

## Silica removal to prevent silica scaling in reverse osmosis membranes



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### HIGHLIGHTS

- Different methods to remove silica from solution were investigated.
- Al<sup>3+</sup> was the most efficient precipitant for silica, removing up to 99% of silica.
- A strongly basic anion exchange resin removed silica up to 94%.
- Monitoring residual silica and Al<sup>3+</sup> is crucial to prevent scaling in membranes.

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### ABSTRACT

Reverse osmosis membranes are increasingly used in drinking water treatment. However, the production of a concentrate stream is the main disadvantage of its application. Increasing the recovery of the membranes in order to have the smallest amount of concentrate possible is an attractive approach. In the absence of bivalent cations in the feed water, silica and silica-derived precipitants are limiting factors in high-recovery reverse osmosis operations. The removal of silica in a separate pretreatment process might be the solution. Several methods were tested to remove silica. Precipitation of silica with Fe(OH)<sub>3</sub>, Al(OH)<sub>3</sub> and silica gel was investigated, and also the removal of silica using a strongly basic anion (SBA) exchange resin.

Al(OH)<sub>3</sub> was the most effective precipitant for silica, removing nearly all of the molecularly dissolved silica. However, a residual amount of aluminum remained in solution, and aluminosilicate colloids were not removed. The use of the SBA exchange resin also showed a good performance, removing up to 94% of the silica. However, further investigations, such as checking whether the residual small amounts of silica and aluminum can still cause scaling in the membrane, need to be conducted.

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### 1. Introduction

Nowadays membrane technology is progressively used to produce drinking water [1]. Nanofiltration (NF) and reverse osmosis (RO) produce high quality water by removing pathogens, organic micropollutants, colloids, natural organic matter and salts. However, NF and RO have several drawbacks, like membrane fouling and the production of concentrate. One of the major foulants of RO membranes is silica. Its ubiquitous presence in natural waters can complicate desalination processes because of its high scaling potential [2–4], and the complex chemistry of silica adds to the difficulty of this problem. Silica solubility is low, about 120 mg/L in water at pH 7 and at 25 °C. It is usually assumed that as long as the

concentration of Si(OH)<sub>4</sub> is below 120 mg/L there is no polymerization, but it could be that solutions with lower concentrations might nucleate less soluble polymeric species of lower solubility [5]. Furthermore, the presence of iron and aluminum decreases the solubility of silica [5]. Once silica polymerizes, deposition on the membranes is likely and it is difficult to remove it by cleaning. Therefore, preventing the occurrence of silica scaling is preferred.

In our previous research we studied the feasibility of achieving a high RO recovery in a system composed of a cation exchange (CIEX) resin, followed by treatment with NF and RO, which treated the NF concentrate [6]. We found that at >94% total system water recovery, silica scaling became the limiting factor for the investigated water type. The silica concentrations in the bulk solution of the RO feed varied between 80 and 200 mg/L SiO<sub>2</sub>.

One of the options to prevent silica scaling consists of removing silica in a separate pretreatment process. Several investigations have been

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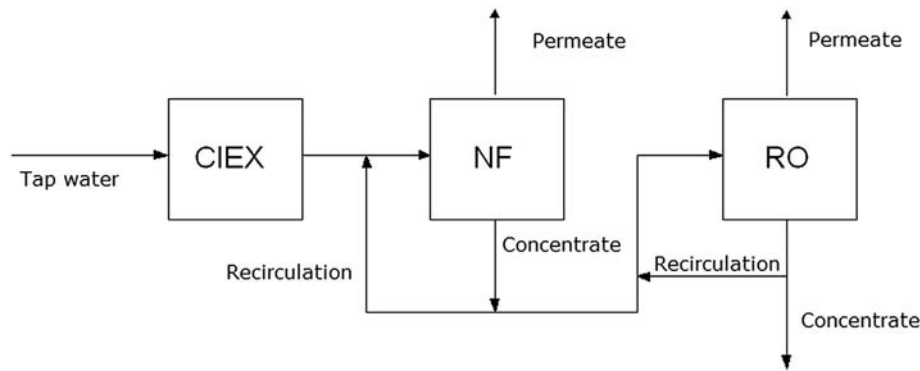


Fig. 1. Simplified scheme of the CIEX–NF–RO system.

conducted in this field. Silica can be removed from water by precipitation with multivalent metal hydroxides, such as  $\text{Fe}(\text{OH})_3$ ,  $\text{Al}(\text{OH})_3$  and  $\text{Mg}(\text{OH})_2$ . This treatment removes both soluble and colloidal silica [3,7,8]. Other methods of silica removal, such as electrocoagulation and in-line coagulation/ultrafiltration have been also investigated and were able to remove up to 65 and 80% of the silica, respectively [9,10]. Silica can also be removed with anion exchange [11] or by chemical (lime or caustic soda) softening at  $\text{pH} > 10$  [12,13]. The removal of silica by chemical softening is in co-precipitation with  $\text{Mg}(\text{OH})_2$ . Thus, to remove silica with chemical softening, it should be applied before CIEX – as CIEX removes nearly all  $\text{Mg}^{2+}(\text{aq})$ .

