

An ultrafiltration- reverse osmosis combined process for external reuse of Weiyuan shale gas flowback and produced water

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Supporting Information:

10 pages

9 figures

1 table

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S1. Scaling potential in the RO unit

The Stiff and Davis saturation index (S&DSI) was used to evaluate the scaling potentials in RO units. S&DSI is defined as the difference between the measured pH and the saturation pH_S :¹⁻³

$$S\&DSI = pH - pH_S \quad (S1)$$

$$pH_S = pCa^{2+} + pAlk + K \quad (S2)$$

where pH is the actual solution pH and pH_S is the pH at saturation; pCa^{2+} and pAlk are the negative of the logarithm of the calcium ion concentration and of the alkalinity, respectively, and K is a factor that accounts for the ionic strength and temperature of the solution.

For the RO influent (i.e, UF permeate), the variations of S&DSI with the RO recovery is shown in **Fig. S1**. The positive values of S&DSI indicate the likely formation of scaling during RO treatment.

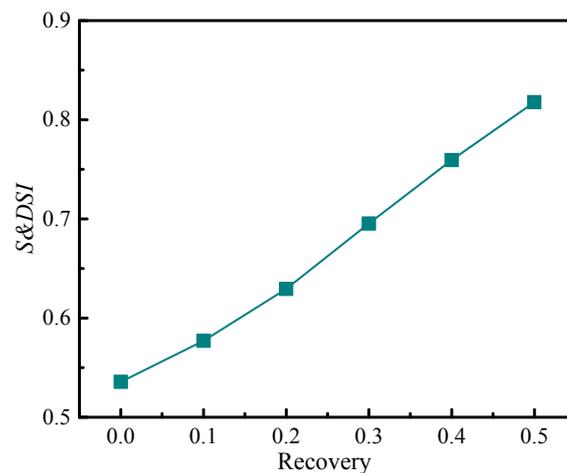


Fig. S1 Scaling potentials (S&DSI) as a function of the RO recovery.

S2. Variations of turbidity and COD in UF permeate

Fig. S2 presents the changes in turbidity and COD of the UF permeate during filtration at a flux of 50 L/(m²·h). The turbidity and COD were both essentially constant throughout the UF, with only slight variations with the filtrate volume (i.e., filtration time).

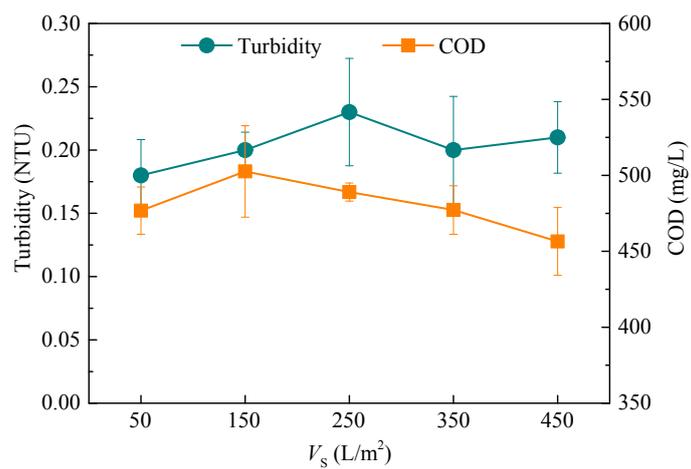


Fig. S2. Variations of turbidity and COD in UF permeate (Flux: 50 L/(m²·h)).

S3. Characteristics of the FPW and its effluent after different treatments

Table S1

Characteristics of the UF feed and its effluent after different treatments.

