

# Supplementary Information

## **Reducing Chloride Ion Permeation during Seawater Electrolysis using Double-Polyamide Thin-film Composite Membranes**

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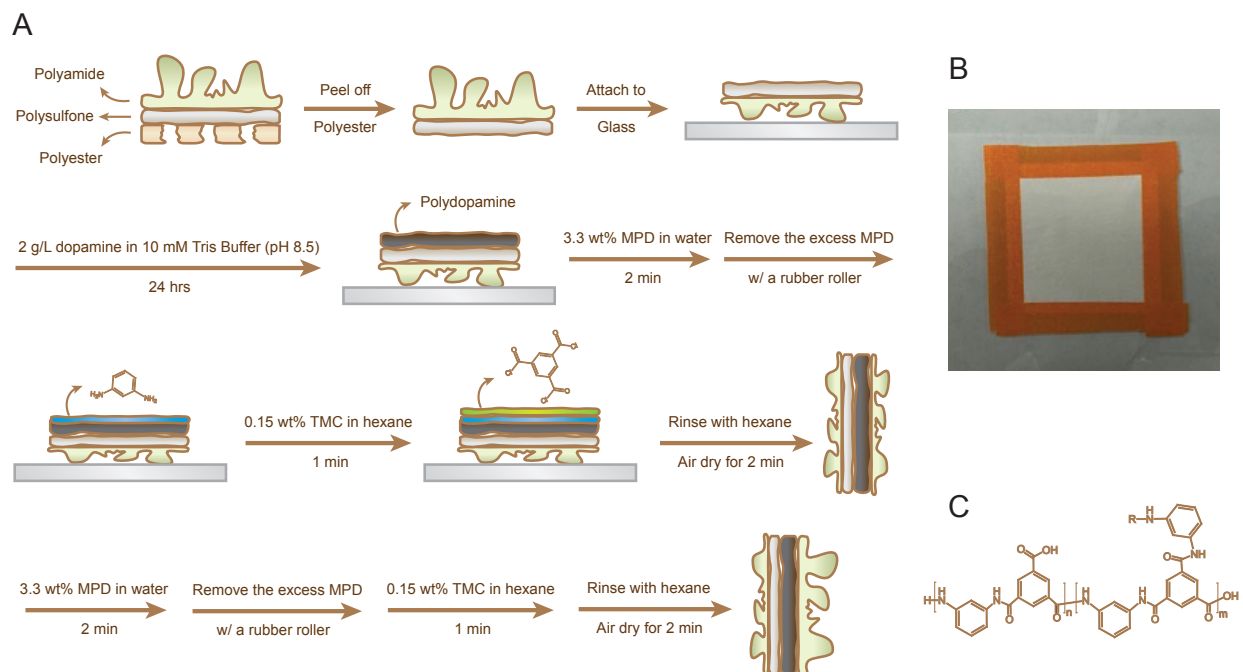
**S1. Ion Permeation Calculation:** Ion permeance ( $P$ ) can be calculated based on the ion concentration on the permeate side ( $C$ ):

$$P = \frac{CV}{tA} \quad (1)$$

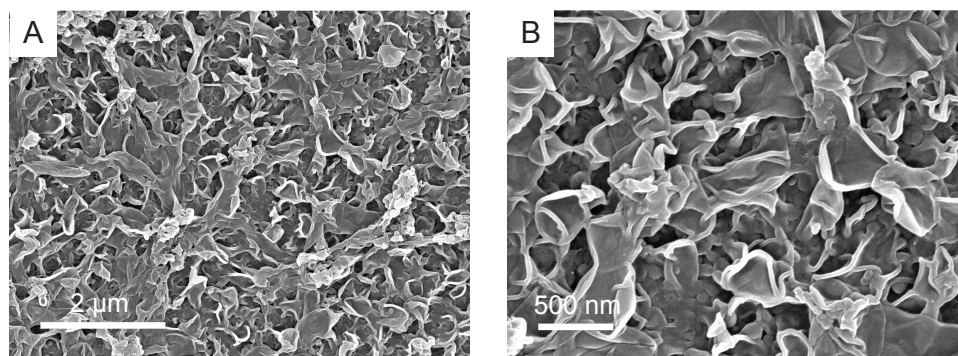
where  $V$  is the electrolyte volume (100 mL),  $t$  is the duration of the experiment, and  $A$  is the membrane area (4 cm<sup>2</sup>). Therefore, the ratio of change in ion permeation can be estimated by:

$$\frac{P_2 - P_1}{P_1} = \frac{\frac{C_2(T)V}{TA} - \frac{C_1(T)V}{TA}}{\frac{C_1(T)V}{TA}} = \frac{C_2(T) - C_1(T)}{C_1(T)} \quad (2)$$

