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Treatment of Medical Radioactive Liquid Waste Using Forward Osmosis (FO) Membrane Process

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Abstract

The use of forward osmosis (FO) for concentrating radioactive liquid waste from radiation therapy rooms in hospitals was systematically investigated in this study. The removal of natural and radioactive iodine using FO was first investigated with varying pHs and draw solutions (DSs) to identify the optimal conditions for FO concentration. Results showed that FO had a successful rejection rate for both natural and radioactive iodine (¹²⁵I) of up to 99.3%. This high rejection rate was achieved at a high pH, mainly due to electric repulsion between iodine and membrane. Higher iodine removal by FO was also attained with a DS that exhibits

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a reverse salt flux (RSF) adequate to hinder iodine transport. Following this, actual radioactive medical liquid waste was collected and concentrated using FO under these optimal conditions. The radionuclides in the medical waste (^{131}I) were removed effectively, but the water recovery rate was limited due to severe membrane fouling. To enhance the recovery rate, hydraulic washing was applied, but this had only limited success due to combined organic-inorganic fouling of the FO membrane. Finally, the effect of FO concentration on the reduction of septic tank volume was simulated as a function of recovery rate. To our knowledge, this study is the first attempt to explore the potential of FO technology for treating radioactive waste, and thus could be expanded to the dewatering of the radioactive liquid wastes from a variety of sources, such as nuclear power plants.

Graphical abstract

Keywords: Forward osmosis; Radioactive liquid waste; Wastewater treatment; Membrane fouling; Radioactive iodine

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1. Introduction

Radioactive materials, also known as radionuclides or radioisotopes, are routinely used in beneficial applications such as electricity generation, genetic modification, non-destructive inspection, and radiation medical therapy [1]. Radioactive material consists of various radioisotopes emitting α , β and γ radiation, which has a seriously negative impact on humans, animals, and plants [2, 3]. For these reasons, the leakage of radioactive material is a critical risk for human health and the environment. Radioactive liquid waste is also generated from the use of radioactive material, and this should be treated to prevent or mitigate harmful impacts, using treatments such as ion exchange [4], membrane distillation [5], reverse osmosis [6], and solvent extraction [7].

In hospitals, radionuclides (e.g., ^{90}Y , ^{125}I , ^{131}I , ^{89}Sr , ^{192}Ir , ^{60}Co , and ^{137}Cs) are utilized to examine and treat cancer patients by shrinking tumors and killing cancer cells [8, 9]. Radiation therapy rooms should be isolated to protect people from undesired radiation exposure. However, because wastewater containing radioactive substances (e.g., urine, feces, and detergent) is produced within radiation therapy rooms, it must be stored in separate septic tanks so that the radioactive material can decay to meet discharge regulations; this requirement limits the number of radiation therapy rooms that can be in operation [10].

Because the use of radionuclides can generate a significant amount of radioactive liquid waste, which requires large storage tanks, concentrating this waste can be beneficial in that it will reduce management costs. Only a limited number of studies have been carried out on the treatment of medical radioactive liquid waste arising from radioimmunoassays (RIAs), which are an in vitro assay technique used to measure the concentration of antigens [11]. RIA waste is composed of various kinds of proteins because they are generated from analysis reagents and samples [12]. Membrane technologies, such as the ultrafiltration–reverse osmosis (UF-

RO) hybrid system, have been considered for the treatment of RIA waste containing ^{125}I . An UF-RO pilot study demonstrated that radioactive liquid waste could be made 15 times more concentrated and the final permeate successfully discharged after further treatment [12].

However, there has yet to be any research on the radioactive liquid waste directly produced from radiation therapy rooms in hospitals. Radioactive liquid waste from radiation therapy rooms differs from in-vitro RIA waste in that it includes all of the liquid waste generated from the examination, radiation, and the patients themselves, while RIA waste contains only the analysis reagents and samples, and typically its radioactivity is much lower than those from radiation therapy room. Medical radioactive liquid waste is a mixture of urine, feces, detergent, and radionuclides [13], which can cause severe membrane fouling in RO [14]. In addition, due to high working pressure (e.g., 20–60 bar), RO membrane fouling tends not to be readily removed by physical or chemical cleaning [15]. Furthermore, the high pressure in RO can also risk the leakage of radionuclides by damaging RO membrane housing, O-rings and fittings and the destruction of the polyamide layer due to increased differential pressure caused by membrane fouling [16]. Therefore, any treatment process for radioactive liquid waste needs to be stable and safe.

Recently, forward osmosis (FO) has been suggested as an alternative to the conventional RO-based desalting process [17-19]. FO utilizes a highly concentrated draw solution (DS) as a driving force to extract pure water from the feed solution (FS) based on the difference in osmotic pressure between the DS and FS. As a result, FO is characterized by low energy consumption, high fouling reversibility, and high recovery rates [20, 21]. In particular, the high fouling reversibility, which arises because of the low working pressure, means FO is suitable for the treatment of wastewater with high fouling potential [22, 23]. FO can also be used to remove toxic ions such as heavy metals and arsenic due to its high rejection rate [24,

25]. In FO, the significant difference in salt concentration between the DS and FS leads to the reverse diffusion of draw solutes from the DS to the FS, which may block the forward transport of feed solutes into the DS [26]. Consequently, FO can be considered safer and more stable for the treatment of radioactive liquid waste compared to a pressure-driven membrane process.

In the present study, we have attempted, for the first time, to apply FO to the treatment of medical radioactive liquid waste produced by radiation therapy. The conventional radionuclide decay system consists of two main septic tanks for the collection of liquid waste and the decay of radionuclides, respectively (**Fig. 1a**). Liquid waste is collected and stored in the first septic tank for the decay of the radionuclides. The liquid waste is then pumped to one of the second septic tank for further decay and discharged after the final water quality (i.e., radioactivity) has met regulations. This system is simple but requires a large area for the tanks, thus improving the management of the radiation therapy facilities. Therefore, to enhance operating efficiency, FO can be integrated with the decay tanks, as depicted in **Fig. 1b**. In this system, FO plays an important role in concentrating the radioactive liquid waste and thus reducing the required size of the septic tanks, allowing an increase in the number of radiation therapy rooms and thus treating more patients if so desired.