Constituents	UF feed	UF permeate ^b	RO permeate ^c			
			2.5 MPa	3.5 MPa	4.5 MPa	5.5 MPa
Turbidity (NTU)	49.8	0.19	0.09	0.06	0.07	0.05
TSS (mg/L)	55.4	-	-	-	-	-
TDS (mg/L)	18,900	18,500	1040	361	192	172
Conductivity (mS/cm)	31.14	30.75	2.29	0.76	0.41	0.37
Alkalinity (as CaCO ₃ , mg/L)	600	490	-	-	-	-
COD (mg/L)	530	481	16.4	10.9	7.5	6.0
pH	7.16	8.39	8.12	7.98	8.04	7.91
Total Fe (mg/L)	2.49	2.45	0.62	0.46	0.28	0.15
Total Cu (mg/L)	6.50	5.88	0.43	0.38	0.22	0.09
Na ⁺ (mg/L)	6,950	6870	258	141	76.1	66.8
Ca ²⁺ (mg/L)	233	229	3.02	1.27	0.59	0.36
Mg ²⁺ (mg/L)	26.9	25.7	0.40	0.11	0.05	0.04
K ⁺ (mg/L)	134.5	129.7	6.50	2.74	1.31	1.27
Sr ²⁺ (mg/L)	72.9	64.2	0.75	0.22	0.09	0.06
Ba ²⁺ (mg/L)	135	133	2.23	BDL	BDL	BDL
NH ₄ ⁺ (mg/L)	92.0	89.9	6.59	3.99	2.37	2.27
Mn ²⁺ (mg/L)	BDL ^a	BDL	BDL	BDL	BDL	BDL
Cl ⁻ (mg/L)	11,000	10900	627	190	97	87
SO ₄ ²⁻ (mg/L)	12.1	11.9	0.31	0.10	0.08	0.06
F ⁻ (mg/L)	2.41	1.94	0.09	0.03	0.02	0.02
Br ⁻ (mg/L)	61.0	60.4	3.65	1.23	0.65	0.59
NO ₂ ⁻ (mg/L)	7.74	6.98	0.69	0.15	0.12	0.10

^aBelow the detection limit (0.01 mg/L).

^bThe UF membrane permeate was obtained under the flux of 50 L/(m²·h).

^cThe water recovery was 30%.

S4. Role of backwash conditions on UF membrane fouling and verification of critical flux

UF tests with different backwash parameters were carried out. The experimental protocol was the same as that used for the UF tests described in the manuscript, except for the backwash parameters. A filtration flux of 50 LMH was employed for all tests, and the same volume of backwash water was used. **Fig. S3** shows the comparison of TFI of UF membranes under different backwash conditions. As presented in **Figs. S3a-b**, there was no statistically significant difference in the calculated values of TFI ($p > 0.05$) when the backwash volume was constant. Previous literature on UF of seawater also indicated that backwashing with permeate water resulted in a similar final TMP when the backwash interval (i.e., filtration duration) was not too large.⁴

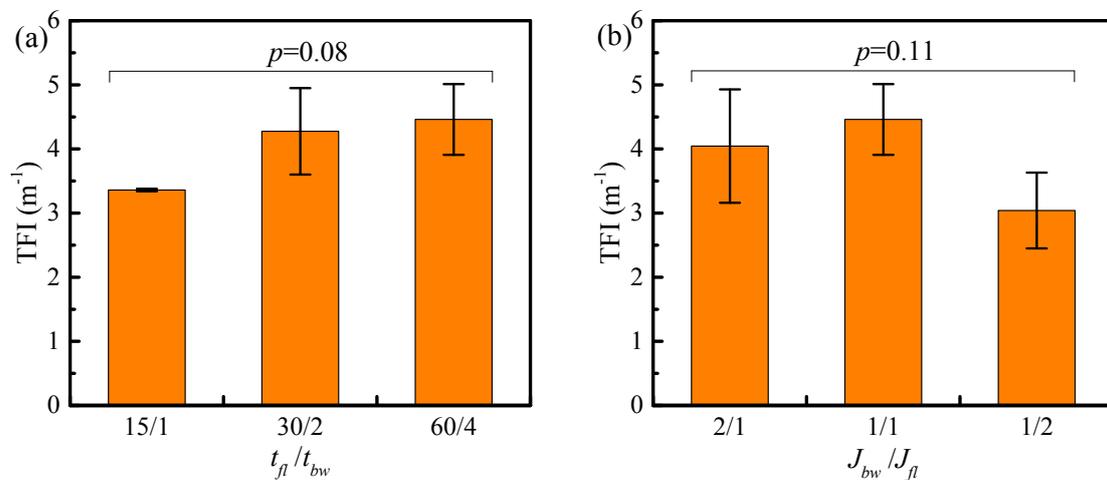


Fig. S3. Comparison of TFI under different backwash conditions with the same volume of backwash water: (a) effect of backwash duration (by maintaining a constant ratio of t_f and t_{bw} , $t_f/t_{bw}=15$, $J_f = J_{bw} = 50$ LMH), and (b) effect of backwash flux ($t_f = 60$ min, $J_f = 50$ LMH; $J_{bw}/J_f = 2/1, 1/1, 1/2$, and corresponding $t_{bw} = 2, 4, 8$ min) (Note: t_f = filtration duration, t_{bw} = backwash duration, J_f = filtration flux = 50

LMH, J_{bw} = backwash flux).

