Supporting Information

Uranium rejection with nanofiltration membranes and the influence of environmentally relevant mono- and divalent cations at various pH

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Bench scale system

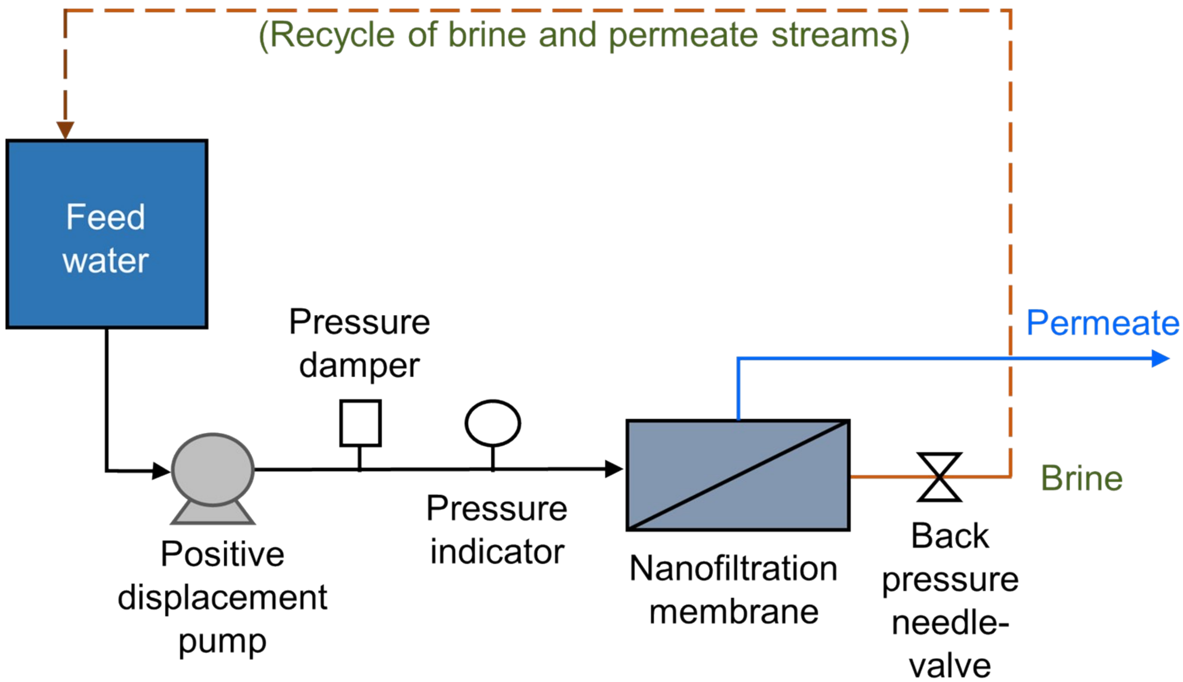

Figure S1: Schematic of bench scale membrane system. During experimental operation, the brine and permeate stream is recycled back into the feed water tank. During sampling and taking permeate flux readings, the permeate stream is collected.

Uranium speciation modeling

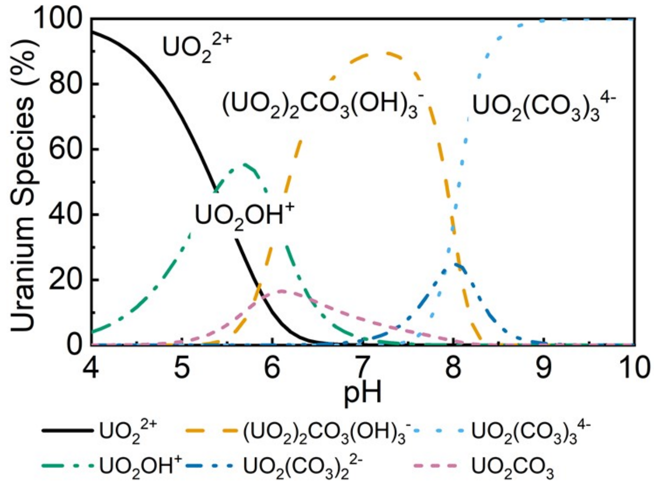

Figure S2: Uranium speciation modeling using Minteq 3.1. Computation input 1g·L^{-1} NaCl with carbonate simulated as open to the atmosphere (0.38 · 10^{-3} atm CO_2), uranium concentration was 150 µg·L^{-1}.

