



Original Article

Fouling and cleaning protocols for forward osmosis membrane used for radioactive wastewater treatment

Xiaojing Liu ^{a, b}, Jinling Wu ^a, Li-an Hou ^c, Jianlong Wang ^{a, b, *}^a Collaborative Innovation Center for Advanced Nuclear Energy Technology, INET, Tsinghua University, Beijing, 100084, PR China^b Beijing Key Laboratory of Radioactive Waste Treatment, Tsinghua University, Beijing, 100084, PR China^c Xi'an High Technology Institute, Xi'an, 710025, PR China

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ABSTRACT

The membrane fouling is an important problem for FO applied to the radioactive wastewater treatment. The FO fouling characteristics for simulated radioactive wastewater treatment was investigated. On-line cleaning by deionized (DI) water and external cleaning by ultrasound and HCl were applied for the fouled membrane. The effectiveness and foulant removing amount by each-step cleaning were evaluated. The membrane fouling was divided into three stages. Co(II), Sr(II), Cs(I), Na(I) were all found deposited on both active and support layers of the membrane surface, resulting in membrane surface became rougher and more hydrophobic, which increased membrane resistance. On-line cleaning by DI water recovered the water flux to 69%. HCl removed more foulants than ultrasound.

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1. Introduction

The radioactive wastewater, which contains various radionuclides (such as ⁶⁰Co, ¹³⁷Cs, ⁹⁰Sr, etc), is harmful to environment and human health. Various technologies have been applied for the treatment of radioactive wastewater, such as precipitation, adsorption, membrane separation and the like [1–5]. Forward osmosis (FO) has the advantage of high rejection, high water flux and low membrane fouling, which is a potential effective process for the treatment of the radioactive wastewater [5]. In our previous study, FO was found effective to separate Co(II) from the aqueous solution. Cellulose triacetate with embedded polyester screen support (CTA-ES) membrane featured the highest water fluxes among the three selected commercial FO membranes. The optimal operation conditions were NaCl concentration of 1.0 M in draw solution (DS), the cross-flow velocities of 11 and 5 cm s^{−1} at the feed solution (FS) and DS sides respectively. However, Co(II) was found on the fouled membrane surface and became the membrane foulants [6], which affects the nuclide retentions. Therefore, membrane fouling is an important problem for FO applied to the radioactive wastewater treatment.

Although FO membrane fouling for the radioactive wastewater treatment is less studied so far, membrane fouling of FO applied for other wastewater treatments was much investigated. Like other membrane technologies, membrane fouling during FO process is inevitable, which involves colloidal fouling, organic fouling, inorganic scaling and biofouling [7]. In the last decade, organic fouling during FO process has been extensively investigated [8–11], which has many similarities with colloidal fouling and can be explained by similar fouling mechanisms [7]. Organic fouling and colloidal fouling are caused by macromolecules and colloidal particles from the influent. The foulants aggregation and/or attachment on the membrane through van der Waals and electrical double layer force [7]. Inorganic scaling is a problem especially for wastewater reclamation and brackish water desalination [7,12–14]. Scaling is caused by the sparingly soluble salts, such as CaSO₄, BaSO₄, and CaCO₃. These salts precipitate near or on the membrane surface when the local concentration is higher than their solubility [15]. Biofouling is complicated and mainly attributed to bacterial adhesion onto the membrane surface to form biofilm [16].

No matter what kind of fouling, it affects the performance of FO. Membrane fouling could reduce the permeate water flux [17]. The foulants deposited on the membrane surface or within the support layer (SL), which made the overall membrane resistance for water transport increased and the extent of concentration polarization changed [7]. The fouling-enhanced concentration polarization and

* Corresponding author. Energy Science Building, Tsinghua University, Beijing, 100084, PR China.

E-mail address: wangjl@tsinghua.edu.cn (J. Wang).

internal concentration polarization self-compensation effect also declined the water flux. Membrane fouling not only affects the water flux, but influences the membrane rejection property [18]. Fouling either enhanced or reduced the contaminants rejection because of the different rejection mechanisms [7]. The increased rejection of contaminants (arsenite [19], trace organic compounds [20,21], micropollutants [18], etc.) was attributed to the membrane fouling enhancing the size sieving, electrostatic repulsion and hydrophobic interaction between contaminants and membrane. However, the decreased rejection of trace contaminants (boron, hydrophilic neutral contaminants) was caused by the fouling-enhanced concentration polarization, which increased contaminants concentration near the membrane surface and eventually enhanced the contaminants diffusion through the membrane [7,18,19].

Membrane fouling in FO process is influenced by operational conditions, influent characteristics, DS composition, membrane properties and membrane orientation [7]. The operational conditions include cross flow velocity [11,22,23] and temperature [13,24], etc. Influent characteristics contain influent composition [9,25] and concentration [26,27], physicochemical properties (e.g. pH [28], ionic strength, etc.) [9]. In addition, the composition and concentration of DS can influence the membrane fouling by affecting the initial water flux [29]. Membrane properties consist of membrane materials, membrane surface properties (surface functional group, hydrophilicity/hydrophobicity, roughness, charge density, etc.), and structural properties [8]. Membrane orientation has an impact on internal concentration polarization, which affects the initial water flux [25,30,31] and then influences the extent of the membrane fouling.

