



Influence of surfactant fouling on rejection of trace nuclides and boron by reverse osmosis



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HIGHLIGHTS

- Surfactants affect trace nuclides and boron rejection and membrane flux.
- Surfactant deposition on membrane surfaces may change surface characteristics.
- Nuclide rejection is correlated with membrane roughness.
- Boron rejection is correlated with membrane hydrophilic property.

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ABSTRACT

Recent studies have shown that contaminant rejection depends greatly on membrane surface morphology and structure. In this paper, we investigate the influence of surfactants on reverse osmosis membrane properties and the rejection of trace nuclides in low level radioactive wastewaters. The results demonstrate that surfactant deposition on membrane surfaces may change surface characteristics such as roughness and contact angle, and therefore affect trace nuclide rejection and membrane flux. After a 12 h fouling test using surfactants sodium dodecyl benzene sulfonate, Tween-80 and cetyltrimethyl ammonium bromide, the permeate flux declined by ~55.8%, 44.86% and 40.61%, respectively. CTAB deposition on the membrane increases nuclide rejection, whereas sodium dodecyl benzene sulfonate and Tween-80 deposition decreases nuclide rejection. All surfactant deposition enhances boron rejection. According to a correlation coefficient analysis, the decrease in flux can be attributed mainly to a variation in membrane surface hydrophilic properties resulting from surfactant deposition. The nuclide rejection and boron transfer ability correlate with membrane roughness and hydrophilic property, respectively.

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1. Introduction

Membrane technology has been used extensively in seawater desalination [1,2] wastewater treatment [3] and water purification [4]. Since it uses tense membranes, reverse osmosis (RO) is capable of rejecting almost all macromolecular substances and charged particles, and is often considered for the treatment of trace contaminants. For example, RO has been used extensively for the removal of trace heavy metals from drinking water resources [5–7]. The rejection of contaminants depends greatly on membrane surface morphology and structure, such as membrane pore size, active skin layer thickness [8] and membrane roughness [9].

However, in some cases, target contaminant concentrations are several orders of magnitude lower than the non-target components, which are

defined as “background composition” in this paper. The background composition may change the membrane surface characteristics, including roughness, and contact angle, therefore, may affect the transport of trace contaminants [10]. For instance, the rejection of N-nitrosamines increases when membranes are fouled by background compositions in the tertiary effluent [11]. The rejection of boron increases when the contact angle of the membrane is changed by humic acid after fouling [12].

The influence of membrane surface morphology and structure transformation resulting from the background composition may be enormous if RO is used to treat low level radioactive wastewaters (LLRWs) from nuclear power plants (NPPs). In LLRWs, the radioactive nuclides exist in trace amounts, whereas other radiologically inactive ions and organic compounds exist in excessive amounts. According to regulations of the International Atomic Energy Agency, LLRWs are defined as solutions with a total radioactivity below 4.0×10^6 Bq/L. Therefore, the concentrations of radio-nuclides are far below 1 μg per liter, or even lower than 1 ng per liter. However, organic compounds and non-radioactive ions exist in LLRWs in excessive concentrations, far above

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1 mg per liter and in some cases even in amounts up to several moles per liter [13,14]. In addition, boron released from primary coolant during the outage period in pressurized or boiling water reactors amounts to an average concentration of $500 \text{ mg} \cdot \text{L}^{-1}$ [15]. These background compositions may change membrane surface morphology, and affect the rejection of trace nuclides.

Research on the treatment of LLRWs by RO was conducted in the 1980s, which enabled design of a full-scale system consisting of cross-flow microfiltration, spirally wound reverse osmosis, and tubular reverse osmosis [16]. Generally, RO system can provide α and β rejection of above 90% [17]. The treatment of ^{137}Cs -contaminated water by RO demonstrated an efficient removal, with ^{137}Cs rejection of above 90%, even higher than salt rejection [18,19]. In addition, RO technology can exhibit a marked superiority of reliability over some emerging technologies like capacitive deionization [20–23]. Although RO technology has been used in several nuclear facilities, such as Chalk River Laboratories in Canada [24], Nine Mile Point station, Comanche Peak NPP and Dresden NPP in the United States [25–27], for the treatment of reactor coolant, floor drain or liquid from steam generator chemical cleaning, a study of the influence of organic compound fouling on trace nuclide rejection by reverse osmosis is still lacking.

In this paper, the influence of surfactant on the rejection of typical nuclides such as cesium, strontium and cobalt in boron-containing water was investigated using a commercial RO membrane material. Three surfactant types (cationic, nonionic and anionic) were used to reflect all possible LLRW states. Correlations with membrane surface properties and flux declines were identified. The results will provide an improved understanding of wastewater treatment design in NPPs and expand RO technology applications in the nuclear industry.

