



## Enhanced rejection of cations by low-level cationic surfactant during ultrafiltration of low-level radioactive wastewater



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### ABSTRACT

The influence of cationic surfactant hexadecyl trimethyl ammonium bromide (CTAB) on the rejection of metal salts was investigated during ultrafiltration of low-level radioactive wastewater (LLRW). In the presence of CTAB below the critical micelle concentration (CMC), the rejection of nuclides significantly increased from 24%–33% to 50% for Cs(I) and above 90% for Sr(II), Co(II), and Ag(I). These increased rejections are attributed to CTAB fouling layer, which led to a more hydrophilicity and more positively-charged membrane surface. The nuclide content on the fouling layer increased with increasing CTAB concentration, and 55–80% of Co(II), Sr(II), and Ag(I) were adsorbed on the membrane at 400 mg/L CTAB. The high deposition of Ag(I) is partially due to the formation of AgBr. Based on these results, low-levels of cationic surfactant can significantly enhance the UF rejections of cations, but membrane fouling and nuclide deposition should not be ignored.

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## 1. Introduction

Nuclear power plants produce large quantities of hazardous low-level radioactive wastewater (LLRW), such as wastewater from laundry and floor sweepings [1]. LLRWs always contain trace levels of nuclides, such as Sr(II), Co(II), Cs(I), and Ag(I), and comparably higher concentrations of surfactants that are generated from the decontamination process. These surfactants can complex with ions and other dissolved pollutants which results in synergistic effects on the migration and dispersion of the pollutants [2,3]. Serious problems caused by surfactants have been found during the LLRW treatment processes, including foaming and entrainment during the evaporation process and serious membrane fouling during membrane treatments [4,5]. Membrane fouling leads to decreased membrane efficiency and shorter membrane lifetime. As membrane technologies have been increasingly used in LLRW treatment [6,7], a more detailed understanding of the influences of surfactants on membrane performance would be beneficial.

Anionic, cationic and nonionic surfactants are the three types of surfactants based on electronic charge and each surfactant influence membrane filtration in a different way. The surfactants can

adsorb onto the membrane surface or into the membrane pores through electrostatic effects or hydrophobic-hydrophilic surface interactions. This ultimately leads to membrane fouling [8–10], which can change the hydrophilic properties of the membrane surface and reduce the porosity via pore blocking [11,12]. Variations in membrane flux are a function of porosity reduction and hydrophilic modification of membrane surface [13]. Cationic surfactants have been reported to have a larger influence on the permeability of reverse osmosis and ultrafiltration (UF) membranes compared to anionic surfactants [5,11,14]. During ultrafiltration of anionic surfactant-containing LLRW, the flux varies depending on the concentration of surfactant and nuclides [4]. However, it is still unknown how the flux varies during the ultrafiltration of LLRWs that contain cationic surfactants.

In addition to the variation in flux caused by surfactants, UF membranes can also reject ions when surfactants are present. For example, micellar enhanced ultrafiltration (MEUF) can reject an extremely high number of dissolved metal ions and other pollutants [15,16]. Anionic surfactants are used to remove cationic metal ions such as Cd<sup>2+</sup> and Cu<sup>2+</sup> [17,18], and cationic surfactants are used to remove anionic ions such as perchlorate (ClO<sub>4</sub><sup>-</sup>) and chromate (CrO<sub>4</sub><sup>2-</sup>) [11,19]. The complexation of surfactants with pollutants plays an important role during these treatments. However, it is not known whether the presence of cationic surfactants can enhance the rejections of cation ions, since no complexation

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is formed. The presence of cationic surfactants may also influence the deposition behavior of nuclide ions and thereby affect the radiation level of the membrane waste.

In this work, we investigate the impact of cationic surfactants on membrane fouling and nuclide rejection during ultrafiltration of cationic surfactant-containing LLRW. Hexadecyl trimethyl ammonium bromide (CTAB) was used as the cationic surfactant at concentrations lower than the critical micelle concentration (CMC) value. The Sr(II), Co(II), Cs(I), and Ag(I) nuclide concentrations ranged from 0 to 5 mg/L. Flux, rejection and distribution of nuclides and membrane fouling were investigated under various conditions. The interactions between CTAB, the nuclides, and the membrane surface are discussed. Potential issues regarding the UF treatment of cationic surfactant-containing LLRW are revealed.

## 2. Experiment

### 2.1. Chemical reagents and membranes

The chemicals used in this study were of analytical grade and obtained from Beijing Chemical Works, China. Feed solutions were prepared by dissolving the required amount of the four salts (Sr(NO<sub>3</sub>)<sub>2</sub>, CsNO<sub>3</sub>, Co(NO<sub>3</sub>)<sub>2</sub>, and AgNO<sub>3</sub>) and CTAB in highly demineralized water (conductivity <1 μS/cm, pH = 7.4). The four salts of Sr(II), Co(II), Cs(I) and Ag(I) were added simultaneously into the solution, with concentrations of 0.1–5 mg/L. The concentration of CTAB ranged from 0 to 400 mg/L. A flat polyether sulfone (PES) UF membrane (SEPRO Membranes, Inc., California USA) with molecular weight cut off (MWCO) of 5 kDa and a contact angle of 92° was used after being soaked in deionized water for 24 h.