Considering fouling is inevitable, membrane cleaning is required including physical cleaning, chemical cleaning and biological cleaning [7]. Physical cleaning can remove the deposited foulants on the membrane surface by enhancing shear force [8,26,32]. The water flux was even recovered close to 100% by simple surface flushing [8]. When FO was used to treat complex wastewater, chemical cleaning is also required because of the irreversible fouling caused by the complicated foulants from the wastewater [11,33]. The typical cleaning chemicals include alkali (NaOH), acids (citric acid, nitric acid, etc.), chelating agents (EDTA) [11], oxidants (NaClO [16], H₂O₂, peroxyacetic acid, etc.), surfactants (detergents) and so on [15]. The recovery efficiency of water flux was affected by different cleaning chemicals [33]. The chemical agents of alkali, oxidants and surfactants are generally used to remove the organic fouling. Acids and chelating agents have advantages in removing the inorganic scaling. For biological cleaning, bioactive agents are applied to remove the foulants [7,34,35]. In fact, every cleaning method has its limitations. Generally, it is necessary to establish a combined cleaning protocol to overcome their limitations and mitigate the membrane fouling.

When the FO process is specially applied for the radioactive wastewater treatment, membrane fouling characteristics would be significantly different from that for the general wastewater treatment. Firstly, in some radioactive wastewater, inorganic substances are the major parts instead of organics. Fouling here was mostly attributed to the inorganic substances. However, the effect of

inorganic fouling on the FO performance was not investigated as fully as that of organic fouling. Secondly, inorganic fouling, especially the fouling caused by radionuclides would make the membrane module radioactive. The used membrane when disposed might become new solid radioactive wastes. Thirdly, which nuclides are prone to attach on the membrane and become the foulants is unclear. The proper cleaning protocols have to be proposed on the basis of this question. As a result, the characteristics of fouling during FO process applied for the radioactive wastewater should be investigated in details for answering all the above questions. An effective cleaning method should be proposed to reduce fouling and facilitate the disposal of radioactive wastes.

Aiming at these questions, in this study, fouling and cleaning for the CTA-ES membrane used in FO process for radioactive wastewater treatment were studied. The fouling characteristics were investigated during 30 d FO operation. The combined cleaning protocols involved on-line cleaning by deionized (DI) water streams and external cleaning by ultrasound and HCl were proposed. The effectiveness of the various cleaning steps were compared and discussed. In addition, the residual foulants on the membrane surface after each cleaning step were analyzed.

2. Materials and methods

2.1. Chemicals, feed and draw solutions

All the chemicals, including CoCl₂, SrCl₂, CsCl, NaCl and HCl, used in this experiment were of analytical reagent grade. FS was simulated radioactive wastewater and prepared by dosing the following chemicals successively to DI water: 20 mg L⁻¹ CoCl₂, 20 mg L⁻¹ SrCl₂ and 20 mg L⁻¹ CsCl. DS was prepared by dissolving NaCl salt in DI water.

2.2. FO membrane and characterization

A commercial flat-sheet CTA-ES membrane was purchased from Hydration Technologies Innovations (HTI). According to the manufacturer, the membrane has asymmetric structure with embedded polyester screen support, containing a thin active layer (AL) and a thick mechanical support layer (SL) [36–38]. The operating pH of membrane was 3–8. The membranes were soaked in DI water for 18 h before use to remove the glycerin on the surface of membrane. The characteristic parameters of CTA-ES membrane are summarized in Table 1.

The original membrane, fouled membrane, cleaned membrane were observed and further analyzed by a field emission scanning electronic microscope (SEM) (QUANTA 200 FEG, FEI, USA) attached by an energy dispersive spectrometer (EDS) (Oxford Inca EDX 300). The detailed structure of the AL of the membrane was observed by an atomic force microscope (AFM) (Dimension, Bruker AXS, Germany). The roughness was evaluated by three parameters, including mean-square surface roughness (R_q), mean surface roughness (R_a) and maximum vertical distance between the highest and lowest data points (R_{max}). Contact angle of membrane was analyzed by a goniometer with the sessile drop method (Contact Angle System OCA20, Data Physics Instruments GmbH, Germany).

Table 1
Changes of the membranes properties.

Roughness (nm)				Contact angle(°)	
	R_q	R_a	R_{max}	AL	SL
Original	5.2	4.1	41.8	65.6 ± 4.5	72.9 ± 4.0
Fouled	253	194	1587	84.0 ± 1.3	101.9 ± 3.2
Cleaned	3.81	2.85	45.8	79.0 ± 5.0	99.5 ± 10.2

Membrane samples were dried by vacuum freeze dryer (LGJ-18, China) for 24 h prior to measurement.

