Electrocoagulation process considerations during advanced pretreatment for brackish inland surface water desalination: Nanofilter fouling control and permeate water quality

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HIGHLIGHTS

- Nanofiltration (NF) is capable of desalinating inland brackish surface water.
- Integrated electrocoagulation-microfiltration (MF) pretreatment controls NF fouling.
- NOM fouling dominated NF after MF-only pretreatment.
- FTIR and XPS successfully diagnosed dominant NF foulants and fouling mechanisms.
- NF achieves excellent (>90%) strontium removal from brackish surface water.

GRAPHICAL ABSTRACT
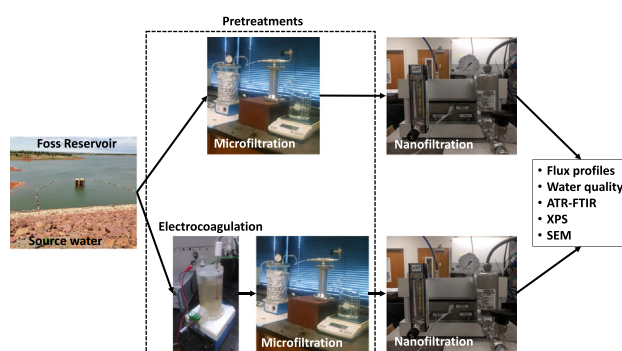
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ABSTRACT

Pretreatment with coupled aluminum electrocoagulation (EC) – microfiltration (MF) successfully mitigated colloidal and organic fouling during nanofiltration (NF) of an inland natural brackish surface water. Natural organic matter (NOM) removal by EC was enhanced due to (i) greater degree of NOM protonation by maintaining a slightly acidic pH during electrolysis, compared with that of minimum aluminum solubility (pH of 5.5 compared to 6.2), (ii) providing sufficient contact time during electrolysis and flocculation by utilizing intermediate current density (10 mA/cm^2), and (iii) combination of charge neutralization and sweep coagulation by increasing aluminum dosage. Direct evidence for removal of the hydrophobic fraction of NOM along with a portion of the hydrophilic moieties by EC-MF compared to MF-only was provided by attenuated total reflectance-Fourier transform infrared spectroscopy. NF flux profiles with EC-MF pretreatment of natural saline water were nearly identical to that of a model solution with similar ionic composition with no added NOM suggesting insignificant role of organic fouling. X-ray photoelectron spectroscopy showed trace amounts of CaCO$_3$ precipitates after EC-MF pretreatment, confirmed by electron microscopy, that caused only minor flux decline. Finally, NF achieves excellent strontium removal as well as other divalent ions and NOM.

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1. Introduction

Desalination is becoming an increasingly discussed alternative in our toolkit of available treatment options to provide drinking water [1]

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especially in times of drought when water supplies increase in salinity [2]. To this end, a significant body of research has demonstrated the capability of reverse osmosis (RO) to desalinate seawater and brackish groundwater [3]. However, RO requires very high transmembrane pressures making it economically less attractive in many occasions. Importantly, energy costs can be significantly reduced by implementing nanofiltration (NF) [4] especially in cases when divalent ions are the dominant component of salinity as in many lakes and reservoirs in West-Central United States [5,6]. Fouling is an important impediment to NF implementation, especially during surface water treatment [7–10].

Microfiltration (MF) or ultrafiltration (UF) pretreatment is necessary to mitigate colloidal fouling during surface water desalination [8,11–13]. However, low-pressure membrane pretreatment may not be sufficient for waters with severe organic fouling potential, necessitating coagulation as an additional pretreatment step [14–16] to maintain flux. Electrocoagulation (EC) is an attractive alternative to conventional chemical coagulation especially for smaller plants given its many operational advantages including (1) portability and modularity, (2) reduced handling of corrosive chemicals, (3) lower alkalinity consumption, and (5) ease of automation [17–19]. To date, the available investigations on membrane fouling control by EC have focused largely on MF and UF [18,20–24] consequently shedding little light on its impacts on NF performance in an integrated dual-membrane system. To our knowledge, only one investigation has coupled both EC and MF as pretreatment for surface water NF [16]. This work demonstrated significantly lower NF fouling with electrochemical pretreatment compared with conventional coagulation for a non-saline "freshwater" supply. Based on this recent finding, we hypothesized that EC would be highly efficient in combating membrane fouling during desalination of high salinity surface water by MF-NF since (i) electrolysis can be performed at very high current density given the high conductivity of the source water thereby substantially reducing electrolysis time (i.e. allowing for very small and portable EC systems), and (ii) generation of a greater number of smaller-sized hydrogen bubbles at high current density [25] will facilitate natural organic matter (NOM) removal by increased hydrophobic interactions thereby reducing organic fouling.