2. Experimental

2.1. Materials and installation

Experiments were carried out in a laboratory-scale installation as shown schematically in Fig. 1. Raw water was pumped into an intermediate buffer vessel equipped with a temperature-control system. Then it was conducted across a cross-flow membrane-filtration apparatus (SEPA CF II) equipped with flat sheet membrane. The membrane-filtration apparatus was manufactured by General Electric Corporation with a flow channel of $146 \text{ mm} \times 96 \text{ mm} \times 0.86 \text{ mm}$ and an effective membrane area of 140 cm^2 . The instantaneous flux was monitored continuously and recorded in real time on a computer, and then recirculated to the feed vessel.

At the beginning of each experiment, the membrane sample was compacted using demineralized water at 0.4 MPa for 6 h to obtain a stabilized flux. Following membrane compaction, fouling tests were carried out under four different conditions within 12 h. Each experiment was repeated three times, with the operating pressure, temperature and flow rate controlled at 1.2 MPa, $25 \text{ }^\circ\text{C}$ and 0.4 m s^{-1} , respectively.

Table 1
Properties of LE membrane.

Membrane	Roughness (Ra, nm)	Contact angle ($^\circ$)	Flux ($\text{L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$)	Rejection (%)
LE	73 ± 0.5	85 ± 0.8	49.55	99.30

During the fouling experiments, feed and permeate samples were collected hourly.

A low-pressure LE membrane (Dow Chemistry) was studied. Its properties are summarized in Table 1.

2.2. Chemicals and raw water

CsNO_3 , $\text{Sr}(\text{NO}_3)_2$ and $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ bearing solutions of demineralized water were used as raw water with cesium (I), strontium (II) and cobalt (II) concentrations of $\sim 5 \text{ mg} \cdot \text{L}^{-1}$. The boron concentration was $500 \text{ mg} \cdot \text{L}^{-1}$. To investigate the influence of organic compound fouling on the rejection of trace nuclides by reverse osmosis, the raw water was spiked with three different kinds of surfactants (sodium dodecyl benzene sulfonate (SDBS), Tween-80 and cetyltrimethyl ammonium bromide (CTAB)). In general, these surfactants exist in LLRWs because of drain pipe cleaning, surface decontamination, floor cleaning and showers. The organic compound concentration was $0.1 \text{ mmol} \cdot \text{L}^{-1}$. At this concentration, there is no formation of micelles in each surfactant. All chemicals in the raw water were of analytical grade.

For the fouling tests, the feed solution was raw water, or raw water spiked with SDBS, Tween-80 or CTAB. After the fouling tests, membrane samples were washed carefully with pure water, dried in a vacuum, and labeled as M-Blank, M-SDBS, M-Tween-80 and M-CTAB for further analysis. The fresh membrane was labeled as M-virgin.

2.3. Membrane characterization

The chemical properties of the fouled membranes were measured by Fourier transform-infrared spectroscopy (Spectrum One, PerkinElmer). Fouling membrane surface imaging was performed by scanning electron microscopy (SEM, HITACHI S-5500) and atomic force microscopy (AFM, SPA-300HV, SEIKO). Membrane surface roughness was determined by AFM surface image analysis. The membrane hydrophobicity was estimated by water drop contact angle measurement using a contact angle analyzer (DSA₁₀₀, Germany).

2.4. Analytical methods

Cesium (I), strontium (II) and cobalt (II) concentrations were analyzed using a Thermo XII inductively coupled plasma-mass spectrometer based on general rules for inductively coupled plasma-atomic emission spectrometry (JY/T 015-1996). Boron was analyzed by UV-1800 spectrophotometer (SHIMADZU) based on the methylene

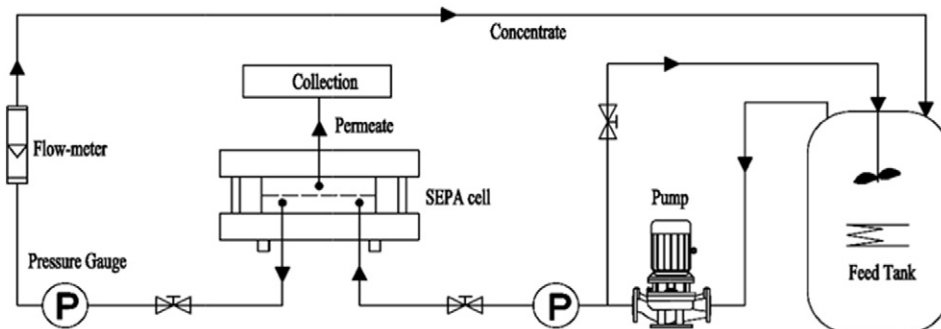


Fig. 1. Schematic of RO setup.

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