2.3. FO system

The fouling and cleaning experiments were carried out with a bench-scale FO system. The schematic diagram of FO system is shown in Fig. 1. The membrane cell with two flow channels of 2 mm in height was separated by FO membrane. The effective area of membrane was 40.5 cm². Two gear pumps (BT600-2J, China) were used for FS and the DS circulation. Feed and draw streams flowed countercurrently in each channel at the flow velocity of 11 cm s⁻¹. The FO process was operated with the AL of the membrane facing the FS. All the FO experiments were conducted in an air-conditioned room with the temperature of 25 ± 2 °C. Digital balance (ML6001, Mettler Toledo, Shanghai, China) and data logging system recorded the weight change of the FS as a function of time to determine the water flux. Samples from FS and DS were taken to analyze the concentration of Co(II), Sr(II), Cs(I) and Na(I).

The fouling experiment was carried out with the FO system operated under the optimal conditions (DS of 1.0 M NaCl, FS and DS flow velocities of 11 cm s⁻¹) for 30 d (FO I) [5]. FS contains 20 mg L⁻¹ CoCl₂, 20 mg L⁻¹ SrCl₂ and 20 mg L⁻¹ CsCl. There was no other foulants to be added in FS. The fouled membrane (Membrane I) was taken to analyze the fouling characterization.

The cleaning experiment was conducted with the FO system after 14 d operation (FO II). The fouled membrane (Membrane II) was on-line cleaned with DI water recycling by both FS and DS sides of the membrane at the flow velocity of 11 cm s⁻¹. For external cleaning, ultrasonic and HCl cleaning were successively employed as follows: 1) The Membrane II was soaked in DI water (60 mL) for 2 h with ultrasonic cleaning at 600 W power (KQ5200DB, Kunshan, China). 2) The Membrane II was soaked in HCl solution (55 mL, pH ~3) for 2 h.

2.4. Indicators of FO performance

The water flux, the nuclide retentions, and the reverse Na(I) flux were the indicators of FO performance. Effects of fouling and cleaning on FO performance were investigated in terms of water flux, nuclide retentions and the reverse Na(I) flux.

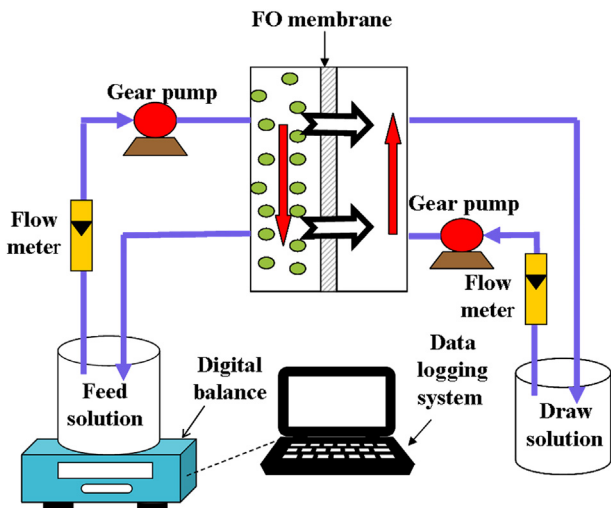


Fig. 1. The schematic diagram of the FO system.

2.4.1. Water flux

Water flux can be calculated by equation (1) [5,36,37]:

$$J_W = \Delta m / (\rho \cdot A \cdot \Delta t) \quad (1)$$

where A (m²) is the effective area of membrane, Δt (h) is the measuring time interval, ρ (g cm⁻³) is the density of water. Δm (g) is the measured permeated water weight interval from the FS to the DS.

2.4.2. Nuclide retentions

The retention of each nuclide ion is determined by equation (2) [5,36]:

$$R(\%) = (1 - C_{f,DS} V_{f,DS} / V_p C_{i,FS}) \times 100 \quad (2)$$

where $V_{f,DS}$ (mL) and V_p (mL) are the final volume of DS and permeated water, respectively. $C_{f,DS}$ (mg L⁻¹) is the final concentration of nuclides ions in DS, $C_{i,FS}$ (mg L⁻¹) is the initial nuclide ions concentration of FS.

2.4.3. Reverse Na(I) flux

The reverse Na(I) flux can be defined as(3):

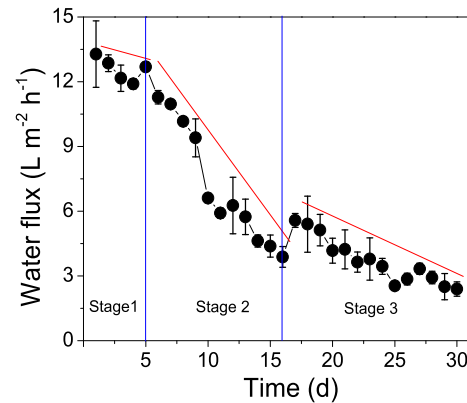


Fig. 2. Effect of membrane fouling on water flux. The experimental conditions were as follows: the FS was 20 mg L⁻¹ nuclides solution, the DS was 1 M NaCl solution, the flow velocity was 11 cm s⁻¹ on FS and DS sides, and the temperature was 25 ± 2 °C for both sides.