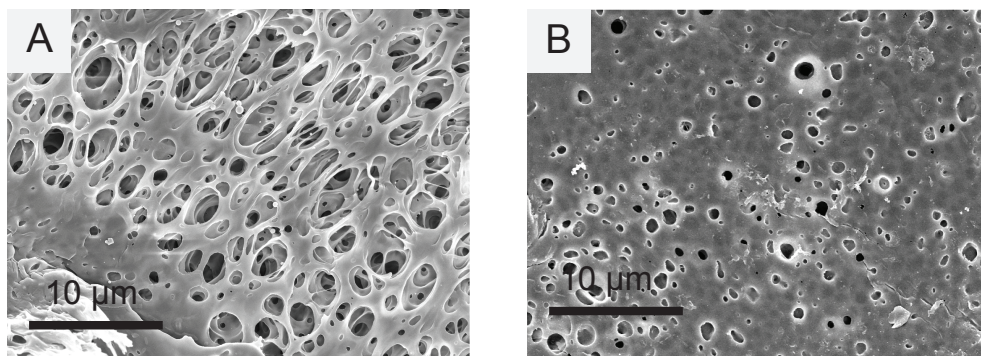
where  $C(T)$  is the ion concentration at a determined experimental duration ( $T$ ; 360 min in our case).



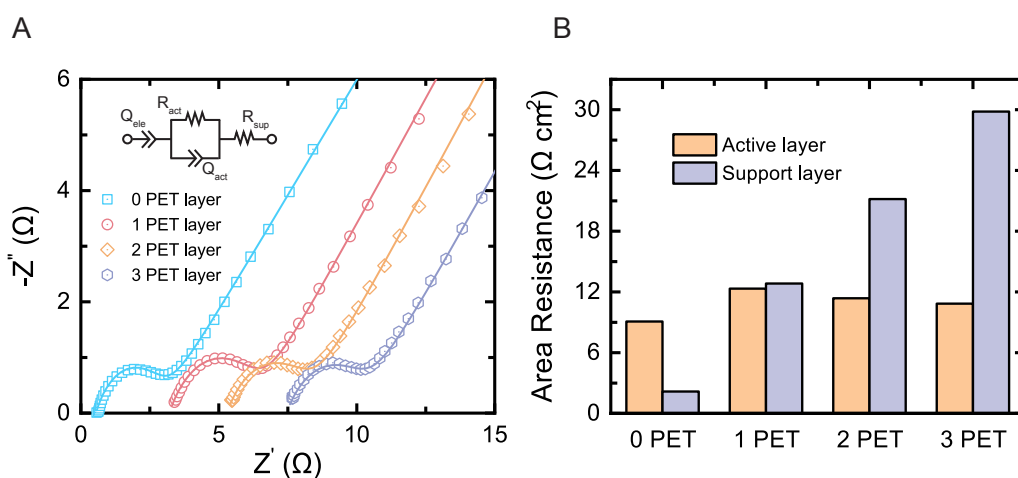
**Figure S1.** (A) Schematic diagram showing the construction of double-polyamide thin-film composite membranes with the intermediate polydopamine layer. (B) Photograph showing the attachment of a pristine polyamide membrane to the glass plate with the polysulfone layer facing up. (C) Chemical structure of the fully aromatic polyamide layers on both sides of the double-polyamide membranes.



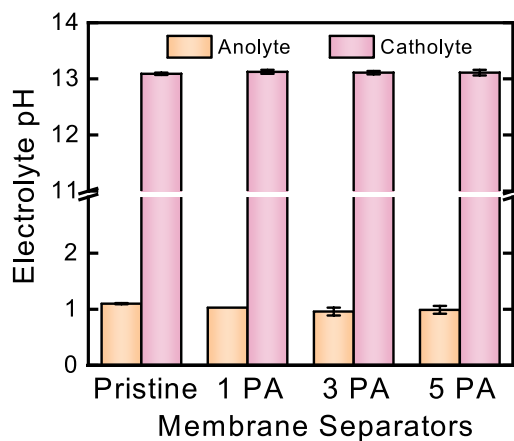
**Figure S2.** SEM images of the top surface of the pristine PA membranes at the magnitude of (A) 50 k and (B) 120 k.



