Rotating Reverse Osmosis for Wastewater Reuse

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Background: Reverse osmosis (RO) has long been in use as a physical membrane separation technology, and it may be useful for wastewater reuse for long-term space missions. However, concentration polarization decreases the flux of solvent through the membrane and the rejection of contaminants as a result of an increase in the solute concentration near the membrane surface. Urea, sodium chloride, and detergent (Geropon TC-42) are major contaminants in spacecraft wastewater. In addition, numerous organic contaminants such as 2-(2-butoxyethoxy) ethanol, caprolactam, 2-propanol, formaldehyde, and methanol have also been found at low concentrations in condensate collected from the cabin of the spacecraft. As the length of space missions increases and wastewater is reclaimed for use as potable water, it is necessary to remove all of these contaminants.

Objectives: Our previous work established the concept of a low-pressure rotating reverse osmosis membrane system. The rotation of the cylindrical RO filter produces shear and Taylor vortices in the annulus of the device that decrease the concentration polarization and fouling commonly seen with conventional RO filtration techniques. A mathematical model based on the film theory and the solution-diffusion model agrees well with the experimental results obtained using this first generation prototype. However, based on the model, the filtrate flux and contaminant rejection depend strongly on the transmembrane pressure. Therefore, the goal of our current work is to improve the flux of the device by increasing the transmembrane pressure by a factor of 3 to 4. In addition, the rejections for a wider variety of inorganic and organic compounds typically found in space mission wastewater are measured.

Rejection of Target Contaminants by Selected Membranes: Flat sheet samples of commercially available reverse osmosis, low pressure RO (LPRO), and nanofiltration (NF) membranes have been tested using a dead-end stirred-cell to remove conventional wastewater contaminants (sodium chloride, urea, and ammonium carbonate) and organic contaminants found in spacecraft condensate. By combining experimental rejection results for various compounds with a model based on the size and electrostatic exclusion properties of the membranes, the pore sizes of the membranes are estimated to be 0.33 nm for RO, 0.34 nm for LPRO, and 0.44 nm for NF membranes. The rejections for both organic and inorganic compounds for these membranes are shown in Figure 1. The rejections of 2-(2-butoxyethoxy) ethanol (BEE) and caprolactam are approximately 80% for the RO and LPRO membranes, because their molecular weights/molecular radii, 162 Da/0.32 nm for BEE and 113 Da/0.28 nm for caprolactam, are large enough to be rejected due to size exclusion. The rejection of these compounds is also relatively high (over 60 %) for the NF membrane. The rejection of ionic compounds is also high (over 80 %) for all membranes due to electrostatic exclusion effects. The rejection of 2-propanol is lower than that of NaCl even though these compounds have similar molecular weights due to electrostatic exclusion of the ionic compound. Urea, formaldehyde, and methanol rejections are quite low because the molecules are small and uncharged. As a result, they are difficult to reject...
by size exclusion or by electrostatic exclusion. Furthermore, the rejection of urea is substantially lower than 2-propanol even though they have the same molecular weight of 60.1 Da. This is because the molecular radius of urea (0.18 nm) is smaller than that of 2-propanol (0.26 nm).

![Figure 1](image1.png)

**Figure 1.** Rejection of different compounds for RO, LPRO, and NF. Operating conditions: $\Delta P=800$ kPa; stirring speed=400 rpm; feed concentration=1 mM; recovery=60 %. (a) RO (AK), (b) LPRO (ESPA), and (c) NF (ESNA) ($\blacktriangle$, urea; ■, ammonium carbonate; ○, sodium chloride; ○, methanol; □, 2-(2-butoxyethoxy) ethanol; Δ, caprolactam; ∇, formaldehyde; ◊, 2-propanol).

**Rotating Reverse Osmosis:** A second generation rotating reverse osmosis system has been designed and fabricated to function at a much higher transmembrane pressure than the original system. The new device operates at 500 psi (3450 kPa) compared to the first generation prototype that operated at 150 psi (1035 kPa). The second generation prototype and fluid circuit (Figure 2a) have also been designed so that testing can be conducted for much longer time periods: tests lasting 4 weeks or more compared to a maximum of a 6-hour test conducted with the first-generation prototype.

Preliminary three day tests exhibit high flux (Figure 2b) and high rejection (over 70 % for NaCl, 80 % for $\left(\text{NH}_4\right)_2\text{CO}_3$, 97 % for detergent) for the duration of the experiment while maintaining a high recovery ranging from 75 to 90 %. This recovery is significantly higher than the average of recovery of 25 % for typical spiral wound RO systems, a property that is particular advantageous for maximum water recovery. The second generation device exhibits a flux four times greater than that of the first generation prototype primarily due to the higher operating pressure. These experiments are the first step in the validation of rotating reverse osmosis at high transmembrane pressures over long time periods.