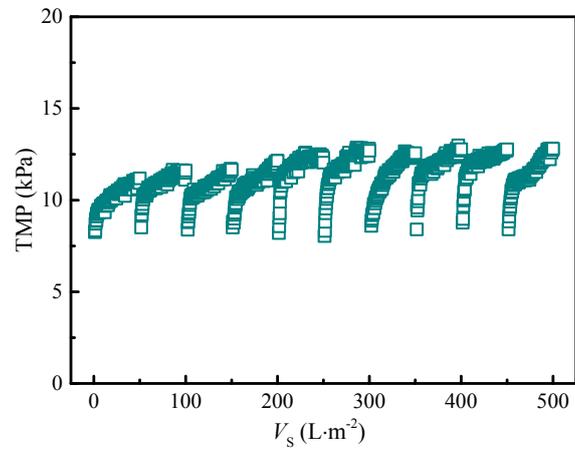


Fig. S4. UF membrane fouling at a flux of 15 LMH.

S5. RO flux and its decline with recovery

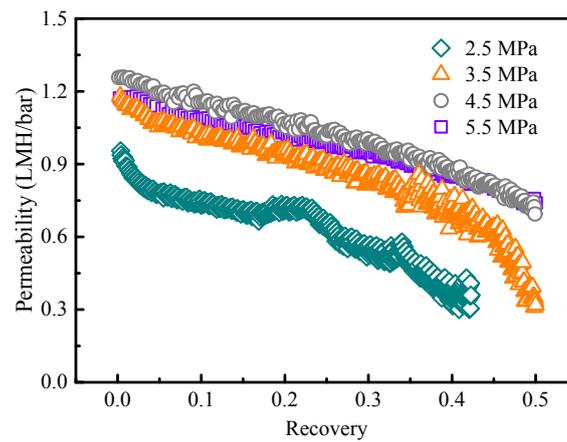


Fig. S5. Plot of RO permeate flux per net pressure driving force as function of recovery.

The flux decline (ΔJ) in the RO unit was calculated by subtracting the measured flux from the pure water flux. Using Eq. (7) in the manuscript, the flux values can be determined by product of hydraulic permeability and osmotic pressure differential, and subtracting these fluxes from the pure water flux led to the flux decline due to osmotic pressure ($\Delta J_{osmotic\ pressure}$). Thus, the ratio of flux decline due to osmotic pressure to RO flux decline was determined, as presented in **Fig. S6**.

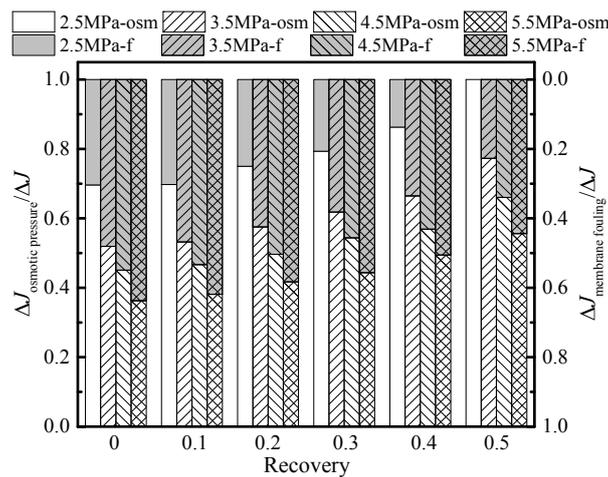


Fig. S6. The ratio of flux decrease due to osmotic pressure in total RO decline flux under different recoveries and pressures (In legend, osm: osmotic pressure; f: membrane fouling).

S6. Determination of solute permeability coefficient

Figs. S7-S8 present the plot of permeate flux as a function of rejection for RO system to determine the solute permeability for TDS and COD of Weiyuan shale gas FPW, respectively. The solute permeability coefficients ω determined using linear regression were 5.1×10^{-8} and 8.4×10^{-8} m/s for TDS and COD, respectively.