Figure 1. Conceptual diagram of (a) the conventional radionuclide decay system and (b) the novel forward osmosis treatment system for the radioactive liquid waste generated by radiation therapy rooms in hospitals. Treated liquid wastes should be lower than 30 Bq/L for discharge to sewage.

The objective of this study is to evaluate the feasibility of FO for the treatment of the medical radioactive liquid waste produced by radiation therapy in a hospital. First, the transport mechanism of radioactive iodine in FO was analyzed by comparing it with that of natural iodine to determine the optimal FO operating conditions (i.e., pH and the characteristics of the draw solution). After this, actual radioactive liquid waste was collected and concentrated using FO under these operating conditions. Finally, the management of radioactive liquid

waste (i.e., the volume of storage tanks) was simulated based on theoretical models derived from mass balance. This study has potential implications for the concentration of radioactive liquid waste produced from other sources (e.g., nuclear power plants, legacy wastes, industries, and naturally occurring radioactive materials).

2. Materials and methods

2.1 FO membrane and draw solutions

The thin-film composite (TFC) polyamide (PA) FO membrane used in this study was provided by Porifera, Inc. (Hayward, CA, USA). This membrane was specifically fabricated with a flat-sheet form to be thinner for use with non-pressurized osmosis. The detailed specifications of this membrane can be found in other studies [27, 28]. The membranes were stored in their dried form in a desiccator in accordance with the manufacturer's recommendations. The membrane coupon was immersed in deionized (DI) water for 30 min prior to all FO experiments.

Sodium chloride (NaCl) and magnesium chloride (MgCl_2) were received at reagent grade (Junsei Chemical, Japan) and used as draw solutes. The DS was prepared by dissolving these chemicals in DI water. Thermodynamic properties of the two chemicals were obtained using the OLI Stream Analyzer 3.2 (OLI System Inc., USA). Despite lower concentration, 0.48 M MgCl_2 exhibited higher osmotic pressure than 0.6 M NaCl. On the other hand, NaCl showed higher diffusivity than MgCl_2 . Detailed information about the DS chemicals is provided in **Table S1**.

2.2 Radioactive feed solutions

Natural iodine (^{127}I) was received in powder form (NaI ; Sigma Aldrich, Korea) and radioactive iodine (^{125}I) was received in the form of a highly concentrated solution (99.5% Na^{125}I ; PerkinElmer, USA). Their physicochemical properties are summarized in **Table 1**. The ^{125}I was selected as the representative form of radioactive iodine because its half-life is much longer than that of ^{131}I (**Table 1**). Both forms of radioactive iodine are utilized in hospitals but radiation therapy rooms generally produce radioactive liquid waste containing ^{131}I .

Table 1 Physicochemical properties of natural iodine (^{127}I), ^{125}I , and ^{131}I . Source: <https://pubchem.ncbi.nlm.nih.gov>.

	^{125}I	^{127}I	^{131}I
Molecular formula	HI	I_2	I_2
Molecular weight	125.91 g/mol	253.81 g/mol	261.81 g/mol
Half-life	59.40 d	Stable	8.02 d
Decay mode	Gamma emission	Stable	Beta emission

The radioactive liquid waste used in this study was sampled from the 1st septic storage tank at Soonchunhyang University Hospital in Korea. The liquid waste was mainly composed of urine, feces, and detergent, with a significant amount of ^{131}I present due to its use in thyroid cancer treatment. The concentration of ^{131}I in the sample of the radioactive liquid waste was $3,363,500 \pm 1,149,402$ cpm/L. The physicochemical properties and water quality of the radioactive liquid waste used as a FS are presented in **Table 1** and **Table 2**, respectively.

Table 2 Water quality of the radioactive liquid waste used in this study. The waste was collected from the 1sts septic storage tank at Soonchunhyang University Hospital (Bucheon, Korea). The analysis was conducted repeatedly.

Components	Values	Components	Values
Chemical oxygen demand (mg/L)	20.85 (± 22.98)	Na ⁺ (mg/L)	5.71 (± 1.49)
Total dissolved solids (mg/L)	1.68 (± 0.28)	Mg ²⁺ (mg/L)	0.63 (± 0.56)
Suspended solids (mg/L)	25 (± 26.87)	Ca ²⁺ (mg/L)	2.84 (± 2.34)
TP (mg/L PO ₄ -P)	2.18 (± 2.98)	Cl ⁻ (mg/L)	66.3 (± 13.58)
TN (mg/L N)	23.27 (± 12.25)	SO ₄ ²⁻ (mg/L)	9.2 (± 0.42)
NH ₃ -N (mg/L)	13.04 (± 3.08)	HCO ₃ ⁻ (mg/L)	4.2 (± 0.57)
pH	7.66 (± 0.03)		

2.3 FO experiments

All FO experiments were carried out using a lab-scale FO system similar to the one described in our previous work [29]. The FO cell had two symmetric channels that were 77 mm long, 26 mm wide and 3 mm deep on both sides of the membrane. Variable speed gear pumps (ISMATEC, Germany) were used to provide crossflow in a co-current direction at a crossflow rate of 8.5 cm/s, with a solution temperature of 25 ± 1 °C and an AL-FS (active layer facing FS) membrane orientation. The concentrations of NaCl and MgCl₂ were 0.6 M and 0.48 M, respectively, in order to ensure their initial water flux was approximately equal. In addition, 0.6 M NaCl was determined to simulate typical seawater as the DS since this study can be expanded to treating radioactive liquid waste generated from a nuclear power

plant located near the sea. Both solutions having 1 L initial volumes were recirculated in a closed-loop system, resulting in batch mode operation. The DS tank was placed on a digital weighing scale (AND KOREA, Korea) and weight changes were recorded with a computer in real time at 3-minute intervals to determine the water flux.