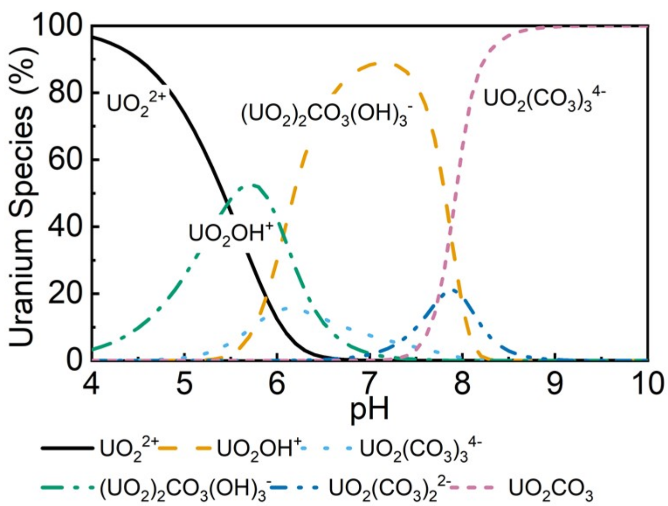

Figure S3: Uranium speciation modeling using Minteq 3.1. Computation input 1g·L^{-1} $MgCl_2$ with carbonate simulated as open to the atmosphere ($0.38 \cdot 10^{-3}$ atm CO_2), uranium concentration was 150 µg·L^{-1}.

Determination of mass transfer coefficient for calculation of concentration polarization.

The concentration polarization modulus (**Equation 1** in main text) was determined by first obtaining the mass transfer coefficient (*k*) from the Sherwood number (**Equation S1**) based on turbulent flows in the following equation.

$$S_h = 0.04Re^{0.75}Sc^{0.33} \qquad \text{(S1)}$$

Where *Re* is the Reynolds number (6346.93), *Sc* is the Schmidt number (476.827). The Sherwood number is then used to calculate the mass transfer coefficient (*k*) from the following equation.

$$k = \frac{S_h D}{d_h} \qquad \text{(S2)}$$

Where *D* is the diffusion coefficient ($2.1 \cdot 10^{-9}$ m^2·s^{-1}), d_h is the hydraulic radius ($3.91 \cdot 10^{-3}$ m). Results of concentration polarization modulus (**Eq.3** in main text) are shown in **Table S1** and **S2**.

Table S1: Concentration polarization modulus for NF90 for each experimental condition across a range of pH.

	pH				
Solution	4	5.5	7	8.5	10
U	1.137	1.144	1.144	1.147	1.143
U + 1 g/L NaCl	1.084	1.092	1.088	1.086	1.087
U + 5 g/L NaCl	1.025	1.031	1.029	1.028	1.027
U + 1 g/L $MgCl_2$	1.106	1.106	1.103	1.101	1.099
U + 5 g/L $MgCl_2$	1.050	1.052	1.050	1.051	1.052
U + 1 g/L $CaSO_4$	1.095	1.102	1.101	1.104	1.100

Table S2: Concentration polarization modulus for NF270 for each experimental condition across a range of pH.

	pH				
Solution	4	5.5	7	8.5	10
U	1.235	1.216	1.217	1.205	1.220
U + 1 g/L NaCl	1.218	1.203	1.171	1.171	1.176
U + 5 g/L NaCl	1.192	1.167	1.142	1.145	1.152
U + 1 g/L $MgCl_2$	1.162	1.167	1.169	1.176	1.175
U + 5 g/L $MgCl_2$	1.090	1.098	1.112	1.125	1.131
U + 1 g/L $CaSO_4$	1.180	1.173	1.168	1.164	1.161

Solute Flux

Solute flux (*Js*) was calculated employing the following equation:

$$j_s = C_{permeate} \cdot J_w \qquad \text{(S3)}$$

Where $C_{permeate}$ is the permeate feed concentration, and J_w is the water flux.