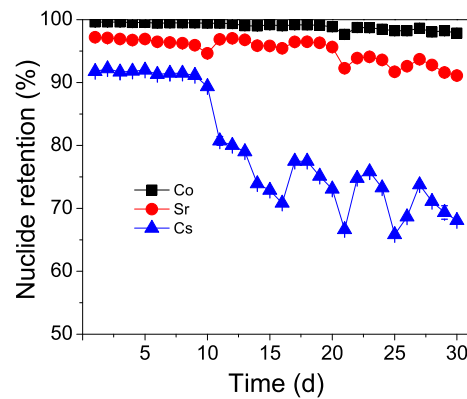


Fig. 3. Effect of membrane fouling on nuclide retentions. The experimental conditions were as follows: the FS was 20 mg L⁻¹ nuclides solution, the DS was 1 M NaCl solution, the flow velocity was 11 cm s⁻¹ on FS and DS sides, and the temperature was 25 ± 2 °C for both sides.

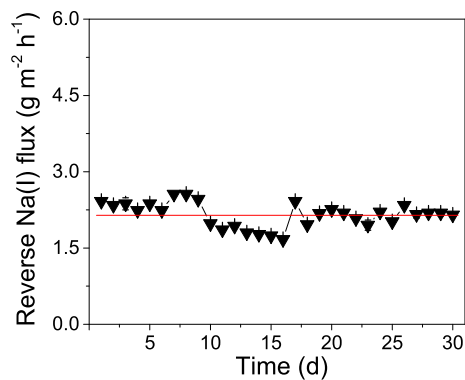


Fig. 4. Effect of membrane fouling on reverse Na(I) flux. The experimental conditions were as follows: the FS was 20 mg L⁻¹ nuclides solution, the DS was 1 M NaCl solution, the flow velocity was 11 cm s⁻¹ on FS and DS sides, and the temperature was 25 ± 2 °C for both sides.

$$J_{\text{NaCl}} = (C_{f,\text{FS}}V_{f,\text{FS}} - C_{i,\text{FS}}V_{i,\text{FS}}) / A\Delta t \quad (3)$$

where $V_{i,\text{FS}}$, $V_{f,\text{FS}}$ (mL) are the initial and final volumes of FS, respectively; $C_{i,\text{Na,FS}}$, $C_{f,\text{Na,FS}}$ (mg L⁻¹) are the initial and final Na(I) concentration in FS, respectively.

2.5. Analytical methods

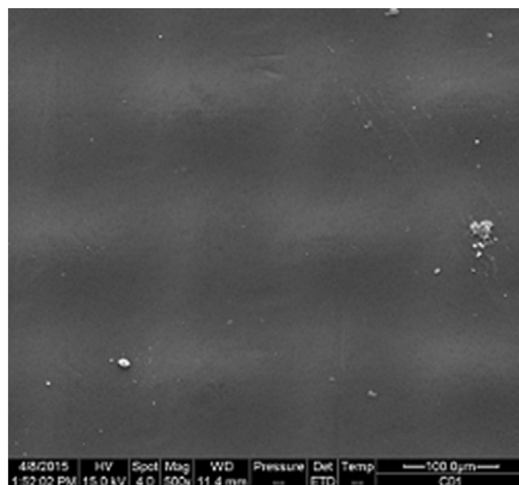
The concentrations of Co(II), Sr(II) and Cs(I), Na(I) in FS and DS were determined by atomic absorption spectrometry (ZA3000, HIT, Japan).

3. Results and discussion

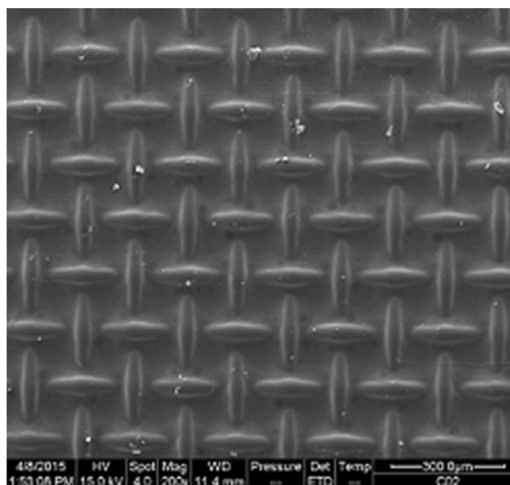
3.1. Fouling characteristics

3.1.1. Decline of water flux

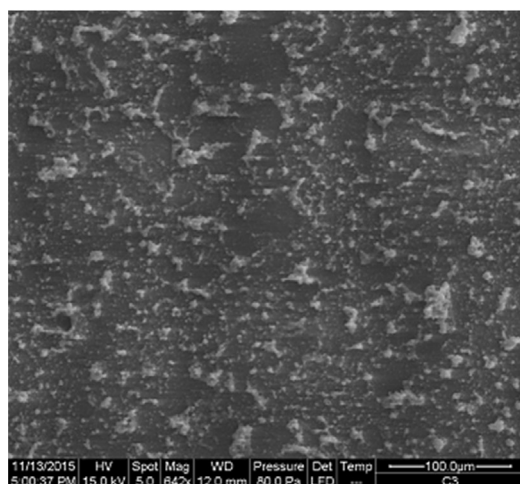
The water flux of Membrane I was investigated during the 30 d