![Figure 2](image2.png)

**Figure 2.** (a) Photograph of second generation rotating reverse osmosis filter and fluid circuit and (b) Flux as a function of time for a 3 day experiment. Operating conditions: LPRO (ESPA); $\Delta P=500$ psi; rotation rate=90 rpm; recovery=75 to 90 %; wastewater composed of NaCl (1,000 mg/L), $\left(\text{NH}_4\right)_2\text{CO}_3$ (3,429 mg/L), and detergent (2,000 mg/L).

Funded by NASA.
Rotating Reverse Osmosis for Wastewater Reuse

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Contributors:
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Filtration Team
Laboratory for Applied Fluid Dynamics

Water Recycle System

Total 11.45 kg

Solid Waste

Food

Condensate

2.27 kg

Hygiene
Flush Water Wash

7.18 kg

Urine 2 kg

Wastewater 9.45 kg

Detergent
Organic Carbon
Inorganic Salts
Microorganisms

Urea

Organic Carbon
Inorganic Salts
Microorganisms

Final 11.45 kg

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## Total Water Requirement for Several Human Space Missions

<table>
<thead>
<tr>
<th>ID</th>
<th>Crew Size</th>
<th>Transit Duration, Days</th>
<th>Surface Stay Duration, Days</th>
<th>Total Number of Duration Days</th>
<th>Water Requirement per Person (kg)</th>
<th>Total Water Requirements (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lunar Human-Mission</td>
<td>3</td>
<td>7</td>
<td>1</td>
<td>8</td>
<td>233</td>
<td>698</td>
</tr>
<tr>
<td>Space Station</td>
<td>3</td>
<td>171</td>
<td>0</td>
<td>171</td>
<td>4,976</td>
<td>14,928</td>
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<tr>
<td>Mars Short Visit</td>
<td>4</td>
<td>1,100</td>
<td>7</td>
<td>1,107</td>
<td>32,214</td>
<td>128,854</td>
</tr>
<tr>
<td>Mars Long-Term Mission</td>
<td>6</td>
<td>1,100</td>
<td>90</td>
<td>1,190</td>
<td>34,629</td>
<td>207,774</td>
</tr>
<tr>
<td>Evolutionary Space Station</td>
<td>10</td>
<td>3,650</td>
<td>0</td>
<td>3,650</td>
<td>106,215</td>
<td>1,062,150</td>
</tr>
</tbody>
</table>
Removal of Molecules and Ions by Reverse Osmosis

Compact
Easy to control
Small energy consumption compared to evaporation (and fewer contaminants)
Independent of gravity

Concentration polarization and membrane fouling are issues

Microfiltration
Ultrafiltration
Nanofiltration
Reverse Osmosis

Suspended Particles
Macromolecules
Dissociated acids
Divalent salts
Sugars
Undissociated acids
Monovalent salts
Water
## Key Contaminants

### Urine and Flush:

<table>
<thead>
<tr>
<th>Compound</th>
<th>MW (g/mol)</th>
<th>Radius (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Urea</td>
<td>60.1</td>
<td>0.18</td>
</tr>
<tr>
<td>Ammonium carbonate</td>
<td>96.1</td>
<td>Cation: 0.125  Anion: 0.133</td>
</tr>
<tr>
<td>Sodium chloride</td>
<td>58.5</td>
<td>Cation: 0.184  Anion: 0.121</td>
</tr>
</tbody>
</table>

### Condensate:

<table>
<thead>
<tr>
<th>Compound</th>
<th>MW (g/mol)</th>
<th>Radius (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2-(2-Butoxyethoxy) ethanol</td>
<td>162.2</td>
<td>0.32</td>
</tr>
<tr>
<td>Caprolactam</td>
<td>113.2</td>
<td>0.28</td>
</tr>
<tr>
<td>2-Propanol</td>
<td>60.1</td>
<td>0.26</td>
</tr>
<tr>
<td>Formaldehyde</td>
<td>30.0</td>
<td>0.22</td>
</tr>
<tr>
<td>Methanol</td>
<td>32.0</td>
<td>0.19</td>
</tr>
</tbody>
</table>
Rejection Test: Organic and Inorganic Contaminants

Molecular weight (Da)

- Sodium chloride
- 2-(2-Butoxyethoxy) Ethanol
- Ammonium Carbonate
- Caprolactam
- 2-Propanol
- Formaldehyde
- Urea
- Methanol