### 2.2. Experimental setup and design

Experiments were performed using a dead-end UF filtration cell (Amicon 8200, Millipore, Massachusetts USA) that was connected to a 4 L feed reservoir [20]. Feed solutions were pressurized from the reservoir to the UF cell under 0.1 MPa nitrogen gas at room temperature (25 °C). The permeate flux was minutely recorded using a balance that was connected to a computer, from which the flux were calculated. A new piece of UF membrane with an effective membrane filtration area of 28.7 cm<sup>2</sup> was used for each test. The pure water flux of each membrane was determined by filtering deionized water through the membrane until a stable permeate flux was obtained. The feed solution was then filtered through the membrane for 15 h, and the solution left in the filtration cell (ca. 200 mL) at the end of each experiment was considered as the concentrate. The concentrations of the nuclides and CTAB were obtained by sampling the filtrate collected in the first hour and at the end of the experiment, as well as the concentrate and the feed solution.

Four levels of CTAB concentration were used in the feed solutions (0, 50, 200 and 400 mg/L) and the concentrations of the nuclides (Sr(II), Co(II), Cs(I) and Ag(I)) ranged from 0 to 5 mg/L. The components of each feed solution are given in Table 1. A solution containing 1 mg/L nuclides means that Sr(II), Co(II), Cs(I) and Ag(I) concentrations are each 1 mg/L. Preliminary experiments were performed in duplicate to evaluate the reproducibility and the maximum error in the permeate flow rate was 5%.

### 2.3. Analytical methods

The Sr(II), Co(II), Cs(I) and Ag(I) ion concentrations were determined by inductively coupled plasma mass spectrometry (iCAP Q, ThermoFisher Scientific, Massachusetts USA). The concentration of

**Table 1**

Fouling resistance and contact angle of membrane fouled with various feed solutions.

Feed solution		R <sub>f</sub> (1/m)	Contact angle (°)
CTAB (mg/L)	Nuclide <sup>a</sup> (mg/L)		
0	1	-6.2 × 10 <sup>11</sup>	79 ± 3.4
50	1	-5.2 × 10 <sup>11</sup>	85 ± 2.0
200	1	2.9 × 10 <sup>12</sup>	89 ± 4.6
400	1	8.8 × 10 <sup>12</sup>	92 ± 1.6
200	0	-3.7 × 10 <sup>12</sup>	63 ± 4.0
200	0.1	2.2 × 10 <sup>12</sup>	73 ± 2.2
200	5	7.7 × 10 <sup>12</sup>	92 ± 1.2

<sup>a</sup> Nuclides included Sr(II), Co(II), Cs(I) and Ag(I), and the concentration of the four nuclides were all 0–5 mg/L.

CTAB was determined by total organic carbon (TOC) analysis (Shimadzu Corporation, Japan).

The surface morphologies and elemental compositions of the freeze-dried fouled membranes were monitored using scanning electron microscopy and an energy dispersive spectrometer (SEM/EDS, Sirion 200, FEI Inc., Maryland USA) at an accelerating voltage of 10.0 kV. The contact angle of the membrane and the surface tension of the CTAB solution was measured using a contact angle analyzer (DSA100, Hamburg Germany).

### 2.4. Data analysis

The membrane flux ( $J$ , m<sup>3</sup>/(m<sup>2</sup> s)) was calculated as

$$J = \frac{\Delta m / 60}{\rho \cdot A \cdot 100} \quad (1)$$

where  $A$  is the effective membrane area (cm<sup>2</sup>),  $\rho$  is the solution density (1 g/cm<sup>3</sup>), and  $\Delta m$  is the effluent mass per minute (g/min).

A normalized flux was calculated in order to eliminate the differences of membrane samples from the ratio of the feed solution flux ( $J$ , m<sup>3</sup>/(m<sup>2</sup> s)) to the pure water flux ( $J_0$ , m<sup>3</sup>/(m<sup>2</sup> s)).  $J_0$  was around  $7 \times 10^{-6}$  m<sup>3</sup>/(m<sup>2</sup> s).

The fouling resistance ( $R_f$ , 1/m) was calculated as

$$R_f = \frac{\Delta P}{\eta J} - R_m \quad (2)$$

where  $\Delta P$  is equal to the transmembrane pressure (N/m<sup>2</sup>),  $\eta$  is the kinetic viscosity of the solution (N s/m<sup>2</sup>), and  $R_m$  is the membrane resistance (1/m) [21].

The rejection of nuclides and CTAB ( $R$ , %) was calculated by

$$R = 100 \cdot \left( 1 - \frac{c_1}{c_0} \right) \quad (3)$$

where  $c_0$  and  $c_1$  are the concentrations of the nuclide ions or CTAB in the feed water and the effluent during the first hour (mg/L for CTAB and μg/L for nuclides), respectively.

A difference analysis for these calculations was conducted using a Student's  $t$ -test.

## 3. Results and discussion

### 3.1. Flux variation

The initial values and the variations over time of the UF flux varied with the feed solutions (Fig. 1). In Fig. 1A, the feed solutions contained 1 mg/L nuclides and different concentrations of CTAB, and the UF flux varied with CTAB concentration. Among runs when the feed solutions contained CTAB of 0 and 50 mg/L, the flux remained constant. This indicates that 1 mg/L of Sr(II), Co(II), Cs(I) and Ag(I) and 50 mg/L of CTAB did not cause serious membrane fouling. For the filtration of the feed solution containing 200 mg/L CTAB, the normalized flux was 72% at the beginning of filtration,

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