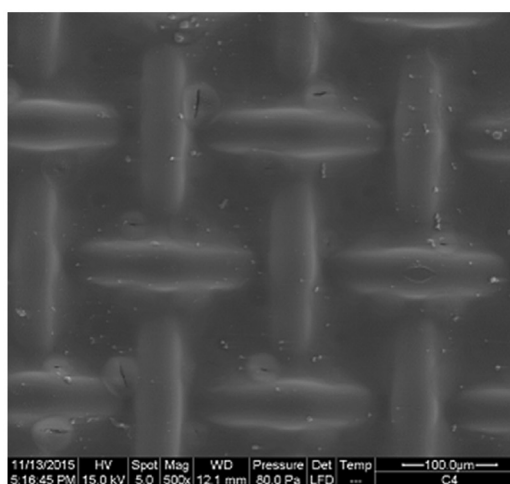
(a)



(b)



(c)



(d)

Fig. 5. SEM images of the AL (a) and the SL (b) of the original membrane; the AL (c) and the SL (d) of the fouled membrane.

operation, which is shown in Fig. 2. The decrease of the water flux could be divided into three stages: Stage 1: a gradual decrease, Stage 2: a sharp decline and Stage 3: a slight decrease. A gradual decrease (from 13.3 to 12.7 L m⁻² h⁻¹) in water flux was found in the first 5 d. The gradual decline could be attributed to the decrease in osmotic driving force with time because of dilution of the DS and concentration of the FS [12]. Then, a sharp decline (from 12.7 to 3.9 L m⁻² h⁻¹) in water flux followed during the following 11 d, which suggested that membrane fouling occurred and fast developed. After about 16 d of operation, the water flux declined slowly from 3.9 to 2.4 L m⁻² h⁻¹ for 14 d. The slow decline of flux indicated a slow developed fouling process. With the development of fouling, the foulants increased the membrane resistance (Please see 3.1.4 in details), which hindered the water molecules through membrane from the FS to DS. In addition, more and more nuclide ions were rejected by membrane as the operation time, which eventually enhanced the osmotic pressure near the membrane surface at FS side. The enhanced osmotic pressure at FS side reduced the effective osmotic pressure of membrane on both sides. Thereby a slight decline in water flux occurred [39].

3.1.2. Decline of nuclide retentions

The effect of membrane fouling on nuclide retentions was

investigated. Fig. 3 shows the nuclide retentions with time. Although severe fouling occurred, the retention of Co(II), Sr(II), Cs(I) maintained higher than 65% during the whole operation time. During the process, the retention of Co(II) and Sr(II) were always higher than Cs(I), because Co(II) and Sr(II) possessed larger hydration radius than Cs(I) and had more difficulties to pass the membrane [40]. The Co(II) retention and Sr(II) retention were 97.7–99.6% and 91.1–97.2%, respectively. Only a slight decrease of Co(II) and Sr(II) retention were observed during the whole operation.

Unlike Co(II) and Sr(II), the retention of Cs(I) changed a lot with fouling occurred. The retention of Cs(I) was initially 92.2%, but declined to 65.8% after 30 d. Fouling-enhance concentration polarization could be an important reason. More and more Na(I) attached on the membrane surface of AL with time (3.1.4 in details), which increased the reverse transfer of Na(I) from DS to FS (3.1.3 in details). The CTA-ES membrane was of negative charge, which worked as a cation exchange membrane. Therefore, more and more nuclide cations transferred from FS to DS to keep the electric balance [40,41]. Among the three cations, Cs(I) with the smallest hydrated ionic radius and lowest charge was easier to be exchanged by Na(I).

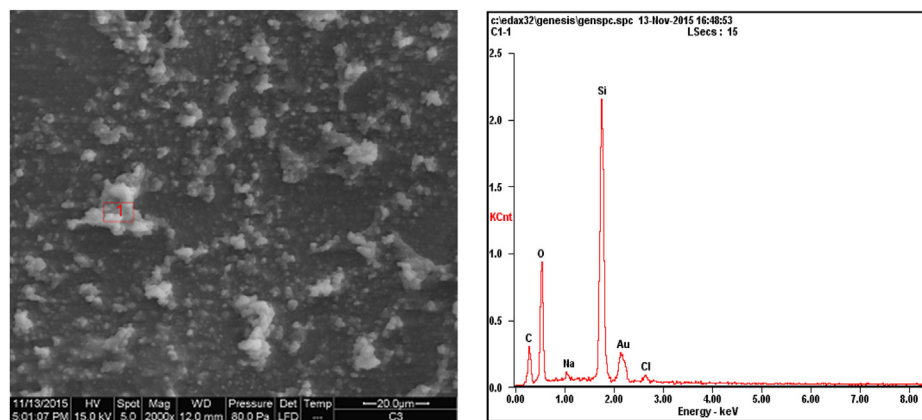


Fig. 6. SEM image and EDS spectrum of the AL of fouled membrane.

