Supplementary Information

Self-Crosslinked Alkaline Polymer Electrolyte Exceptionally Stable at 90°C

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| Membrane | TA% | IEC (mmol/g) | Mechanical strength (MPa) | Ionic conductivity (mS/cm) | |
|----------|-----|--------------|------------------------------|----------------------------|------|
| | | | | 20°C | 80°C |
| 1 | 30 | 1.49 | 7.6 | 17.5 | 43.9 |
| 2 | 50 | 1.34 | 20.5 | 15.2 | 39.5 |
| 3 | 70 | 1.21 | 15.3 | 10.7 | 29.6 |

Table S1 Mechanical and conducting properties of xTQAPS membranes with different TA%.[†]

[†] TA% is the nominal fraction of TA in total functional groups of *x*TQAPS.

Experimental details

The synthesis and characterizations of QAPS have been reported in our previous paper^{4a}, synthetic procedures and measurements that have been described therein are not repeated here. Below are new synthetic processes and key measurements involved in this report.

TAPS powder. Chloromethylated polysulfone (CMPS) was dissolved in N,N-dimethylformamide (DMF) to form a solution of 15 wt%, into which diethylamine (50 mol% of the benzyl chloride group on CMPS) was added dropwise, followed by stirring the solution at 40 $^{\circ}$ C for 4 h. The resulting TAPS solution was poured into a beaker containing deionized water, the precipitate was washed repeatedly with deionized water, and then the resulting TAPS powder was dried in a vacuum oven at 40 $^{\circ}$ C for 72 h.

*x***TAPS membrane.** Rather than pouring the as-prepared TAPS solution into water for precipitation, casting it onto a clean, flat glass plate and drying in oven at 60 $^{\circ}$ C for 16 h, and the resulting *x*TAPS membrane was further dried in a vacuum oven at 80 $^{\circ}$ C for 10 h.

TQAPS solution. CMPS was dissolved in DMF to form a solution of 15 wt%, into which diethylamine (30, 50, or 70 mol% of the benzyl chloride group on CMPS, corresponding to Membrane **1**, **2**, **3** in Table 1, respectively) was added dropwise, followed by stirring the solution at 40 °C for 4 h. Cooling down the reaction system to 30 °C, then bubbling the solution with trimethylamine gas for 0.5 h.

*x***TQAPS membrane.** The TQAPS solution was cast onto a clean, flat glass plate, and dried in oven at 60 °C for 16 h and then further dried in a vacuum oven at 80 °C for 10 h.

XPS measurement. XPS measurements were carried out using a Kratos XSAM-800 spectrometer with an Mg K_{α} radiator. The N(1s) signal was collected and analyzed using the software XPSPeak.

Ionic conductivity measurement. The OH⁻ conductivity of fully hydrated TQAPS membranes were measured as a function of temperature ranging from 20°C to 90°C using AC impedance

spectroscopy (IviumStat, The Netherlands). A TQAPS membrane was cut to a square of 2 cm^2 and sandwiched between two electrodes made of Teflon-bound carbon film. The membrane resistance was measured under the open-circuit mode over the frequency ranging from 1 Hz to 1 MHz with oscillating amplitude of 5 mV. The ionic conductivity was calculated as:

$$\sigma = \frac{L}{R_{\rm mem} \times A}$$

where L is the membrane thickness in cm, A the electrode area in cm², and R_{mem} the membrane resistance in Ω .