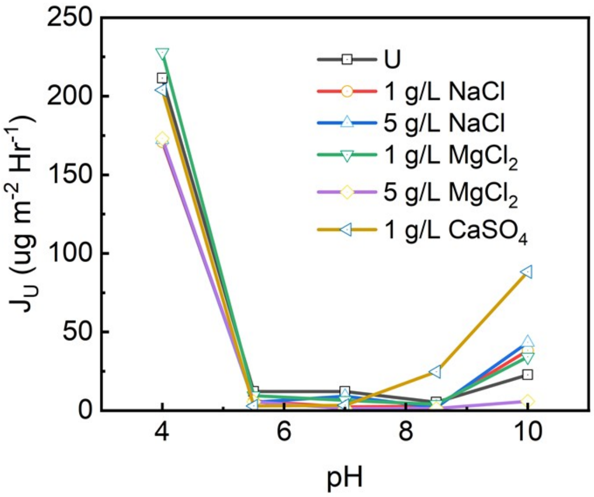

Figure S4: Influence of uranium-containing salt solutions on uranium flux for NF90 membrane. Separate experiments were conducted for each salt solution. All solutions in these tests also contained 150 $\mu g \cdot L^{-1}$ of uranium, operated at 22°C and 75 psi.

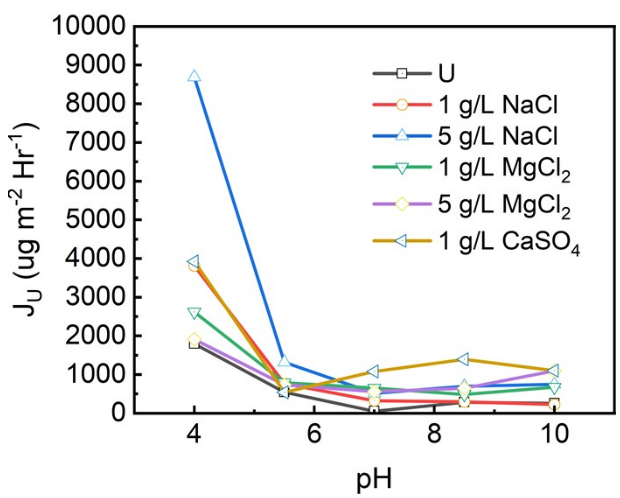

Figure S5: Influence of uranium-containing salt solutions on uranium flux for NF270 membrane. Separate experiments were conducted for each salt solution. All solutions in these tests also contained 150 μg·L^{-1} of uranium, operated at 22°C and 75 psi.

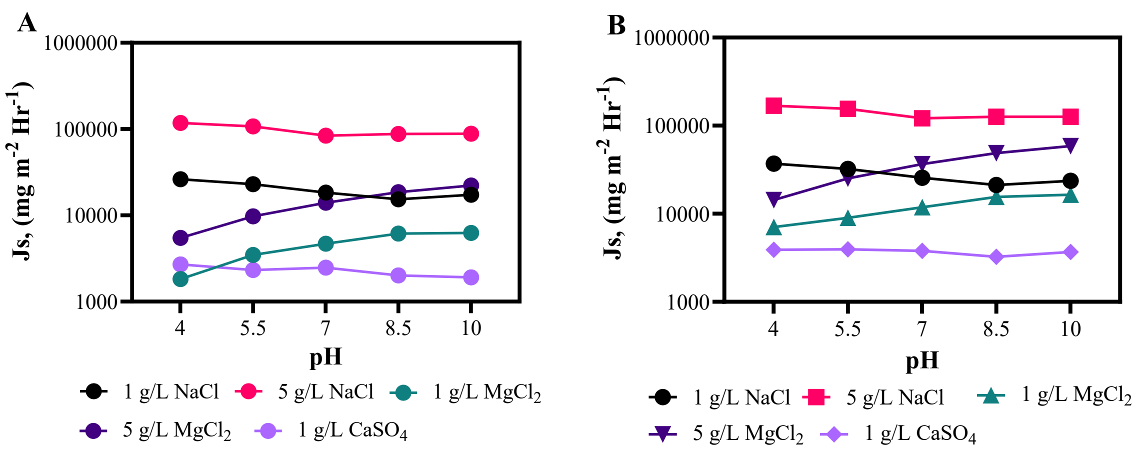

Figure S6: NF 270(A) Cation, (B) Anions. Influence of uranium-containing salt solutions on solute flux. Flux is calculated for Na^+ and Cl^- in 1 g·L^{-1} NaCl solution for NF90 membrane across a range of pH. All solutions in these tests also contained 150 μg·L^{-1} of uranium, operated at 22°C and 75 psi.

