



Two-stage sulfate removal from reject brine in inland desalination with zero-liquid discharge



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HIGHLIGHTS

- Two-stage process for SO₄ removal from NF-reject brine
- Successful removal at reasonable lime and aluminum ratios
- Effective removal at a high pH range
- The experimental model developed accurately predicted experimental results.

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ABSTRACT

This research proposes a two-stage process for the removal of sulfate from a nano-filtration (NF) reject stream. In Stage 1, calcium chloride is used to partially remove the majority of sulfate as calcium sulfate precipitate while the remaining sulfate is removed in Stage 2 as calcium sulfoaluminate precipitate. Kinetics of the removal process in both stages was investigated and the results indicated that the removal kinetics is not a limiting factor for the practicability of the process. Equilibrium characteristics of sulfate precipitation were evaluated and optimum conditions for maximum sulfate removal were obtained. Sulfate removal in Stage 1 was independent of the pH while efficient sulfate removal in Stage 2 was found to be above pH 11. Efficient sulfate removal was observed at practical ranges of lime and sodium aluminate doses. An equilibrium model was developed to describe the chemical behavior in Stage 2. A valid explanation for the mechanism of sulfate removal in Stage 2 was the formation of a solid mixture consisting of ettringite and monosulfate as confirmed by X-ray diffraction (XRD) analysis. This approach is expected to reduce the volumes of the overall precipitated solids and in turn improve the overall efficiency of the ZLD treatment system.

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1. Introduction

The deterioration of groundwater quality due to seawater intrusion and other human activities in the Gulf region and many arid countries has led to the use of inland desalination techniques for the production of good quality water. Clearly, any desalination process produces two streams; a clean water product stream and a reject concentrate stream (called brine) that must be disposed of. Reject brine disposal into the sea is not an option for inland desalination. Improper disposal of reject brine from inland plants results in the following problems [1]: (1) Pollution of groundwater resources that are used as feed water for desalination plants due to high salt content and the presence of other harmful chemicals in the concentrate; (2) production decline in agricultural lands caused by the deposition of airborne salts from dried concentrate;

(3) formation of eyesores caused by the improper disposal of concentrates on nearby land; and (4) rendering treated municipal sewage effluent unsuitable for agriculture due to high total dissolved solids (TDS). Environmental issues caused by the improper disposal of reject brine pose major challenges for inland desalination and urge the need for zero liquid discharge (ZLD) [2,3].

Most ZLD systems in operation today use thermal processes, evaporation ponds, or a combination of these methods [4]. While thermal evaporation is a proven process, it is energy-intensive and suffers from high capital and operating costs. On the other hand, evaporation ponds typically require large areas of land, and their construction costs are high. Furthermore, water evaporated from a pond is a lost resource.

Given the need for ZLD and the drawbacks of existing ZLD methods, it is imperative to find alternative ZLD treatment technologies that provide more affordable concentrate management. This can be achieved through the development of inexpensive treatment techniques that maximize water recovery while minimizing the volume of concentrated

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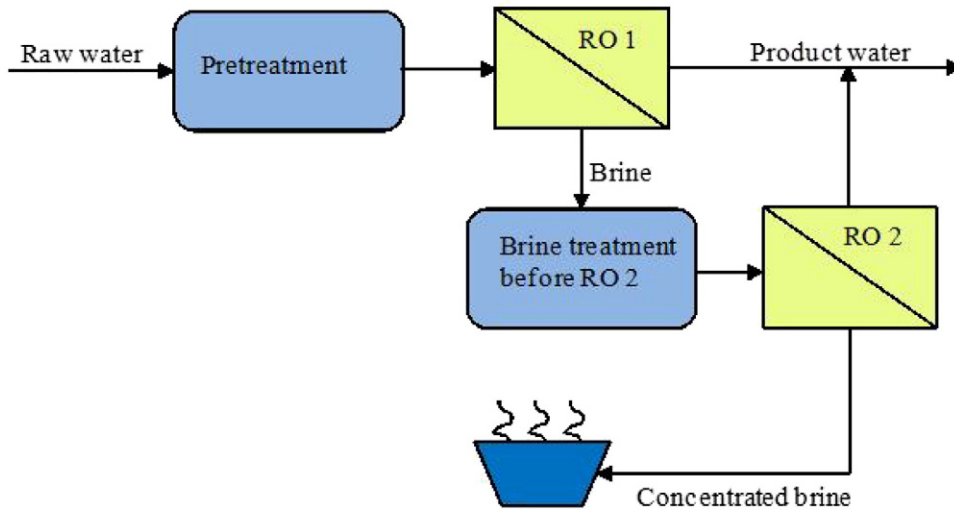


Fig. 1. Schematic diagram of the two-stage RO with intermediate brine treatment.

brine that needs to be vaporized. When brackish groundwater is desalinated using reverse osmosis (RO), the constituents of the water are concentrated in the reject brine by a factor of 3–10 times that of raw water, causing membrane fouling depending on the desalinated water recovery [5,6]. Sulfate salts, such as barium sulfate or calcium sulfate, form a scale on the membranes during water recovery. Although the calcium sulfate scale is more soluble than other mineral scales such as CaCO_3 , BaSO_4 , and SrSO_4 , once formed, it is difficult to eradicate mechanically and is insoluble in mineral acids and other conventional solvents [7–9]. Sulfate is present in high concentrations (~700 mg/L as SO_4^{2-}) in brackish groundwater in Qatar, resulting in limited water recovery in desalination systems.