To investigate the transport behavior of natural iodine in FO, a NaI solution with a concentration of 150 mg/L was prepared for the FS. The FO experiments were conducted for 4 h under different pHs (pH 4, 7, and 10) and different DSs (0.6 M NaCl and 0.48 M MgCl₂) to evaluate their effects on transport behavior. The solution pH was adjusted with 0.1 M of NaOH or HCl stock solution. The transport behavior of radioactive iodine was then evaluated using the radioisotope ¹²⁵I. Prior to the experiments, 1 mL of the stock solution was spiked into 1 L of FS to obtain a final concentration of 0.5 mCi/L. The FO experiments were conducted by following the same protocol as that used for natural iodine.

Finally, actual radioactive liquid waste generated by patients exposed to ¹³¹I was treated under optimal conditions, as determined by the natural iodine and ¹²⁵I experiments. Before starting, the radioactive liquid waste was pre-screened using 250 µm mesh (US size 60 mesh) to reduce fouling potential by particulates. The FO experiments were conducted for 24 h with 0.6 M NaCl DS without adjusting the feed pH (i.e., pH 7.66) because the pH of the radioactive liquid waste was quite close to pH 7 which exhibited high rejection rate in the present study. To evaluate the effect of hydraulic washing on the recovery of water flux by the FO membrane after fouling, DI water was flushed inside the DS and FS channels at a crossflow velocity that was three times higher than normal (25.5 cm/s) for 15 min. After this, the stability of FO was evaluated by conducting FO experiments with a fresh FS and DS for 24 h. To determine the dilution effect of the DS on the water flux, a baseline test was carried out for 24 h with DI water as the FS and 0.6 M NaCl as the DS; the resulting flux curves were

utilized to establish a baseline for comparison with the water flux obtained during the treatment of the liquid waste. All FO experiments were carried out at least two times and average values were presented.

2.4 Membrane surface characterization

The zeta potential for the active layer of virgin FO membranes was analyzed using a streaming current electrokinetic analyzer (SurPass, Anton Paar GmbH, Austria). Repeated measurements were conducted with the pH varying from 3 to 10 with 0.01 M KCl as the background electrolyte solution. The average zeta potential for the FO membranes is reported in **Fig. 2**.

Membrane surface characterization was conducted by collecting the membrane coupons after FO treatment of radioactive liquid waste, soaking them in DI water for a few seconds to remove the FS and DS, and then drying them in a desiccator for 1 day. The surface and cross-sectional morphologies of the fouled FO membrane were observed and analyzed using scanning electron microscopy (SEM, Quanta 250 FEG, FEI, USA) and energy dispersive X-ray spectroscopy (EDX). Samples taken from each membrane were first lightly coated with Pt and then SEM imaging was carried out at an accelerating voltage of 15 kV.

Figure 2. Zeta potential of FO membranes as a function of pH (pH 3 to pH 10.5). The isoelectric point of the FO membranes used in this study was around pH 4.

2.5 Analytical methods for natural and radioactive iodine

The natural iodine in the samples were analyzed following the procedures described in other studies [30]. Concentration was measured using inductively coupled plasma mass spectrometry (ELAN DRC II, PerkinElmer, USA). Natural iodine rejection in FO can be calculated using permeate concentration, yielding [25]:

$$R_{iodine} = \left(1 - \frac{C_{iodine} \times V_D / V_P}{C_{iodine,f}} \right) \times 100\% \quad (1)$$

where R_{iodine} is the rejection rate (%) of natural iodine, C_{iodine} and $C_{iodine,f}$ are the concentration of natural iodine (mg/L) in the DS and FS, respectively. To take the dilution effect of DS into account for the rejection rate calculation, a ratio of the permeate volume to the total DS volume was employed in **Eq. (1)**. Concentrations of both forms of radioactive

iodine (i.e., ^{125}I and ^{131}I) were analyzed in terms of radioactivity (counts per minutes, cpm) with a gamma well counter (Captus 3000, CAPINTEC. USA). For ^{125}I , the rejection rate was obtained from **Eq. (1)** because its half-life is relatively long (59.4 d) compared to the duration (4 h) of the experiment. However, the half-life of ^{131}I is very short (8.02 d), thus the rejection rate needs to be calculated using **Eq. (2)**,

$$R_{I-127} = \left(1 - \frac{C_{I-127} \exp(\lambda t_2) \times V_D / V_P}{C_{I-127, f} \exp(\lambda t_1)} \right) \times 100\% \quad (2)$$

where t_1 and t_2 are time that has passed (d) since the start of the experiment, and λ is decay constant (d) of the radionuclides (0.0864 d^{-1} for ^{131}I).

2.6 Simulation of radioactive liquid waste management

The proposed radioactive liquid waste management system consisted of a FO system for the concentration of the liquid waste and two main septic tanks (**Fig. 1**). The liquid waste was firstly concentrated by the FO system. The concentrated liquid waste was then filled in the first septic tank before being moved to the second tank for further decay to meet discharge regulations. The mass balance of the volume for the radioactive liquid waste in FO and the recovery rate of FO can be written as **Eq. (3)** and **Eq. (4)**, respectively. The inflow of the radioactive liquid waste from radiation therapy rooms can be obtained using **Eq. (5)**. The final volume of the radioactive liquid waste after FO concentration can be given in the form of **Eq. (6)** by substituting **Eq. (4)** and **Eq. (5)** into **Eq. (3)**.