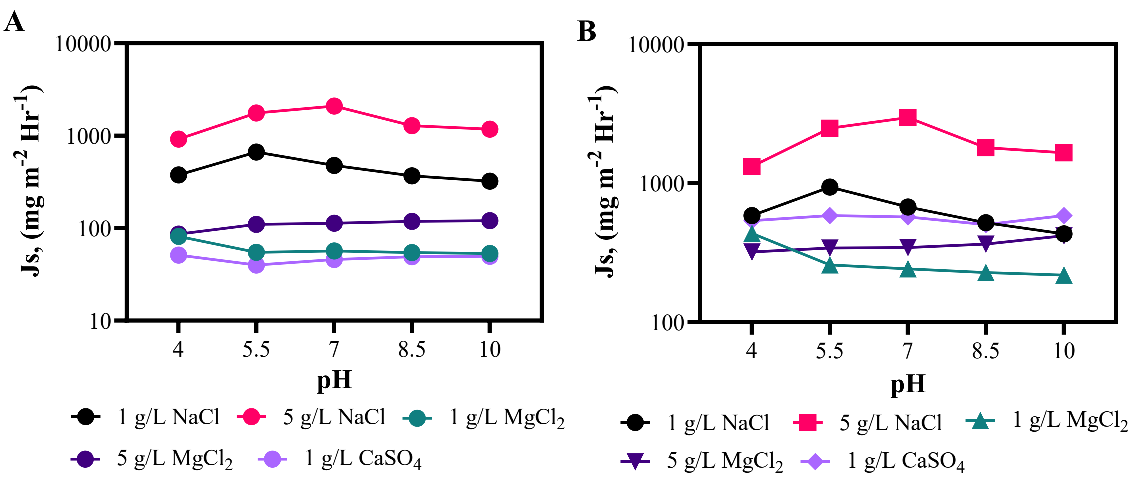

Figure S7: NF 90(A) Cation, (B) Anions. Influence of uranium-containing salt solutions on solute flux. Flux is calculated for Na^+ and Cl^- in 1 g·L^{-1} NaCl solution for NF90 membrane across a range of pH. All solutions in these tests also contained 150 µg·L^{-1} of uranium, operated at 22°C and 75 psi.

Water Flux Performance

Water flux (Jw) was calculated employing the following equation:

$$J_w = \frac{V}{t \cdot A} \qquad \text{(S4)}$$

Where V is the volume (L) of permeate water collected during time t (h^1) and A is the area (m^2) of the membrane surface.

Water volumetric flux can also be stated as a function of solvent permeability constant (A_w), hydrostatic pressure (P), and osmotic pressure (π) through the following equation:

$$J_w = A_w(\Delta P - \Delta\pi) \qquad \text{(S5)}$$

Where ΔP is the applied pressure difference between the feed solution and the permeate solution. $\Delta\pi$ is the calculated osmotic pressure difference between the feed and the permeate streams.

$$\pi = icRT \tag{S6}$$

Where *i* is the Van Hoff coefficient, R is the gas constant (8.314 kPa·m^3·mol^{-1}·K^{-1}), and *T* is the temperature (*K*).

