

**Supplemental Information**

**Promoting water-splitting in Janus bipolar ion-exchange resin wafers for electrodeionization**

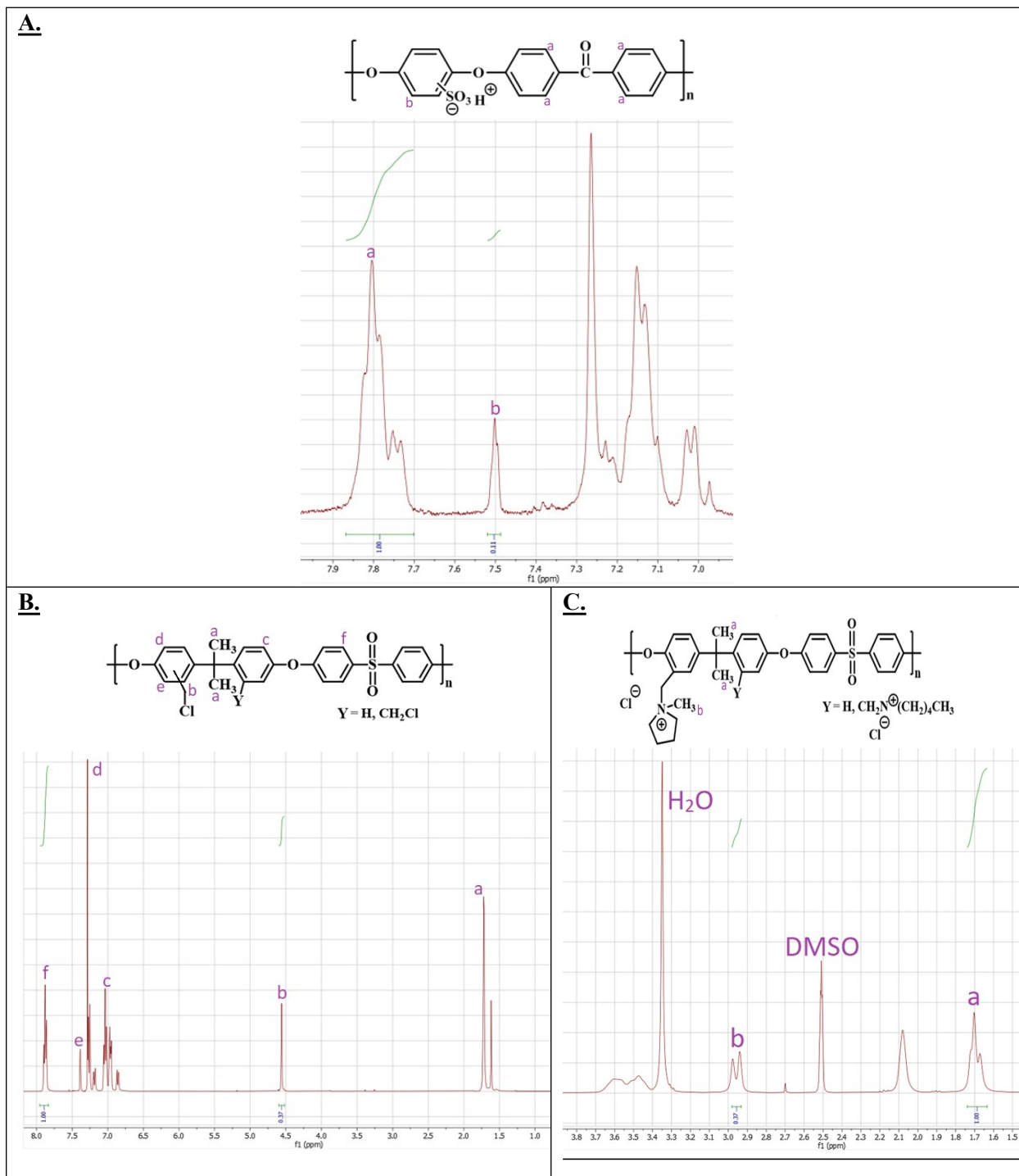
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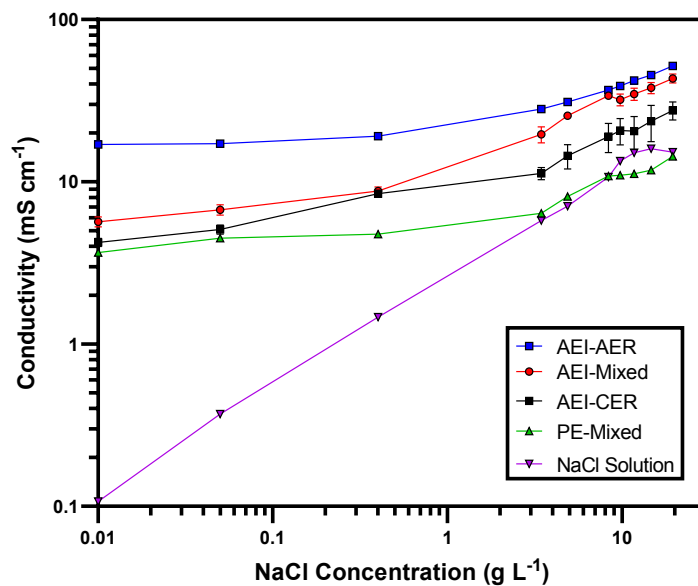
**Figure S1.** (a)  $^1\text{H}$  NMR spectra for sulfonated poly(arylene ether ether ketone) (SPEEK). (b)  $^1\text{H}$  NMR spectra for chloromethylated poly(arylene ether sulfone) (CMPSf). (c)  $^1\text{H}$  NMR spectra for quaternary benzyl n-methyl pyrrolidinium chloride poly(arylene ether sulfone) (QAPSf).