In addition to being capable of high removals of divalent ions, NF provides an excellent barrier against disinfection by-product (DBP) precursors [8], contaminants of emerging concern [26,27], and many other currently regulated contaminants while providing treated water with sufficiently low dissolved solids concentrations [28]. Of particular interest to this research is strontium, which is a top priority for the United States Environmental Protection Agency and categorized as Group I in its third Contaminant Candidate List (CCL3) for regulatory consideration as a primary drinking water standard [29]. Sr^{2+} occurs widely in public water systems [30] along with other divalent ions such as Ca^{2+} and Mg^{2+} but its removal during drinking water treatment is only beginning to be investigated. For example, Sr^{2+} was recently shown to be better removed from groundwater by lime softening and NF [31,32] than from surface water by conventional treatment processes [31]. It has also been artificially spiked to a freshwater source and shown to be very well removed by low-pressure RO [33]. To our knowledge, there are no reports of the removal of naturally occurring Sr^{2+} from saline surface water by NF.

The principal objectives of this research focusing on desalination of brackish water from the Foss Reservoir in West-Central Oklahoma, USA are to: (i) optimize EC operating parameters to maximize NOM removal and (ii) evaluate the effectiveness of coupling EC and MF pretreatment to mitigate NF fouling. We also report data on strontium removal by NF. Transient NF fouling profiles following pretreatment by EC-MF and MF-only were compared with a synthetic feed water, which was formulated to closely mimic the ionic composition of Foss Reservoir water without any added NOM to operationally establish the role of NOM on fouling. All experiments were performed at bench-scale using a commercial NF membrane (NF270, Dow), whose surfaces were characterized in detail by Fourier Transform Infrared (FTIR) Spectroscopy and X-ray Photoelectron Spectroscopy (XPS) after different pretreatments to identify dominant foulants and establish fouling mechanisms.

2. Materials and methods

2.1. Source water

A sample was collected in November 2015 from the Foss Reservoir in Oklahoma, USA serving as a model inland brackish surface water. The source water was slightly alkaline (pH 8.0 ± 0.2), very hard (total hardness 1620.7 ± 10.61 mg/L as $CaCO_3$ and calcium hardness 720.3 ± 3.54 mg/L as $CaCO_3$), and high in buffering capacity (alkalinity 115.8 ± 2.0 mg/L as $CaCO_3$) and salinity (conductivity 2766 ± 235 μS/cm). Major anions were quantified by ion chromatography (IC) revealing sulfate (1623.5 ± 90 mg/L) as the dominant one with lesser amounts of chloride (174.5 ± 21 mg/L), and bicarbonate (141.2 ± 2.4 mg/L). Calcium (286.9 ± 7.7 mg/L), magnesium (218.1 ± 12.9 mg/L), sodium (199.2 ± 8.2 mg/L) were the major cations measured by inductively coupled plasma–mass spectrometry (ICP-MS) with lower concentrations of potassium (10.5 ± 1.2 mg/L), strontium (8.8 ± 1.0 mg/L) and aluminum (0.5 ± 0.002 mg/L). Silicon was also present (13.8 ± 0.2 mg/L) in the water sample.

The NOM concentration was measured as dissolved organic carbon (DOC) concentration and UV_{254} absorbance. The DOC concentration and UV_{254} absorbance were 9.4 ± 0.5 mg/L and UV_{254} 0.11 ± 0.004 cm^{-1}, respectively, despite being low in turbidity (2.2 ± 0.6 NTU). The low specific UV absorbance value (SUVA, defined as the ratio of UV_{254} in m^{-1} and DOC concentration in mg/L) of 1.19 ± 0.08 m·L/mg indicated that the NOM was predominantly non-humic, highly hydrophilic, and low in molecular weights [34]. Fractionation using Supelite DAX-8 resin (Sigma Aldrich) confirmed that only 24% of NOM was hydrophobic; the remaining 76% comprising hydrophilic/transphilic fractions.

A model solution formulated to have similar ionic concentration and pH as Foss Reservoir water but without added NOM was also employed as a negative control to evaluate organic fouling. It was prepared by dissolving 3.29 mM Na_2SO_4, 8.98 mM $MgSO_4$, 2.52 mM $CaCO_3$, 4.63 mM $CaSO_4$, 2.08 mM NaCl, and 2.84 mM HCl in nanopure water.

2.2. NF pretreatment

Two NF pretreatment processes were evaluated; MF-only and EC followed by MF. Dead-end MF was performed at constant pressure (1.38 bar) using a 300-mL stainless steel stirred cell (SEPA ST, Osmonics) and 0.22 μm modified PVDF membranes (GVWP04700, Millipore).

Batch EC experiments were performed in galvanostatic mode using a 500 mL custom-made Perspex cell fitted with an Al anode (99.9965%, Alfa Aesar) with initial effective area of 16.39 cm^2 and a perforated cylindrical 316-stainless steel cathode. The anode was mechanically scrubbed and thoroughly rinsed prior to electrolysis. The entire cell was rinsed with dilute HNO_3 to remove traces of precipitated coagulant after each set of experiments (i.e. five consecutive electrolysis). Operating conditions were optimized by systematically varying the current density (10 or 40 mA/cm^2), pH (5.5 and 6.2) and target aluminum concentrations (0–40 mg/L) by adjusting the electrolysis time, cell potential, and adding H_2SO_4 as necessary. After electrolysis, the suspension was flocculated for 30 min and allowed to settle for 30 min. The total aluminum concentration was measured by ICP-MS after acidification with HNO_3, which agreed to within 5% of the value obtained by simply weighing the anode before and after electrolysis. The zeta potential of suspensions was measured using an electrophoretic light scattering