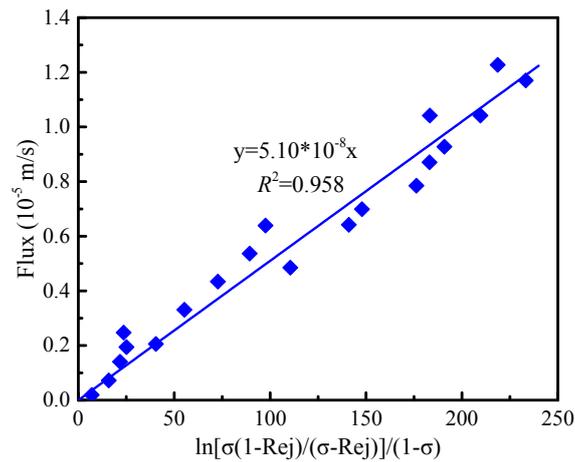


Fig. S7. Plot of permeate flux as a function of TDS rejection for RO system to determine the solute permeability of Weiyuan shale gas FPW.

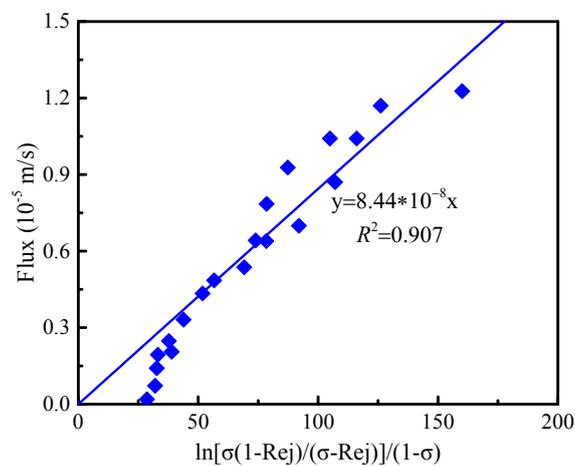


Fig. S8. Plot of permeate flux as a function of COD rejection for RO system to determine the solute permeability of Weiyuan shale gas FPW.

S7. Specific energy consumption of RO unit

The specific energy consumption (SEC) and normalized SEC (SEC_{norm}) for a single-pass RO process were calculated using Eqs. (S4)-(S5):⁵

$$SEC = \Delta P/r \quad (S4)$$

$$SEC_{norm} = R_{ej}/r(1-r) \quad (S5)$$

The SEC and normalized SEC under different recoveries and pressures are illustrated in **Fig. S9**. As presented in **Fig. S9a**, when the recoveries of RO step were 0.3-0.5, the SEC values ranged from 1.4 to 5.1 kWh/m³. These values are consistent with literature reports showing that the SEC was 2.5-7.0 kWh/m³ for RO treatment of shale gas FPW.⁶⁻¹⁰

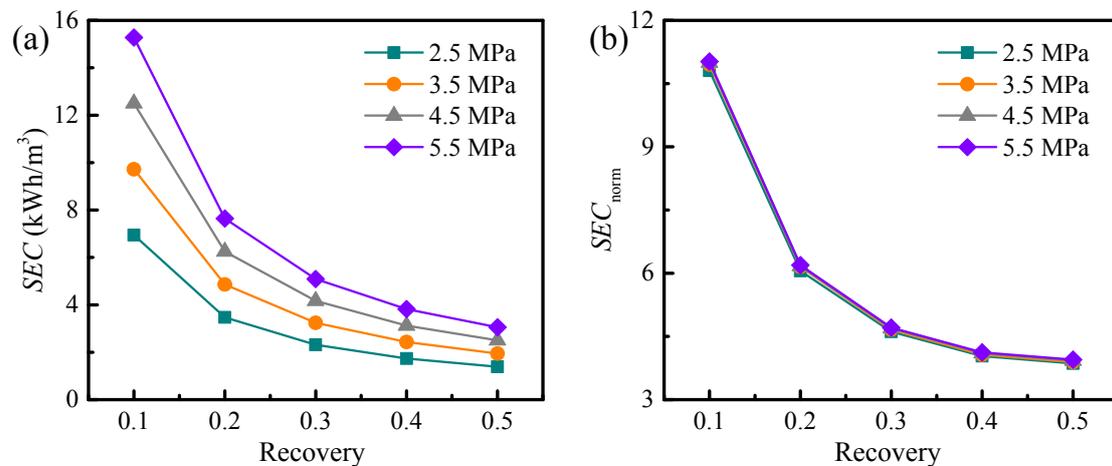


Fig. S9. The SEC and normalized SEC of RO unit under different recoveries and pressures

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