$$Q_{out,FO} = Q_{in,FO} - Q_p \quad (3)$$

$$Re_{FO} = \frac{Q_p}{Q_{in,FO}} \quad (4)$$

$$Q_{in,FO} = N_{room} Q_{room} \quad (5)$$

$$Q_{out,FO} = N_{room} Q_{room} (1 - Re_{FO}) \quad (6)$$

where, N_{room} is the number of radiation therapy rooms, $Q_{in,FO}$ and $Q_{out,FO}$ are the respective flow rate of the inflow and the outflow in FO (m^3/d), Q_p is the permeate flow rate (m^3/d), Q_{room} is the production rate of the radioactive liquid waste (m^3/d), and Re_{FO} is the recovery rate (defined as a ratio of the permeate to the FS) for FO. The mass balance for the concentration of the radioactive liquid waste in FO and the iodine rejection rate of FO can be written as **Eq. (7)** and **Eq. (8)**, respectively. The final concentration of the radioactive liquid waste after FO concentration can be given in the form of **Eq. (9)** by substituting **Eq. (3)**, **Eq. (4)** and **Eq. (8)** into **Eq. (7)**.

$$Q_{out,FO} C_{out,FO} = Q_{in,FO} C_{in,FO} - Q_p C_p \quad (7)$$

$$R_{iodine} = 1 - \frac{C_p}{C_{in,FO}} \quad (8)$$

$$C_{out,FO} = \frac{C_{in,FO} - C_{in,FO} Re_{FO} (1 - R_{iodine})}{1 - Re_{FO}} \quad (9)$$

where, $C_{in,FO}$ and $C_{out,FO}$ are respective initial and final concentrations of the radioactive liquid waste (cpm/L), C_p is the iodine concentration in the permeate (cpm/L), and R_{iodine} is the iodine rejection rate for FO. The first septic tank is theoretically same as the sequencing batch reactors (SBR) due to the accumulation of water (no outflow) in the reactor [31]. Thus, the mass balance for radionuclides in the first septic tank can be calculated from **Eq. (10)** and modified as **Eq. (11)**:

$$\frac{d(V_{S1}(t)C_{S1}(t))}{dt} = Q_{S1,in}C_{S1,in} - Q_{S1,out}C_{S1,out} - V_{S1}(t)r \quad (10)$$

$$\frac{dC_{S1}(t)}{dt} + \frac{C_{S1}(t)}{V_{S1}(t)} \frac{dV_{S1}(t)}{dt} = \frac{Q_{S1,in}}{V_{S1}(t)} C_{S1,in} - r \quad (11)$$

where $V_{S1}(t)$ is the volume of the first septic tank (m^3), $C_{S1}(t)$ is the concentration of radionuclides in the first septic tank (cpm/L), $Q_{S1,in}$ and $Q_{S1,out}$ are the respective inflow and outflow of radioactive liquid waste (m^3/d), $C_{S1,in}$ and $C_{S1,out}$ are the concentration of radionuclides in the inflow and the outflow, respectively (cpm/L), r is the decay rate of radionuclides (cpm/L/d), and t is the operation time (d). Due to the unique properties of SBR, the change in volume as a function of time should be identical to the inflow rate, which is $dV_{S1}(t)/dt = Q_{S1,in}$ or $V_{S1}(t) = Q_{S1,in}t$. Therefore, **Eq. (11)** can be modified to become **Eq. (12)**. Because the decay rate can be written in the form of **Eq. (13)**, **Eq. (12)** can be modified to become **Eq. (14)**:

$$\frac{dC_{S1}(t)}{dt} = \frac{C_{S1,in} - C_{S1}(t)}{t} - r \quad (12)$$

$$r = \lambda C_{S1}(t) \quad (13)$$

$$\frac{dC_{S1}(t)}{dt} = \frac{C_{S1,in} - C_{S1}(t)}{t} - \lambda C_{S1}(t) \quad (14)$$

For the second septic tank, the mass balance of the radionuclides can be written as **Eq. (15)** and summarized as **Eq. (16)** due to the lack of inflow, the lack of outflow, and the constant tank volume (which is the same as the volume of the first septic tank):

$$\frac{d(V_{S2}(t)C_{S2}(t))}{dt} = Q_{S2,in}C_{S2,in} - Q_{S2,out}C_{S2,out} - V_{S2}(t)r \quad (15)$$

$$\frac{dC_{S2}(t)}{dt} = -r \quad (16)$$

where $V_{S2}(t)$ is the volume of the second septic tank (m^3), $C_{S2}(t)$ is the concentration of radionuclides in the second septic tank (cpm/L), $Q_{S2,in}$ and $Q_{S2,out}$ are the respective inflow and outflow of radioactive liquid waste (m^3/d), and $C_{S2,in}$ and $C_{S2,out}$ are the concentration of radionuclides in the inflow and the outflow, respectively (cpm/L). A set of ordinary differential equations (ODEs) was numerically solved to simulate the proposed management system for radioactive liquid waste.

3. Results and discussion

3.1 Basic FO performance: Water flux and rejection of natural iodine

To determine the optimal operating conditions for the concentration of radioactive liquid waste, the transport behavior of radioactive iodine needed to be evaluated for different solution pHs and DS types. Prior to this, however, the behavior of natural iodine was examined to discover its transport mechanism through an FO membrane.

3.1.1 Influence of pH on FO performance

FO experiments were carried out with FSs of varying pH (from 4 to 10); the results are presented in **Fig. 3a**. Water flux increased only slightly (from 20.4 ± 1.2 L/ m^2 /h to 20.8 ± 2.1 L/ m^2 /h) with an increase in solution pH (from pH 4 to pH 10, respectively), suggesting pH had no significant influence, associated with external concentration polarization (ECP).

The rejection rate for natural iodine is shown in **Fig. 3a**. pH 7 and pH 10 exhibited similar rejection rates (97.47% and 96.06%, respectively), while pH 4 had a much lower rejection rate (26.01%). This can be explained by the zeta potential of the FO membrane in relation to

variations in solution pH. As presented in **Fig. 2**, the FO membrane surface is negatively charged at both pH 7 and pH 10 (-20 mV and -28 mV, respectively), while it is neutrally charged at pH 4. Consequently, when operating FO under a pH of 7 or 10, the natural iodine ions are rejected by the negatively charged membrane surface due to the strong electric repulsion between the negative ions and the membrane surface [26, 32, 33]. On the other hand, natural iodine ions could be more readily transported to the DS at a pH of 4 due to the neutrally charged membrane surface. Therefore, it can be concluded that the dominant mechanism for rejecting ions is the electric repulsion rather than the steric hindrance by very tight TFC FO membrane.