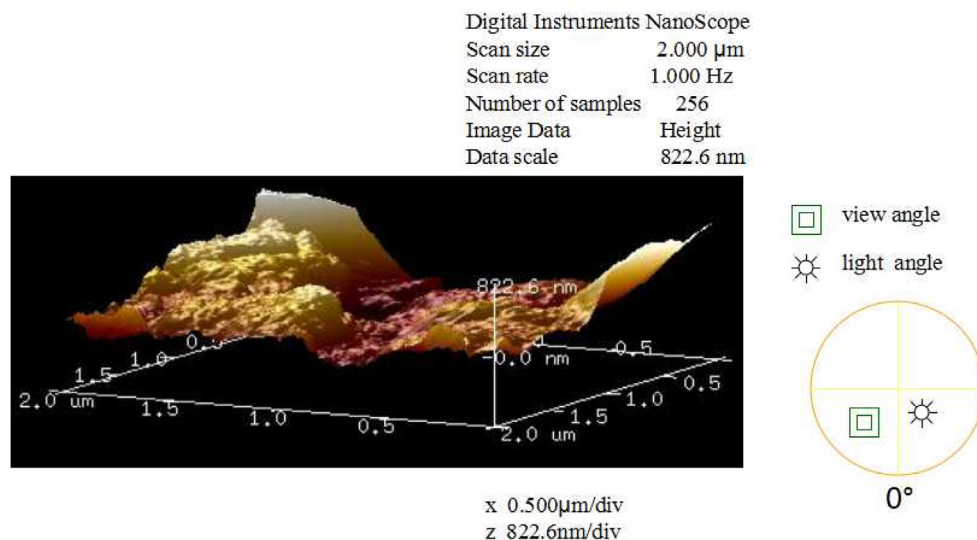


Fig. 7. AFM image of the AL of fouled membrane.

3.1.3. Increase of reverse Na(I) flux

The reverse Na(I) flux, shown in Fig. 4, increased slightly from 1.67 to 2.56 g m⁻² h⁻¹ during the entire operation time, indicating that the reverse permeation of Na(I) enhanced. It was related to the continuously-increased amount of Na(I) accumulated on the membrane surface due to the fouling-enhanced concentration polarization, which was proved in section 3.1.4. The increased Na(I) concentration on the membrane surface increased reverse flux of Na(I) with the smaller hydrated radii (see Fig. 5).

3.1.4. Characterization of fouled membrane

From the SEM images, it can be seen that some crystals attached on membrane surface, especially the surface of AL (Fig. 5a–d). The composition of foulants was analyzed by EDS (Fig. 6). Si, Na, Cl were found deposited on the membrane. Si element was from natural water. Na(I) and Cl(I) were the major foulants, which was consistent with the ion concentration in cleaning solutions (refer to section 3.3). The accumulation of NaCl on the membrane was the reason for the increase of reverse Na(I) flux and the decline of Cs(I) retention. The CTA-ES membrane was of negative charge, which worked as a cation exchange membrane [40,41]. A great quantity of Na(I) increased the reverse transfer of Na(I) from DS to FS. For the electric balance, more nuclide cations transferred from FS to DS, which caused the decline of the nuclide retentions. Cs(I) retention reduced faster than Co(II) and Sr(II) retentions because Cs(I) featured the smallest hydrated ionic radius and lowest charge, which made the Cs(I) easier to be exchanged by Na(I) to keep the electric balance.

Within the selected area observed in SEM images, none of the nuclides ions including Co(II), Sr(II) and Cs(I) were found on the either surface of the fouled membrane. It suggested that the concentration of three nuclides were much lower than NaCl concentration on the membrane surface.

The AL of fouled membrane became rougher with R_{\max} value increased from 41.8 to 1587 nm (Fig. 7, Table 1). The deposition of foulants on the membrane surface increased the membrane roughness. The contact angle of AL increased from 65.6 to 84.0° and that of SL increased from 72.9 to 102.9°, which indicated that the fouled membrane became more hydrophobic (Table 1), so that the membrane resistance increased and then the water flux reduced.

3.1.5. Foulant amounts

The foulants accumulated on the membrane were evaluated by the total amounts of the ions removed from the following cleaning steps. The total amount of Co(II), Sr(II), Cs(I), Na(I) were 53.2, 52.2, 45.5, 27100 mg m⁻², respectively (Table 2). It is observed that Na(I) is the major foulant for the membrane fouling. Cs(I) amount is lower than Co(II) and Sr(II) since Cs(I) with smaller hydrated ionic radius and lower charge is easier to go through the membrane into the DS.

3.2. On-line cleaning by DI water

The fouled Membrane II was subjected to on-line cleaning by DI water after 14 d operation when the water flux rapidly reduced at the end of Stage 2 (Fig. 2).

Table 2
Cleaning efficiency of various cleaning protocols.

Elements	Foulant amount removed from per membrane area (mg·m ⁻²)				
	DI water		Ultrasound	HCl	Total
	AL	SL			
Co	51.5	1.17	0.15	0.43	53.2
Sr	47.5	2.32	1.16	1.22	52.2
Cs	42.6	2.37	0.07	0.46	45.5
Na	1990	24500	607	26.3	27100

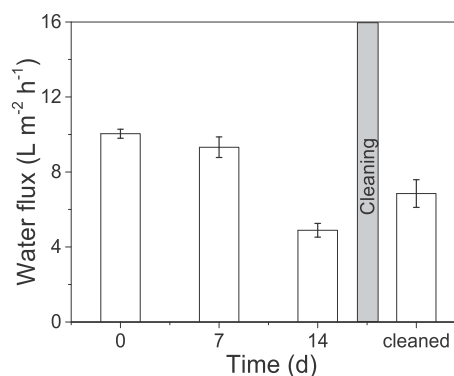


Fig. 8. Water flux before and after cleaning. The experimental conditions were as follows: the FS was 20 mg L⁻¹ nuclides solution, the DS was 1 M NaCl solution, the flow velocity was 11 cm s⁻¹ on FS and DS sides, and the temperature was 25 ± 2 °C for both sides.