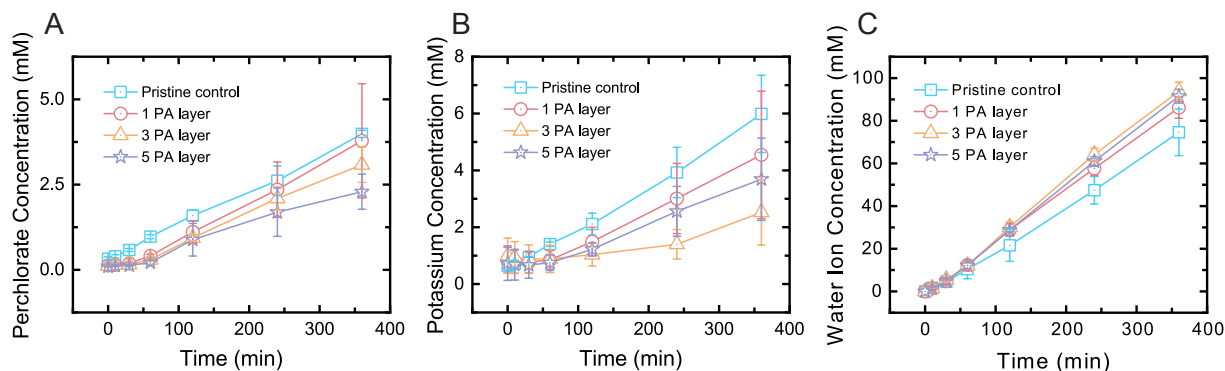
**Figure S3.** SEM images of (A) the pristine PSF control and (B) PDA-coated PSF layer.



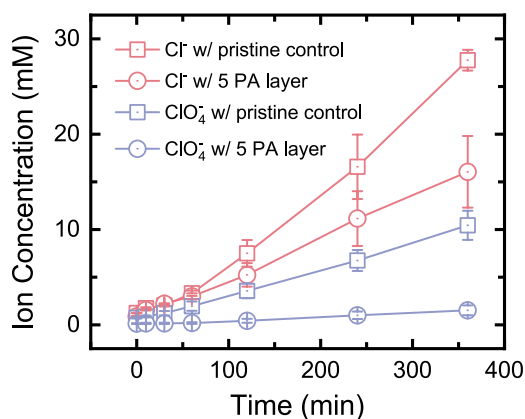
**Figure S4.** (A) EIS spectra of the electrolyzer assembled with PA membranes with varied PET layers at the initial of the operation of electrolysis. (B) Resistance of the active layer (PA layer) and the support layer (PSF and PET layer) obtained through fitting the EIS spectra.



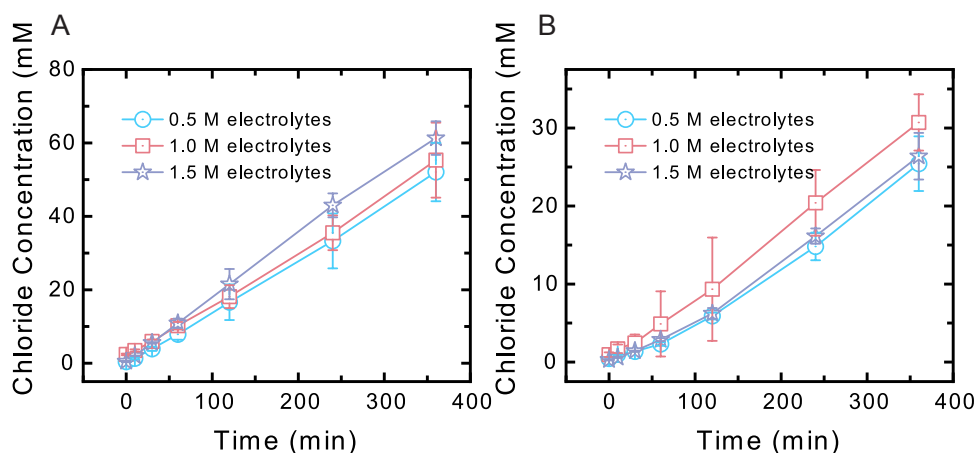
**Figure S5.** pH of the anolyte and catholyte after the operation of the electrolyzers assembled with double-polyamide TFC membranes with varied PA thickness.



**Figure S6.** (A) Perchlorate and (B) potassium permeation, and (C) proton and hydroxide accumulation within the electrolyzers assembled with double-polyamide thin-film composited membranes with varied polyamide thickness.



**Figure S7.** Anion permeation (chloride and perchlorate) within the electrolyzers assembled with double-polyamide thin-film composited membranes with varied polyamide thickness. The anolyte was sodium perchlorate, and the catholyte was sodium chloride.



**Figure S8.** (A) Sodium and (B) chloride permeation through the pristine membrane at the electrolyte concentrations of 0.5, 1.0, and 1.5 M.