The addition of NaCl, $MgCl_2$, and $CaSO_4$ were chosen as typical mineral solutes found in natural groundwater (Fortune et al. 2009). The NF90 (**Figure S8A**) exhibited water fluxes ranging between 8 – 45 (L·m^{-2}·hr^{-1}) between pH 4 -10. The NF270 (**Figure S8B**) exhibited water fluxes between 35 – 97 (L·m^{-2}·hr^{-1}) between the same pH 4 – 10. The pH variation resulted in marginal changes to water production (flux). Therefore, next we examined how the ionic strength of the solution affects water flux. The addition of ions had a negative effect on both membrane (NF90 and NF270) water production performances. This observation is expected as the osmotic pressure for each solution increases by the addition of ions. Osmotic pressures were calculated **(Equation S6)** for each solution and listed in decreasing osmotic pressure: 5 g·L^{-1} NaCl, 5 g·L^{-1} $MgCl_2$, 1 g·L^{-1} NaCl, 1 g·L^{-1} $MgCl_2$, 1 g·L^{-1} $CaSO_4$, U, DI. (30.5psi, 18.7psi, 6.1psi, 3.7psi, 2.6psi, 0.0psi). From the NF90 membrane experiments, water flux in increasing order for the experimental solutions are as follows: 5 g·L^{-1} NaCl, 5 g·L^{-1} $MgCl_2$, 1 g·L^{-1} NaCl, 1 g·L^{-1} $CaSO_4$, 1 g·L^{-1} $MgCl_2$, U, DI. The increasing order of water flux for NF 270 membrane experiments is as follows: 5 g·L^{-1} $MgCl_2$, 5 g·L^{-1} NaCl, 1 g·L^{-1} NaCl ~ 1 g·L^{-1} $CaSO_4$ ~ 1 g·L^{-1} $MgCl_2$, U, DI.

Our results showed a correlation between the reduction of water flux to an increase of solute concentration in the feed solutions. The addition of solute ions increased the solution osmotic pressure, followed by the reduction of water flux due to the need for additional external pressure to exceed the osmotic pressure to generate water flux, which correlates with **Equation S6.** Depending on the ionic concentration of groundwater, a decrease in water production can be expected in areas with groundwater with high total dissolved solids.

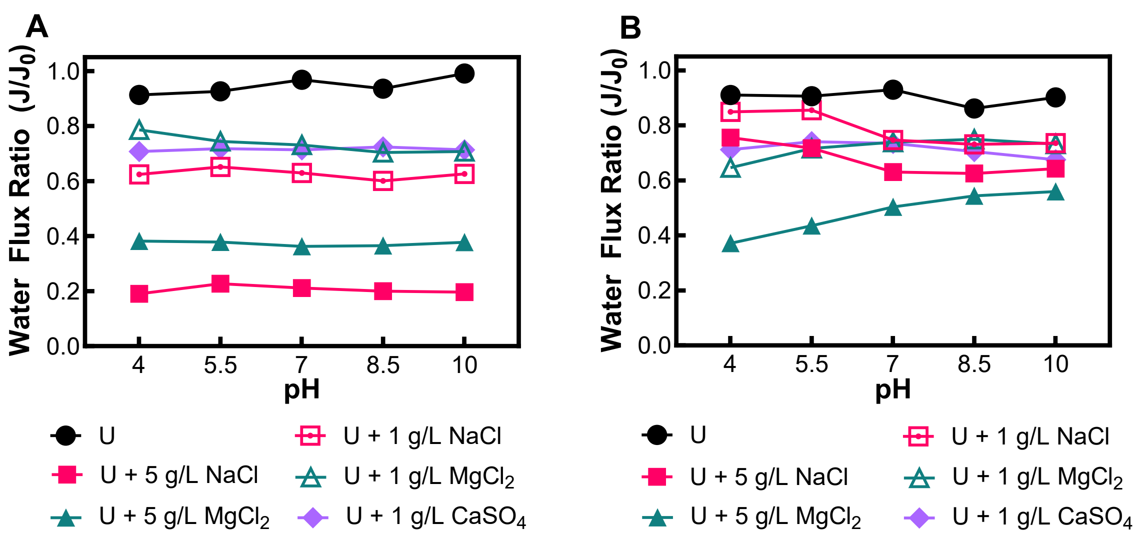

Figure S8. Influence of uranium-containing salt solutions on water permeate production for A) NF90 and B) NF270. All solutions in these tests contained 150 μg·L^{-1} of uranium, operated at 22°C and 75 psi.