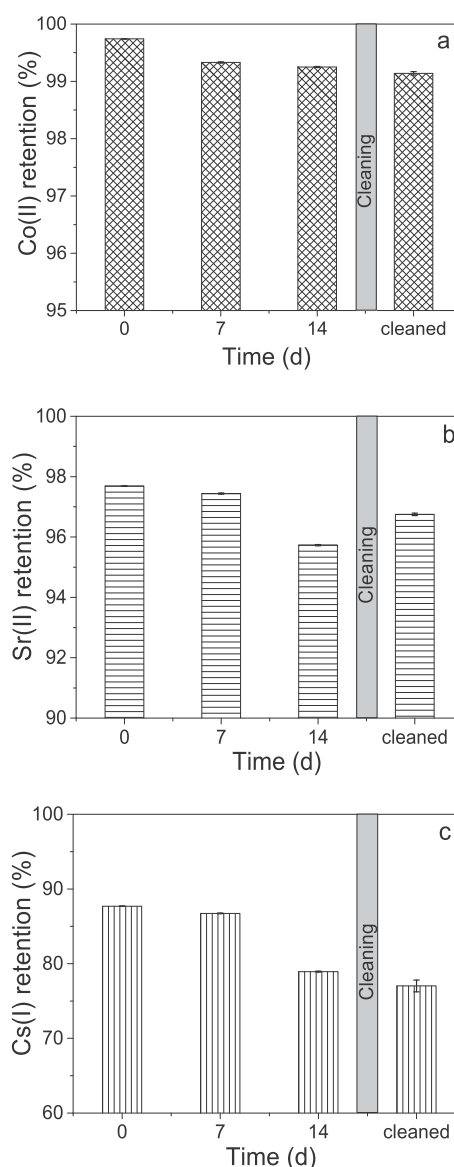


Fig. 9. Co(II) retention (a), Sr(II) retention (b) and Cs(I) retention (c) before and after cleaning. The experimental conditions were as follows: the FS was 20 mg L⁻¹ nuclides solution, the DS was 1 M NaCl solution, the flow velocity was 11 cm s⁻¹ on FS and DS sides, and the temperature was 25 ± 2 °C for both sides.

3.2.1. Recovery of water flux

The efficiency of on-line cleaning on the recovery of water flux was shown in Fig. 8. The water flux decreased from 10.0 to 4.9 L m⁻² h⁻¹. After on-line cleaning by DI water, the water flux recovered from 4.9 to 6.9 L m⁻² h⁻¹. The water flux was recovered to 69%. Therefore, the on-line cleaning by only DI water was not enough to obtain a good flux recovery.

3.2.2. Change of nuclide retentions and reverse Na(I) flux

The changes of nuclide retentions after on-line cleaning by DI water were shown in Fig. 9. The Co(II) and Cs(I) retentions were decreased by 0.11% and 1.91% respectively. Sr(II) retention increased from 95.7% to 96.8%. The changes were small and could be almost neglected.

The reverse Na(I) flux after on-line cleaning was investigated (Fig. 10). The reverse Na(I) flux was slightly increased from 1.57 to 1.65 g m⁻² h⁻¹.

3.2.3. Characterization of on-line cleaned membrane

After on-line cleaning, a great quantity of crystals were effectively removed from both AL and SL, which resulted in the roughness of the membrane significantly recovered from 1587 to 45.8 nm (Fig. 11, Table 1). But there were still some remnants attached to the

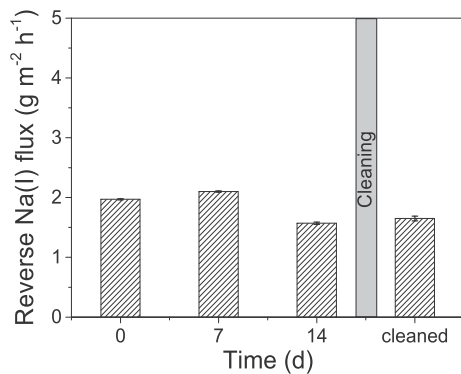


Fig. 10. Reverse Na(I) flux before and after cleaning. The experimental conditions were as follows: the FS was 20 mg L⁻¹ nuclides solution, the DS was 1 M NaCl solution, the flow velocity was 11 cm s⁻¹ on FS and DS sides, and the temperature was 25 ± 2 °C for both sides.

membrane surface (Fig. 12a and b). This result indicated that on-line cleaning by DI water is not sufficient to remove all the fouling layers deposited on the FO membrane. Other cleaning methods are needed to further remove the irreversible fouling and restore the water flux.