Several methods were proposed for sulfate removal from water including chemical treatment by precipitation [10–12], membrane separation using nanofiltration [13–15], ion-exchange and/or adsorption [16–18] and biological treatment [11,18] or a combination of these processes. However, these methods suffer from limited removal efficiency and/or high cost. In addition, ion exchange and membrane separation involve the generation of a liquid waste stream that would require proper management.

Sulfate can also be removed by precipitation with calcium and aluminum as calcium sulfoaluminate which has a low solubility product.

Important evidence to consider is the performance of sulfate in the chemistry of Portland cement. The stability of calcium sulfoaluminate, commonly known as ettringite ($\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12}$), at equilibrium relies on adequate activities of calcium, aluminum, and sulfate [19]. It also depends on the pH, temperature, and sulfate availability. It has been reported that the most favorable pH conditions for the formation and stability of ettringite are in the domain of 11 and 12.5 [19,20]. Increasing the pH above the stability range will cause the ettringite to convert to calcium hydroxide leaving behind a solution of sulfate and aluminate ions. Damidot and Glasser reported from thermodynamic calculations that ettringite was stable at all times relative to monosulfate ($\text{Ca}_4\text{Al}_2(\text{SO}_4)(\text{OH})_{12}$) at 25 °C [19,21]. In order for ettringite to develop over monosulfate, the aluminum-to-sulfate ratio has to be lower than 1.0 [19]. The availability of more aluminum than sulfate will result in sulfate developing into monosulfate and excess aluminum existing in the hydroxyl-substituted AFm (alumina, ferric oxide, monosulfate), hydroxy-AFm, phase. If there is a slight excess of sulfate, a mixture of monosulfate and ettringite will develop in the system. As the available sulfate level increases, more ettringite and less monosulfate will develop [22,23].

Previous research has shown that sulfate was efficiently removed from recycled cooling water by precipitation with calcium and

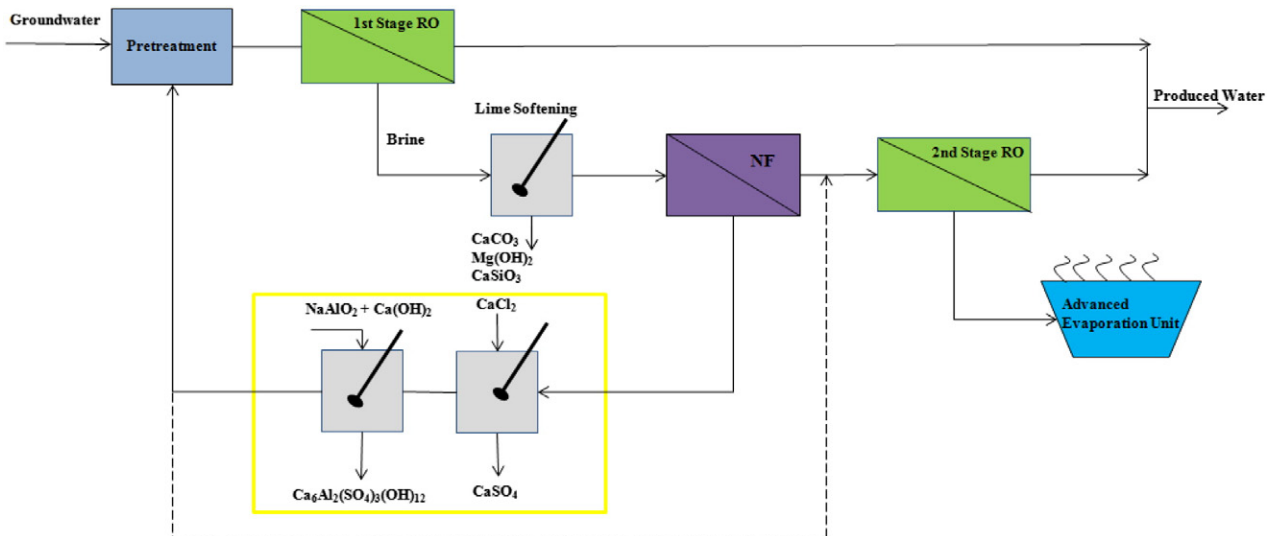


Fig. 2. Schematic of the two-stage precipitation process of NF reject brine.

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