LPPO (ESPA); C₀=1 mM; ΔP= 800 kPa (116 psi); stirring speed=400 rpm; pH=7; recovery=60 %

From Stirred-Cell Test

Laboratory for Applied Fluid Dynamics
Filtration Team
Pore Size Calculation

Solvent Flux

\[ J_v = \frac{r_p^2 \Delta P}{8 \mu (\Delta x/A_k)} \]

Solute Conc.

\[ C_p = \frac{C_m K_c \phi}{1 - \exp \left( - \frac{K_c}{K_d} \frac{J_v \Delta x}{A_k} \right) (1 - \phi K_c)} \]

Concentration Polarization

\[ \frac{C_{i,m} - C_{i,p}}{C_{i,b} - C_{i,p}} = e^{\frac{J_v}{k_i}} \]

Steric Factors

\[ K_{i,d} = 1.0 - 2.30\lambda_i + 1.154\lambda_i^2 + 0.224\lambda_i^3 \]

\[ K_{i,c} = 1.0 + 0.054\lambda_i - 0.988\lambda_i^2 + 0.441\lambda_i^3 \]

Key Parameters Measured:

\[ J_i; J_v; C_{i,p}; C_{i,b}; \Delta P \]

Key Parameters Calculated:

\[ C_{i,m}; k_i \]

(Lee and Lueptow, 2001 ES&T)

(RO Membrane)

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Pore Size Calculation

Solvent Flux

\[ J_v = \frac{r_p^2 \Delta P}{8\mu(\Delta x/A_k)} \]

Solute Conc.

\[ C_p = \frac{C_m K_c \phi}{1 - \exp\left( -\frac{K_c}{K_d} \frac{J_v \Delta x}{A_k} \right)(1 - \phi K_c)} \]

Concentration Polarization

\[ \frac{C_{i,m} - C_{i,p}}{C_{i,b} - C_{i,p}} = e^{J_v/k_i} \]

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Key Parameters Measured:

\[ J_i; J_v; C_{i,p}; C_{i,b}; \Delta P \]

Key Parameters Calculated:

\[ C_{i,m}; k_i \]

(Lee and Lueptow, 2001 ES&T)
# Membrane Properties Obtained from Experiments and Model Calculation

Effective Membrane Pore Size \( r_p \) (nm)

<table>
<thead>
<tr>
<th>Compound</th>
<th>Molecular Radius (nm)</th>
<th>RO (AK)</th>
<th>LPRO (ESPA)</th>
<th>NF (ESNA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2-(2-Butoxyethoxy) Ethanol</td>
<td>0.32</td>
<td>0.333</td>
<td>0.327</td>
<td>0.423</td>
</tr>
<tr>
<td>Caprolactam</td>
<td>0.28</td>
<td>0.324</td>
<td>0.327</td>
<td>0.427</td>
</tr>
<tr>
<td>2-Propanol</td>
<td>0.26</td>
<td>0.334</td>
<td>0.349</td>
<td>0.452</td>
</tr>
<tr>
<td>Formaldehyde</td>
<td>0.22</td>
<td>0.335</td>
<td>0.334</td>
<td>0.440</td>
</tr>
<tr>
<td>Methanol</td>
<td>0.19</td>
<td>0.344</td>
<td>0.336</td>
<td>0.448</td>
</tr>
<tr>
<td>Urea</td>
<td>0.18</td>
<td>0.326</td>
<td>0.343</td>
<td>0.448</td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td><strong>0.333</strong></td>
<td><strong>0.336</strong></td>
<td><strong>0.440</strong></td>
<td></td>
</tr>
</tbody>
</table>
Dependence of Rejection on Solute Radius

From Stirred-Cell Test
Rotating Reverse Osmosis

Taylor-Couette Flow + Reverse Osmosis

Reduces Concentration Polarization and Fouling

Semi Permeable RO Membrane

Concentrate

Feed

Permeate

L

d

ω

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Taylor-Couette Flow

\[ Ta = \frac{r_1 \Omega d}{v} \]

Laminar Couette flow \rightarrow Taylor vortices \rightarrow Wavy vortices
Fouling