3.2.4. Fouling removed by on-line cleaning

Great amount of the foulants were removed by on-line cleaning, especially the Na(I). The foulant amounts removed from AL per membrane area were 51.5 mg m⁻² of Co(II), 47.5 mg m⁻² of Sr(II), 42.6 mg m⁻² of Cs(I) and 1990 mg m⁻² of Na(I). The amounts of Co(II), Sr(II), Cs(I), Na(I) removed from SL per membrane area were 1.17, 2.35, 2.37, 24500 mg m⁻², respectively (Table 2). The nuclides from AL were much more than those from SL because the FS faced the AL. A great quantity of NaCl was also removed from AL, which indicated that the reverse transferred NaCl also accumulated on AL and became the foulants. The nuclides did attach on both sides of membrane although they were not found by SEM-EDS.

3.3. External cleaning

After on-line cleaning, Membrane II was cleaned by another two-step external cleaning methods. The two-step external cleaning is HCl cleaning followed by ultrasound. The cation concentrations in the cleaned solution from each step were examined to investigate the cleaning effectiveness and evaluate the foulants amount on fouled membrane surface.

The amounts of Co(II), Sr(II) and Cs(I) removed by ultrasound were 0.15, 1.16, 0.07 mg m⁻², respectively (Table 2). The amounts

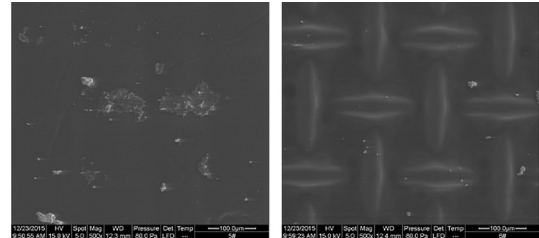


Fig. 12. SEM images of the AL (a) and the SL (b) of the cleaned membrane.

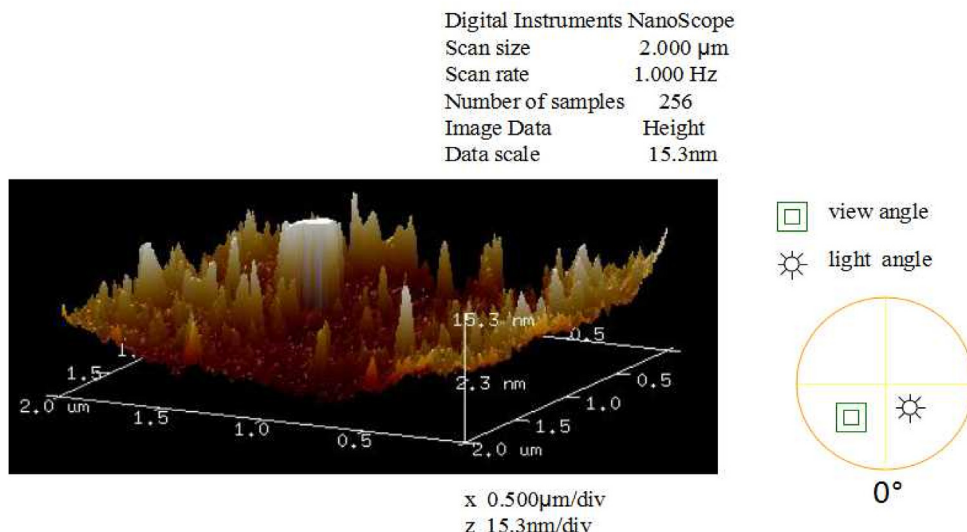


Fig. 11. AFM image of the AL of cleaned membrane.

removed by HCl were 0.43, 1.22, 0.46 mg m⁻² (Table 2), respectively. HCl removed more foulants than ultrasound. Among the three nuclides, Sr(II) amount removed from the membrane by the external cleaning was the highest, which suggested that Sr(II) played a more important role in fouling among the three nuclides. Sr(II) and Ca(II) are the same main group elements in the chemical periodic table and have similar chemical properties. The presence of Ca(II) generally enhanced the membrane fouling by directly depositing and improving the intermolecular adhesion between foulants [9,27]. Therefore, it is reasonable that Sr(II) has a similar fouling effect with Ca(II), which was more prone to foul the membrane than Co(II) and Cs(I).

Although a great quantity of foulants had been removed, the recoveries of nuclide retentions and water flux were insignificant. Therefore, more effective cleaning protocols are required to be proposed in FO process used for radioactive wastewater in the future.

4. Conclusions

This study investigated the fouling and cleaning protocols for CTA-ES FO membrane used for radioactive wastewater treatment. Membrane fouling during FO process for radioactive wastewater was divided into three stages according to the decrease rate of water flux: Stage 1 with slow declined flux, Stage 2 with rapid declined flux and Stage 3 with slightly declined flux. Co(II), Sr(II), Cs(I), Na(I) were all found deposited on both AL and SL of the membrane surface, resulting in membrane surface became rougher and more hydrophobic, which increased membrane resistance. Sr(II) was more prone to foul the membrane than Co(II) and Cs(I). On-line cleaning by DI water recovered the water flux to 69%. HCl removed more foulants than ultrasound.

Further study is needed to explore the fouling mechanisms and propose more effective cleaning protocols for FO process applied of radioactive wastewater treatment.

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