Figure 3. Water flux (column, left axis) and rejection rate (red points, right axis) of natural iodine for (a) varying pHs of the FS with a DS of NaCl 0.6 M and (b) different DSs (0.6 M NaCl and 0.48 M MgCl₂) with a pH of 7. Time-dependent water flux curves are presented in **Fig. S1a and S1b**. The following experimental conditions were used for all FO experiments: 150 mg/L NaI solution as the FS; crossflow velocity 8.5 cm/s; 4 h operation; and temperature 25 ± 1 °C.

3.1.2 Influence of the draw solution on FO performance

FO experiments were conducted with different DSs to investigate the effect of the DS on FO performance. Because water flux has a significant impact on the rejection rate [34], the concentration of each DS was adjusted to ensure that the initial water flux was similar. Interestingly, even though 0.48 M MgCl_2 had a higher osmotic pressure than 0.6 M NaCl, their water fluxes were very similar ($19.6 \pm 2.3 \text{ L/m}^2/\text{h}$ and $20.4 \pm 1.1 \text{ L/m}^2/\text{h}$, respectively). This is because MgCl_2 has a lower diffusivity than NaCl ($0.79 \times 10^{-9} \text{ m}^2/\text{s}$ and $1.45 \times 10^{-9} \text{ m}^2/\text{s}$, respectively) and a higher viscosity (1.06 mPa·s and 0.94 mPa·s, respectively; **Table S1**), thus increasing internal concentration polarization [35]. As a result, the effective osmotic pressure gradient for 0.48 M MgCl_2 DS was similar to that of 0.6 M NaCl DS.

Reverse salt flux (RSF) was also evaluated because it can influence the transport of natural iodine [26, 32, 36]. The results indicated that 0.48 M MgCl_2 DS exhibited a lower RSF ($33.7 \text{ mmol/m}^2/\text{h}$ and $202.7 \text{ mmol/m}^2/\text{h}$, respectively) than 0.6 M NaCl DS (**Table S2**). There are three potential reasons for the lower RSF with MgCl_2 DS: (i) lower bulk concentration, (ii) lower solute diffusivity, and (iii) larger hydrated diameter. Because the concentration of MgCl_2 DS was much lower than that of NaCl DS, RSF, which is a function of the effective concentration gradient across the active layer of the FO membrane, should be lower. The lower solute diffusivity of MgCl_2 could also lead to stronger ICP and thus decrease the effective concentration gradient. Finally, the larger hydrated diameter of Mg^{2+} ions could hinder the back diffusion of draw solutes. Besides, due to the Donnan equilibrium, the reverse diffusion of Cl^- ions in MgCl_2 DS could be also restricted [37].

The rejection rate of natural iodine was further investigated with different DSs (**Fig. 3b**). The rejection rate of natural iodine with MgCl_2 DS was lower than that with NaCl DS (84.99% and 97.47%, respectively). This may be due to the difference in the RSF between the two

DSs, as discussed above. The RSF likely hinders the forward transport of feed solutes, strengthening the rejection properties [36, 38]. Therefore, because the RSF with MgCl_2 DS was lower than that with NaCl, the rejection rate for natural iodine was lower with MgCl_2 DS than with NaCl DS, which is consistent with previous studies [14, 32]. This suggests that a high RSF is preferable for applications which require a very high rejection of feed solutes even though it can have a considerably negative impact on the sustainability of FO operation.

3.2 FO performance with radioisotopes

To investigate the transport behavior of radioactive iodine across the FO membrane, FO experiments were carried out with the radioisotope ^{125}I , which was adopted as a representative radioisotope not only because it is used in radiation treatment, but also its relatively longer half-life (59.4 d) makes the analysis more convenient. Interestingly, **Fig. 4a** shows that the water flux significantly varied (from $18.0 \pm 3.4 \text{ L/m}^2/\text{h}$ to $22.3 \pm 0.2 \text{ L/m}^2/\text{h}$) with a change in solution pH.

This behavior might be attributed to conformational changes of the cross-linked membrane polymer structure, which can be elucidated by increased zeta potential of the active layer with increasing FS pH. Xie et al. suggested that the enhanced electrostatic repulsion between ionizable functional groups of the membrane polymeric matrix can lead to an increased average pore size and higher permeate flux [33]. The results are well agreed with results in other studies [39, 40]. Moreover, the water flux in the presence of radioisotopes was similar to that for natural iodine, indicating that radioisotopes do not significantly influence the intrinsic properties of the FO membrane at low levels of radioactivity.

The rejection rate for the radioisotopes was obtained by analyzing the radioactivity of FS and DS. It was found that the rejection rate at pH 4 (i.e., 24.12%) was much lower than at pH 7 and pH 10 (i.e., 99.11% and 99.3, respectively), which is consistent with the results presented

in **Section 3.1.1**. As discussed above, the zeta potential of the membrane surface changes with solution pH, and thus the rejection rate at high pH increased due to the strong electric repulsion between the membrane surface and the iodine ions (**Fig. 5a**). The water flux and rejection rate results indicate that radioactive iodine does not damage the FO membrane at low levels of radioactivity. Therefore, it can be suggested that FO is a promising technology for the treatment of low-level radioactive liquid waste.

As discussed in **Section 3.1.2**, RSF can significantly influence the transport of radioactive iodine. Therefore, RSF was measured for different pHs and the results are presented in **Table S2**. It is interesting to note that RSF decreased as pH increased (231 mmol/m²/h, 202.7 mmol/m²/h, and 117.7 mmol/m²/h for pHs 4, 7 and 10, respectively). This might be due to the significant increase in the zeta potential with an increase in feed pH. A more negatively charged zeta potential suppresses the back diffusion of draw solutes by inducing larger electrostatic repulsion [33]. Therefore, the decrease in RSF at pH 10 may have led to a slight reduction in the rejection rate compared to that at pH 7 for both natural and radioactive iodine. However, further research is required to fully understand this phenomenon.