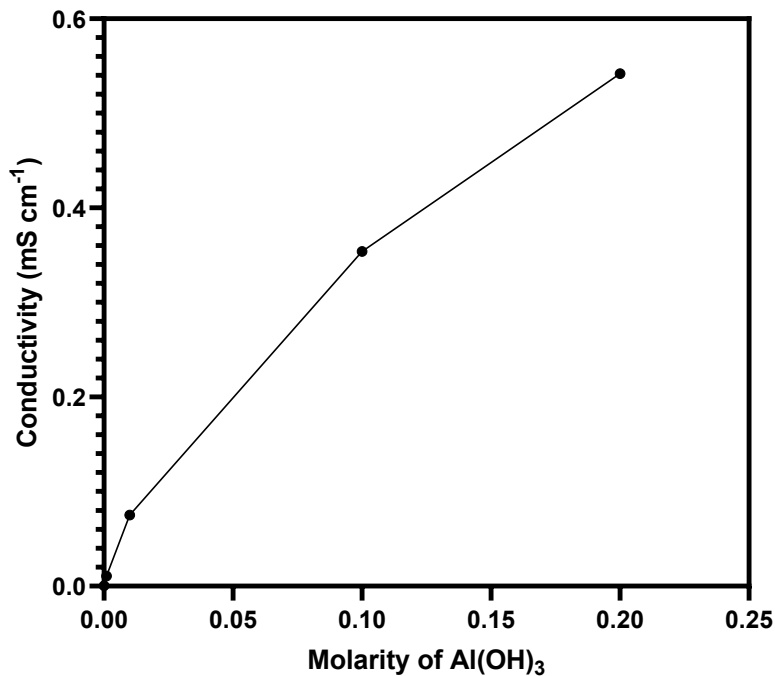
**Figure S2.** CEI-CER RW synthesized without adding polyethylene to the mixture. This RW shows poor mechanical integrity after immersion in water due to chemical incompatibility between the SPEEK CEI and CER.

*Anion exchange ionomer (AEI-AER, AEI-CER, AEI-Mixed) RW*

The resin wafers in Figure S3 were prepared by dissolving QAPSf to a 14 wt % concentration in NMP and mixed with resins and sodium chloride in a 2:2.4:1 ratio by mass. The AEI-AER and AEI-CER RWs were blended with either pure anion or cation exchange resins, respectively, while the AEI-Mixed was blended with a 1:1.3 CER:AER ratio. The mixture was packed into a foil-lined mold and dried at 60°C for 12 hours to remove the residual solvent. The mold was hot pressed to 150°C for 2 hours with 2 metric tonnes of force and then allowed to cool to room temperature under load. The resin wafer was immersed in DI water for 20 minutes and repeated three times to dissolve the sodium chloride porosity.



**Figure S3.** Ionic conductivity of RWs formulated with CER, AER, and a mixture of both types of ion-exchange resins with AEI binder. AEI-AER had the highest ionic conductivity. The addition of CER to the AEI-AER matrix (or replacement of AER with CER) reduced the ionic conductivity.



**Figure S4.** Ionic conductivity of suspension solutions containing Al(OH)<sub>3</sub> nanoparticles in DI water.

## **Additional Experimental Methods**

### *Materials*

Purolite PFC100E cation exchange resins (ion-exchange capacity (IEC) = 1.9 eq L<sup>-1</sup>, density = 1.27 g cm<sup>-3</sup>) and Purolite PFA400 anion exchange resins (IEC = 1.3 eq L<sup>-1</sup>, density = 1.07 g cm<sup>-3</sup>) were acquired commercially and used in the RW formulation. Sodium chloride (NaCl) and 99% N-methyl pyrrolidone (NMP) were obtained from VWR and used as is. Aluminum hydroxide nanopowder (Al(OH)<sub>3</sub>) was sourced from U.S. Research Nanomaterials, Inc. Deionized water (DI H<sub>2</sub>O, 18.2 MΩ, < 20 ppb TOC) was obtained from a Milli-Q Millipore Elix 10 system. Commercially available cation exchange, anion exchange, and bipolar membranes were obtained from Ameridia (Neosepta CMX, AMX, and BP; ASTOM Corporation, Tokyo, Japan) and were used for the RW-EDI experiments.

### *Conventional (benchmark PE-Mixed) RW*

Polyethylene (PE) binder, resins, and NaCl were mixed in a ratio of 2:1:0.5 by mass, with the CER and AER mixed in a 1:1.3 ratio. The mixture was packed into a foil-lined mold and hot pressed to 115 °C with 2 metric tonnes of force for 30 minutes. The resultant RW was cooled to room temperature under the load before removal from the mold. The RW was immersed in DI water for 20 minutes and repeated 3 times to dissolve the NaCl porosigen.