Deposition of Particles or Solutes on the Membrane Surface

No Rotation

Particle Deposition

Axial Flow
Fouling

Rotation Decreases Fouling
Geometry for Rotating RO

\[ Q_{\text{conc}}(t) = C_{b,j}(L,t) \]

\[ Q_{\text{feed}}(t) = Q_{\text{conc}}(t) + J_y(t)A_m \]

\[ C_{f,i} = C_{b,i}(0,t) \]
Mass Transfer Model

\[ \frac{\partial C_{b,i}(x,t)}{\partial t} = -\frac{1}{S_a} \left( Q_{\text{conc}}(t) + 2\pi \int_x^L J_y(x,t) dx \right) \frac{\partial C_{b,i}(x,t)}{\partial x} + \frac{2\pi \cdot J_y(x,t)}{S_a} C_{b,i}(x,t) - \frac{2\pi \cdot J_{s,i}(x,t)}{S_a} \]

- **Unsteady Term**
- **Concentrate Flow**
- **Input Flow**
- **Permeate Flow**

**Water Transport**

**Solute Transport**

**Concentration Polarization**

**Pressure Drop**

\[ J_y = L_y (\Delta P - P_{\text{loss}}) \]

\[ J_{s,i} = J_y C_{p,i} = L_{s,i} (C_{m,i} - C_{p,i}) \]

\[ \frac{C_{m,i} - C_{p,i}}{C_{b,i} - C_{p,i}} = e^{\frac{J_y}{k_i}} \]

\[ P_{\text{loss}} = \sum_{i} \Delta \Pi_i + \Delta P_{\text{rot}} + \Delta P_{\text{axis}} + \rho g x \]
Modeling Operating Conditions

> 40 L/m²-hr Flux
> 80% Rejection
> 80% Recovery

$\omega = 100$ rpm

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Increasing the Flux

Curves of Constant Flux (L/m²/hr)

To Increase Flux: Increase Pressure

*(Lee and Lueptow, 2001 JMS)*
First vs. Second Generation Design

First:
150 psi and 4 to 6 hr tests

Second:
> 500 psi and 24 hr to 3+ month tests
First vs. Second Generation Design

First:
150 psi and 4 to 6 hr tests

Second:
> 500 psi and 24 hr to 3+ month tests
First vs. Second Generation Design

First:
150 psi and 4 to 6 hr tests

Second:
> 500 psi and 24 hr to 3+ month tests
Long Term Testing

Fluid Circuit Diagram

- Reservoir
- Pump
- Concentrate Line
- Motor
- Feed Line
- Pressure Regulator
- Filter
- Filtrate Line

Rotating RO
Comparison of Rotating RO with Non-Rotating RO

Flux

\[ J(t) \text{ (L/m}^2\text{-hr)} \]

\[ \omega=90 \text{ rpm} \]

\[ \omega=0 \text{ rpm} \]

Rejection

\[ R_i(t) \]

\[ \Delta P= 1000 \text{ kPa}, \ Q_{\text{conc}}= 0 \text{ ml/min} \]

\[ (\text{NaCl})_2\text{CO}_3 \]

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Preliminary Results: 24 Hour Test

LPRO (ESPA); ΔP= 3450 kPa (500 psi); rotation speed=90 rpm; recovery=83 %
Microgravity Issue: Bubbles

- Blocking inlet or outlet conduits
- Blocking membrane at inflow regions between vortices
Summary

• Characterization of Membranes
  – Rejection depends on pore radius

• Rejection Mechanisms
  – Size exclusion for organic compounds
  – Electrostatic exclusion for ionic species

• Developed a second generation Rotating RO system
  – High flux
  – High rejection
  – High recovery

• Model for Rotating RO based on the solution-diffusion model with the film theory

• Experimental flux and rejection match the model

\[ J_v = L_v (\Delta P - P_{loss}) \]

\[ J_{s,i} = J_v C_{p,i} = L_{s,i} (C_{m,i} - C_{p,i}) \]
NASA-NCMR 2004

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Funded by NASA

(http://quest.arc.nasa.gov/space/)
Objectives

- Characterization of RO membranes for key contaminants
- Analysis of rejection for key inorganic and organic compounds by RO membranes
- Theoretical model for rotating RO
- Effectiveness of rotating RO experimentally to verify our theoretical model
Rejection Test: Urea and Ammonium Carbonate

Urea Hydrolysis

\[ CO(NH_2)_2 + 3H_2O \overset{Urease}{\longrightarrow} 2NH_4^+ + HCO_3^- + OH^- \]

From Stirred-Cell Test

\[ \Delta P = 800 \text{ kPa (116 psi)}; \text{stirring speed}=400 \text{ rpm}; \text{pH}=7; \text{recovery}=60 \%; \text{urea}=2,000 \text{ mg/L, ammonium carbonate}=3,429 \text{ mg/L} \]
Preliminary Results: 3 Day Test

LPRO (ESPA); ΔP= 3450 kPa (500 psi); rotation speed=90 rpm; recovery=75-90 %