Figure 4. Water flux (columns, left axis) and rejection rate (red symbols, right axis) for radionuclides (¹²⁵I) in relation to (a) pH of the FS with NaCl 0.6 M DS and (b) DS (0.6 M

NaCl and 0.48 M MgCl_2) at pH 7. Time-dependent water flux curves are presented in **Fig. S1c and S1d**. The following experimental conditions were used for all FO experiments: 0.5 mCi/L Na^{125}I solution as the FS; crossflow velocity 8.5 cm/s; 4 h operation; and temperature 25 ± 1 °C.

The effect of DS type on FO performance when treating a solution containing radioisotopes (^{125}I) was investigated and compared with the results from **Section 3.1.2**. Water flux with 0.6 M NaCl DS was slightly higher than that with 0.48 M MgCl_2 DS (21.0 ± 1.7 L/m²/h and 18.7 ± 0.8 L/m²/h, respectively). The rejection rate for radioisotopes was obtained for the different DSs; the results are presented in **Fig. 4b**. MgCl_2 DS exhibited a lower rejection rate than did the NaCl (89.02% and 99.12%, respectively), a similar trend to that in **Fig. 3b**. The higher rejection rate for NaCl DS could be caused by the RSF hindering the transport of feed solutes, as depicted **Fig. 5b**. In these experiments, the water flux with MgCl_2 DS was slightly lower than that with NaCl DS, leading to a decrease in the rejection rate.

Interestingly, the results for the radioisotopes (^{125}I) exhibited a very similar trend to those for natural iodine. This is because there was no significant difference in the physicochemical properties of both forms of iodine. Furthermore, the radiation emitted by the radioisotopes had no apparent impact on the intrinsic properties of the FO membrane primarily due to low radioactivity.

Overall, it can be concluded that FO should operate at pHs higher than 7 and using a DS with an adequate RSF in order for solutions containing radionuclides to be effectively treated. Moreover, when comparing our results with the rejection rates from previous studies [11, 12], it can be seen that stand-alone FO exhibited similar rejection rates (i.e., 99.12% and 99.8%, respectively) to a UF-RO hybrid system. In a lab-scale UF and RO hybrid process, UF, 1st pass RO, and 2nd pass RO could achieve rejection rates of 50~60%, 90% and 95%,

respectively, leading to a total rejection rate of approximately 99.8% [11]. This is similar to the results from a pilot UF and RO hybrid system [12]. Furthermore, since FO can be considered as a non-pressured desalting process (FO working pressure for various water resources: 1 ~ 2 bar), its energy requirement is low compared to other desalting technologies (RO working pressure for brackish water: 15 ~ 30 bar) according to other studies [22], although significant energy is required for draw re-generation. Even because of high fouling potential, RO cannot be utilized to treat radioactive liquid waste produced from the radiation therapy facilities. This suggests that FO is more stable and economically feasible for the treatment of radioactive liquid waste. Even though the FO experiments were conducted with natural iodine (^{127}I) and radioisotope ^{125}I , findings from this study can be applied to the treatment of radioactive liquid waste containing radioisotope ^{131}I because there is no significant difference in their physicochemical properties (**Table 2**).

Figure 5. The effects of (a) FS pH and (b) DS type on radioactive iodine transport behavior across FO membranes. (a) The negatively charged surface can induce the electrostatic repulsion of iodine anions, resulting in a higher rejection rate. (b) The RSF can hinder the forward transport of iodine ions to the DS side, and thus lead to a higher rejection rate.

3.3 Treatment of real radioactive liquid waste from radioactive therapy

3.3.1 Flux decline

For the first time to our knowledge, FO experiments were conducted using actual radioactive liquid waste as a FS and NaCl 0.6 M as a DS. Since FO above pH 7 exhibited better rejection property, FS pH (7.66) was not adjusted. The resulting flux is presented in the form of normalized water flux in **Fig. 6**. Results show that the normalized water flux rapidly declined to 30% of the initial water flux (from 19.4 ± 1.3 L/m²/h to 5.9 ± 0.3 L/m²/h) after 1 day of operation. Because FO was operated in batch mode, in which the FS and DS were recirculated in a closed-loop system, the FS became concentrated and the DS was continuously diluted, reducing the effective osmotic pressure gradient. Therefore, to exclude the influence of this phenomenon on the flux, baseline experiments were carried out using DI water as the FS and 0.6 M NaCl. It was found that membrane fouling occurred after 200 mL of permeate had been produced, corresponding to 6 h of operation. Using SEM imaging, it was observed that the membrane surface was completely covered by organic foulant deposits with a thickness of 6.25 ± 1.09 μ m (**Fig. 7a**).

To remove the fouling layer, hydraulic washing was conducted by increasing the cross-flow velocity of both solutions to 25.5 cm/s for 15 min, and then the FS and DS were replaced with fresh solutions. **Fig. 6** shows that the water flux recovery was only 75% after hydraulic washing. This low recovery rate may originate from the specific mix of foulants. A closer observation of the fouled surface (**Fig. 7b and 7c**) revealed that the fouling layer was composed of a variety of deposits because actual radioactive liquid waste is a mixture of organic and inorganic substances (**Table 3**). Small spherical foulants and organic-like foulants were observed on the membrane surface (**Fig. 7b and 7c**). In FO, when both organic compounds and colloids are deposited onto the membrane surface, this combined fouling can

reduce the efficiency of hydraulic washing [41]. To more accurately identify the composition of the fouling layer, EDX analysis was conducted on the fouled surfaces in **Fig. 7b and 7c**. However, because the fouling layer was a mix of organics and scalants, there was no distinguishable difference in the EDX results (**Fig. S2**). Nevertheless, EDX results suggest that not only organics existed dominantly in the fouling layer but also a considerable amount of inorganics such as silica and some scales was observed. From this result, it can be also drawn that the fouling reversibility of FO might be limited due to severe membrane fouling. However, compared to pressurized desalting process such as RO, FO has better fouling reversibility even though its efficiency is not perfect since high working pressure in other processes makes the fouling layer hardly removed [14, 41, 42].

