) with a support layer of polypropylene was used. The porosity, average pore size and membrane thickness provided by the supplier were 70–80%, 0.2 μm, and 179 μm, respectively [21]. DCMD experiments were carried out at a moderate feed temperature (T_f) of 55 °C. Deionized (DI) water was used as the cooling/initial permeate solution, which was set at a permeate temperature (T_p) of 25 °C. The bulk temperatures were maintained within the variation of ± 3 °C for all experiments. The feed solution (1.5 L of actual and pretreated ROC) and permeate solution (1.5 L deionized water (DI)) were channelled into the membrane cell at a feed and permeate flow velocity of 1.1 m/s in a counter current mode with a gear pump.

The DCMD operation was carried out up to the point where the initial feed volume (1.5 L) was reduced to around 0.5–0.6 L (achieving 60–65% water recovery) or until a significant permeate flux decline occurred. Three repeated cycles of DCMD operation (cycles 1–3) were carried out using the same membrane. After each cycle, DI water (0.1 L) was flushed at low velocity (0.6 m/s) into the feed module to clean the membrane. At the end of cycle 3, the used membrane was removed from the module for further analysis as described in Section 2.4.2.

2.2. Freeze crystallizer (FC)

This study used a batch reactor FC set-up (Fig. 2), comprising of four reactor tanks (bulk, crystallizer, ice and thawing tank). The bulk ROC (1.5 L) was precooled to 0 °C. The ROC was channelled to a slurry tank (placed in a chiller unit), which was maintained at – 8 to – 11 °C based on previous studies [24–27] that established the freezing points of salt

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