The aim of this study is to find the most suitable method and conditions to remove silica in order to be able to reach very high recoveries with RO without silica scaling. In the present paper we report different experiments to remove silica with  $\text{Fe}(\text{OH})_3$ ,  $\text{Al}(\text{OH})_3$ , silica gel and a strongly basic anion (SBA) exchange resin, performed with synthetic water, tap water, and water extracted from a pilot plant [6], paying special attention to the amounts of residual silica and residual precipitant after the treatment. With the results obtained we can design a pretreatment step to remove silica, avoiding silica scaling in the RO membrane.

## 2. Materials and methods

Two different kinds of experiments were performed in an attempt to remove silica in the most efficient way. The first method was the removal of silica by precipitation and the second one by means of a strongly basic anion (SBA) exchange resin.

### 2.1. Silica removal by precipitation

#### 2.1.1. Experimental procedure

Silica removal experiments were performed using 250 mL plastic covered beakers. The solutions were placed in the beakers and the required reagent (see below) was added, if necessary. The pH was checked with a pH meter (Radiometer Copenhagen PHM95) and adjusted to the desired value by addition of (1 M) HCl or (1 M) NaOH (Sigma Aldrich).

Supersaturated silica solutions (200 mg/L) were prepared by dissolving the corresponding amount of  $\text{Na}_2\text{SiO}_3 \cdot 5\text{H}_2\text{O}$  (Sigma Aldrich) in deionized water. The  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$  and silica gel used to precipitate the silica were from JT Baker, Merck and Sigma Aldrich, respectively. The silica gel had a particle size ranging from 9.5 to 11  $\mu\text{m}$  and its pore size ranged from 50 to 76  $\text{\AA}$ .

The solutions were stirred continuously at 600 rpm. The samples for analysis were collected every 10 or 30 min, depending on the experiment, with a plastic syringe and filtrated with a 0.22  $\mu\text{m}$  Millipore filter. All the experiments were done in duplicate. In all the experiments using deionized water, the pH was adjusted to  $8.5 \pm 0.3$ .

Dissolved silica, also known as reactive silica, was analyzed with a spectrophotometer at 452 nm wavelength (Hitachi U-2900).

These analyses were done immediately after sampling, following the silicomolybdate Hach method 8185. Total silica (reactive and non-reactive) was measured by means of Ion Couple Plasma Atomic Emission Spectrometry (ICP-AES), using an ICP analyzer (Spectro Arcos). In the experiments using  $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ , the dissolved silica was too low to be determined spectrophotometrically, thus, the remaining silica was total silica measured only with ICP-AES. The concentrations of aluminum remaining in the treated water were determined after filtration with a 0.22  $\mu\text{m}$  filter, by ICP-AES.

#### 2.1.2. Water type

The experiments were performed using different types of water: deionized water and water extracted from a pilot plant described elsewhere [6]. The pilot achieved high recoveries in the treatment of tap water by subsequent application of CIEX, NF and RO (on the NF concentrate). In the pilot the water was recirculated over both the NF and RO in local loops, in order to get sufficient cross-flow. Three water types were extracted from the pilot, to investigate the efficiency and impact of precipitation using  $\text{Al}^{3+}$  for the removal of silica at three different possible stages in the pilot installation. The extracted water types were: the tap water feeding the pilot ('tap water'), water from the NF recirculation loop ('NF recirculation') and water from the RO recirculation loop ('RO recirculation'). A simplified scheme of the system is shown in Fig. 1. The average composition of these streams, including the concentrations of silica, is given in Table 1.

By performing precipitation experiments in three different water types, the most optimal location for silica precipitation in a CIEX–NF–RO system can be assessed.

#### 2.1.3. Precipitate characterization and particle size distribution

For the design of a separate process for removing silica it is important to characterize the precipitate formed [14]. Information about particle size and shape is needed to design the proper filtration unit to follow the precipitator. Ultrafiltration, for instance, might be required to filter residual colloids after precipitation.

Table 1

Average composition in the different water streams tested (all in mg/L except pH).

	Tap water	NF recirculation	RO recirculation
$\text{Ca}^{2+}$	70	0.40	2.31
$\text{Na}^+$	15	677	4300
$\text{Mg}^{2+}$	5.98	0.03	1.21
$\text{K}^+$	1.23	0.57	3.37
$\text{Al}^{3+}$	0.02	0.12	0.50
$\text{Fe}^{3+}$	0.00	0.03	0.10
$\text{Cl}^-$	10	47.5	285
DOC	1.90	16	101
$\text{HCO}_3^-$	271	1540	10,300
$\text{SiO}_2$	18	20	140
pH	8.1	8.3	8.8

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