The FO recovery rate is one of most important factors to consider when assessing the feasibility of concentrating radioactive liquid waste. In this study, radioactive liquid waste could be successfully reduced down to approximately 40% of its original volume (corresponding to a concentration factor of 2.5) after the 1st cycle (1 day operation) and then approximately 49% (corresponding to a concentration factor of 2.04) after the 2nd cycle (another 24 h) due to the lack of effectiveness of hydraulic washing (**Fig. 6**). It is noteworthy that the limited efficiency of hydraulic washing can lead to the reduction in a lifespan of FO membrane. This suggests that alternative cleaning strategies need to be implemented to ensure efficient operation and a high recovery rate. For example, osmotic backwashing can be considered due to its effectiveness in flushing the fouling layer during FO under AL-FS mode [26]. Citric acid can also be utilized as an efficient chemical agent for the dissociation and removal of the combined fouling layer [22]. Despite the need of further developing cleaning strategies as well as draw regeneration, it can be cautiously said that FO could be economically viable and environmentally feasible for concentrating radioactive wastewater

due to its lower energy requirement [22, 43] and higher fouling reversibility [14, 42] than pressurized desalting technologies.

In addition to cleaning strategies, post-treatment of the produced water from the cleaning should be also taken into account since the fouling layer can contain radioactive iodine. The simplest way is just to return the produced water to the first septic tank. However, since this can influence the operation of the septic decay tanks, the further study is required to develop/optimize the appropriate cleaning method by considering cleaning efficiency as well as radioactivity reduction for the feasible and safe operation of FO to treat the radioactive liquid waste.

Figure 6. Flux curves obtained during FO operation in the treatment of actual radioactive liquid waste produced by radiation therapy rooms. Hydraulic washing was conducted after 1 day of operation by flushing DI water inside the DS and FS channels at a 3-times higher crossflow velocity (25.5 cm/s) for 15 min. Initial fluxes of baseline and fouling experiments were 19.9 L/m²/h and 19.4 L/m²/h, respectively. Experimental conditions for the FO

experiments: radioactive liquid waste containing ^{131}I as the FS; NaCl 0.6 M as the DS; crossflow velocity of 8.5 cm/s; and temperature of 25 ± 1 °C.

Figure 7. SEM images of (a) the fouled membrane surface (i.e., the combined image taken from two different spots) under 10k magnification, (b) organic-like foulants under 70k magnification, and (c) scale-like foulants under 70k magnification.

3.3.2 Rejection rates

To further investigate the feasibility of FO in the treatment of radioactive liquid waste, rejection rates were measured by analyzing the radioactivity of the FS and DS. **Table 3** shows that FO rejected 98.01% of radionuclides (^{131}I), which was a little lower than the results from **Section 3.1 and 3.2**. This might be due to lower water flux which could increase iodine concentration in the permeate, thereby resulting in a reduction in the rejection rate [44].

The rejection rate slightly reduced even after hydraulic washing due to membrane fouling but was still high enough to sustain FO operation. The high rejection of the FO membrane stems from both the membrane itself (i.e., a small mean effective pore size) and the unique characteristics of FO operation (i.e., the hindrance effect by the high reverse diffusion of draw solutes), as discussed previously.

Table 3 presents the radioactivity of the final DS. These results indicate that, despite the significant reduction of radioactivity, substantial dilution or further decay is still required to enable the direct discharge of the DS into the sewage system because the permissible limit for discharge is 1.17 cpm/mL (equivalent to 1.8 dpm/mL or 0.03 Bq/mL) in the Republic of Korea. However, in the re-generation of the DS, additional desalting process (e.g., nanofiltration, reverse osmosis, or membrane distillation) is essential, and thus the final product water can be reused within the hospital. Nevertheless, a further study is required to lower radioactivity of the final product water as well as recover DS efficiently. In addition, since hospital liquid waste is a mixture of various contaminants, a small portion of some contaminants can be possibly diffused into the DS. Results of the water quality analysis showed that FO could reject 96.6% organics while a considerable amount of TN was diffused into the DS. This was because FO hardly removes ammonia which is a main component of urine [45]. However, Liu et al. suggested that the complete rejection of TN can be achieved by combining with MD as the re-regeneration process [46].

Due to high rejection property of the re-generation process, both radioactive iodine and some contaminants can be accumulated in the DS. This can be simply prevented by employing the adsorption technology which can effectively remove both radioactive nuclides and other contaminants [47, 48]. The requirement of the DS re-generation can also make FO as an energy intensive process. However, FO can more readily control membrane fouling than

other membrane-based desalting processes such as RO [14, 33, 41], which possibly lower operation expenses, for instance, from the cleaning chemicals and electricity for increasing working pressure by membrane fouling. This can make FO more advantageous to treat the liquid wastes having high fouling potential. Furthermore, although FO requires the energy for re-generate the conventional inorganic draw solution, FO can be beneficial in terms of the hospital management when considering the cost required for the further installation of radioactive liquid waste handling facility as well as radiation therapy rooms.

Table 3. Rejection rates (%) of radionuclides (^{131}I) in radioactive liquid waste generated by radioactive therapy rooms. The radioactivity of the feed and draw solutions was measured using a gamma well counter, and the half-life was used to correct the radioactivity when calculating the rejection rate.

	1 st cycle	2 nd cycle
Rejection rate (%)	98.01 $\pm$ 0.2	97.36 $\pm$ 0.89
Radioactivity in DS (cpm/mL)	26.62 $\pm$ 11.41	27.61 $\pm$ 4.73

3.4 Simulation of radioactive liquid waste management

To investigate the effect of FO on the management system for radioactive liquid waste, septic tank capacity and retention time were determined using the mass balance equations with decay rate of radioactive iodine isotopes for the simulation conditions shown in **Table S3**. For the simulation, the system was designed to have one first septic tank and two second septic tanks, which is similar to the current design used in the Suncheonhyang University Hospital. The first septic tank played a role to collect radioactive liquid waste and deliver it to

the second tank, and then the second septic tank further reduced radioactivity up to the 30 Bq/L for the discharge. Tank capacity was first simulated as a function of the recovery rate in FO (**Fig. 8**). Results indicate that septic tank capacity can be significantly reduced from 48 m³ to 12 m³ when concentrating the radioactive liquid waste by up to 75% with FO. Furthermore, it is noteworthy that retention time increased only 10 % (from 155 d to 171 d) even though the radioactivity in the liquid waste after FO concentration increased significantly from 18,933 cpm/mL to 75,203 cpm/mL. This is because the total amount of radionuclides is relatively constant due to the high rejection properties of the FO membrane. Based on the simulation, it can be concluded that FO can successfully reduce the space requirements for septic tanks with little impact on retention time, meaning that more radiation therapy rooms could be installed in a hospital (e.g., from 2 rooms to 8 rooms at 75% recovery rate).

Figure 8. Simulated tank capacity (columns, left axis) and retention time (red symbols, right axis) as a function of recovery rate (%) in FO. The simulation models were developed by adopting the decay rate of radioactive iodine isotope (^{131}I).

In order to further elucidate the effect of FO concentration on the discharge of radioactive liquid waste in detail, the retention time was simulated as a function of the number of radiation therapy rooms and recovery rate (%) in FO with a fixed septic tank capacity. Results of **Fig. 9** show that an increase in recovery rate in FO could increase the retention time in septic tanks even though the tank capacity was constant, consistent with results shown in **Fig. 8**. However, interestingly, an increase in the number of radiation therapy rooms could lead to a reduction in retention time. This was due to a naturally increased production of radioactive liquid waste when increasing the number of radiation therapy rooms. In the first septic tank, since the decay and the accumulation of radioactive nuclides occurred simultaneously, an increased amount of liquid waste reduced retention time and thus led to less decay of radioactive nuclides. Therefore, for the further decay of radioactive nuclides to meet the discharge standard, retention time in the second septic tank was increased. As a result, total retention time for the discharge could be decreased when increasing the number of radiation therapy rooms. To understand a decrease in total retention time, the transport of the last radioactive nuclide fed into the first septic tank should be considered. In terms of the last radioactive nuclide, its decay occurred only in the second tank since liquid waste in the first tank was moved to the second septic tank right after the first tank became full. Thus, how fast the first tank filled fully is one of most important factors to determine retention time in the total system. As shown in **Table S4**, when increasing the number of rooms, retention time in the first tank was noticeably increased, while no significant change of retention time

in the second tank was observed. Consequently, the employment of many radiation therapy rooms in a hospital at high recovery rate in FO enables less impact on retention time as well as the treatment of larger amount of radioactive liquid waste, implying that patients can have more opportunities to get radiation treatment as well as hospitals can make more profits by running more radiation facilities.

Figure 9. Simulated retention time as a function of the number of radiation therapy rooms and recovery rate (%) in FO with a fixed tank capacity (48 m^3). Some data not shown in this figure could be not simulated since retention time in the second tank should not exceed to its double in the first tank. The simulation models were developed by adopting the decay rate of radioactive iodine isotope (^{131}I).

Findings from the present study have significant implications for the dewatering of radioactive liquid waste produced by nuclear power plants owing to high rejection property of FO. For example, radioactive liquid waste can be concentrated using FO and more easily

immobilized due to the significantly reduced volume. The DS exhibits low radioactivity due to the high rejection properties of FO membranes, and thus it can be discharged into the sea after concentration of seawater (which has a similar osmotic pressure to that of the DS used in this study) is utilized as the DS. In addition, because radioactive liquid waste rarely contains organics, unlike that from hospitals, FO operation will be more stable due to the lower fouling potential. Despite the many advantages of FO, the impact of high levels of radioactivity on the stability of FO membranes should be assessed because only liquid waste characterized by low-level radioactivity was examined in the present study. For the treatment of radioactive liquid waste by FO, the appropriate measures of radiation protection including a lead barrier, and protective clothing and gloves should be considered due to the concern for health risks.

4. Conclusions

The present study reports for the first time that FO can be successfully used in the concentration of radioactive liquid waste generated by radiation therapy in hospitals. FO was first assessed for the treatment of natural and radioactive iodine (^{125}I) under different pHs and different draw solutions to obtain the optimal operating conditions. FO performance during the treatment of real radioactive liquid waste containing ^{131}I was then investigated in terms of water flux and rejection rate. Finally, the effect of FO concentration on the management system for radioactive liquid waste was simulated. Findings from this study have significant implications for the dewatering of radioactive liquid waste from other sources such as nuclear power plants. The primary findings drawn from this study can be summarized as follows:

- FO rejected both natural and radioactive iodine at a rate of up to 99.85%, which is more than that of the UF and two-pass RO hybrid processes (99.7%) reported in the literature.

- A higher pH leads to higher iodine rejection by enhancing electrostatic repulsion, and NaCl DS achieves higher iodine rejection via the inhibitory effect of RSF on the forward transport of iodine.
- During the treatment of real radioactive liquid waste, the radionuclide ^{131}I was successfully removed by FO, but severe fouling was observed, resulting in a significant reduction in flux.
- Membrane fouling could not be removed by simple hydraulic washing due to the combined presence of organic and inorganic substances on the membrane surface.
- A simulation of required septic tank capacity as a function of recovery rate demonstrated that FO is able to effectively reduce septic tank volume, allowing an increase in the number of radiation therapy rooms (e.g., from 2 rooms to 8 rooms at a 75% recovery rate).

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Highlights

- Medical radioactive liquid waste was treated using FO for the first time.
- FO successfully rejected both natural and radioactive iodine.
- The rejection of FO was optimized for high pH and DS with adequate RSF.
- Actual radioactive liquid waste was effectively concentrated, reducing its volume.
- Severe fouling was observed due to the combined effect of organic and inorganic foulants.