Regulating hydrogel mechanical properties with an electric field

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S1. Materials and methods.

Materials. Acrylamide (Sigma-Aldrich), N,N'-methylenebis(acrylamide) (MBAA, TCI America), ammonium persulfate (APS, VWR), N,N,N',N'-tetramethylethylenediamine (TEMED, Oakwood Chemical), sodium chloride (EMD Millipore), poly(sodium 4-styrenesulfonate) (PSS, average Mw ~ 70,000, Sigma-Aldrich), PSS 30 wt.% solution in water (average Mw ~ 200,000, Sigma-Aldrich), poly(diallyldimethylammonium chloride) 35 wt.% solution in water (PDADMAC, average Mw < 100,000, very low molecular weight, Sigma-Aldrich), PDADMAC 20 wt.% solution in water (average Mw 200,000 - 350,000, medium molecular weight, Sigma-Aldrich), hydrochloric acid (37 wt.%, EMD Millipore), sodium hydroxide (Research Products International Corp), phenolphthalein (Thermo Scientific), ethanol (Koptec), hydrogen peroxide (30%, Fisher Scientific), sulfuric acid (95% - 98%, VWR), FAA-3-PK-75 (Fuelcell Store), Nafion 117 (Fuelcell Store), deionized water (DI water, Labconco WaterPro P5), Pt electrode, adhesive copper tape, 3D printing filament (eSUN, 1.75MM ePA-CF), polytetrafluoroethylene (PTFE) films, UV cure self-leveling silicone (Silicone Solutions,SS-155).

Semi-interpenetrating polymer network (semi-IPN) hydrogel preparation. 3.1200 g acrylamide, 200 µL MBAA solution in water (10 mg/mL), 4.0908 g NaCl, 104 µL APS solution in water (10 wt.%), 3.2960 g PDADMAC Mw 200,000 - 350,000 20 wt.% solution in water (or 1.8834 g PDADMAC Mw < 100,000 35 wt.% solution in water), and 15.2 mL water were added to a round-bottom flask. After all components were dissolved, 2.8027 g PSS Mw 200,000 30% solution in water (or 4.2040 g PSS Mw ~ 70,000 20 wt.% solution in water) was added to the flask drop-wise. When the PSS was fully dissolved, the flask was placed in an ice water bath and the solution was bubbled with argon for 30 min. 30 µL TEMED was added to the solution and then the solution was injected into a U-shape mold (two pieces of glasses sandwiching either 1 mm or 3 mm spacers). The filler mold was kept in an oven at 50 °C for 3 h. After this, the hydrogel was removed from the mold.

Polyacrylamide (PAam) gel preparation. 3.1200 g acrylamide, 200 μ L MBAA solution in water (10 mg/mL), 104 μ L APS solution in water (10 wt.%), and 20 mL water were added to a round-bottom flask. After all components were dissolved, the flask was placed in an ice water bath and the solution was bubbled with argon for 30 min. 30 μ L TEMED was added to the solution and then the solution was injected into a U-shape mold (two pieces of glasses sandwiching either 1 mm or 3 mm spacers). The mold with solution was kept in an oven at 50 °C for 3 h. After this, the PAam gel was removed from the mold.

Salt-gel preparation. This followed the same procedure as PAam gel preparation, except that 4.0908 g NaCl was added at the beginning in addition to other components.

Nafion 117 ion exchange membrane treatment. The Nafion 117 ion exchange membrane was first placed in 80 °C 3% hydrogen peroxide solution for 1 h. After this, the membrane was rinsed with water and then put in 80 °C water for 2 h. Then the membrane was rinsed again and placed in 80 °C 0.5 M sulfuric acid for 1 h. After being rinsed, the membrane was put in 80 °C 1

M NaOH solution for 1 h. Finally, the membrane was rinsed with DI water and cooled to room temperature.

FAA-3-PK-75 ion exchange membrane treatment. The FAA-3-PK-75 ion exchange membrane was placed in 0.5 M NaCl solution at room temperature for 24 h. After this, the membrane was rinsed with water and then put in 1.5 M NaCl solution at room temperature for 72 h. Finally, the membrane was rinsed with DI water.

Using diffusion to remove salt ions (path 1). For semi-IPN hydrogels with high-level NaCl content, no post-synthesis processing is needed. For hydrogels with medium-level NaCl content, the weight of the sample was first measured then it was put in a glass vial with a known weight. Five times of the weight of the sample of DI water was added to the vial, with the sample completely submerged in water. After the sample was fully equilibrated, it was taken out of the vial and placed in open air. The water in the vial was completely dried out and the weight of the residual solid was measured. The sample underwent a continuous weight loss due to water evaporation until it reached the point where the weight of the sample equaled its original weight minus the weight of the residual solid. After this, the sample was placed in a 100% humidity chamber for equilibration. For hydrogels with low-level NaCl content, the weight of the sample was first measured then it was put in a glass container with 200 mL DI water inside. The sample was completely submerged in water. DI water was changed daily and the whole process took 3 days, meaning there were three cycles. At the end of the third cycle, the sample was taken out and temporarily stored in a 100% humidity chamber. In the meantime, the water from three cycles was collected and completely dried out, and the weight of the residual solid was measured. After this, the sample was taken out and placed in open air. The rest of the procedure was the same as the medium-level one. For specimens shown in the Figure 2a photo, they were soaked in DI water for different lengths of time, then taken out of water and put on the table for photographing.

Using electrodialysis to remove salt ions (path 2). The assembly of the multi-hydrogel device is done according to the respective device scheme. For a forward operation, a piece of 12 mm X 12 mm X 3 mm semi-IPN hydrogel was sandwiched between a piece of 12 mm X 12 mm FAA-3-PK-75 ion exchange membrane and a piece of 12 mm X 12 mm Nafion 117 ion exchange membrane. Then on top of the membranes, a piece of 24 mm X 12 mm X 3 mm PAam hydrogel was placed, respectively. The PAam gels were in direct contact with Pt electrodes, which were connected to the power supply (Zurich DC Power Supply DS-304M) through a piece of copper tape and wires. The electrode on the Nafion 117 side was the cathode, connected to the negative output of the power supply, while the electrode on the FAA-3-PK-75 side was the anode, connected to the positive output of the power supply. An oscilloscope (Digilent Analog Discovery 2) was used to monitor and record the voltage and current during the operation. After all connections were set, the power supply was turned on and the output voltage was set to maximum, which was around 14 V. As the current gradually increased and exceeded 100 mA, the output voltage was adjusted downward to keep the current at around 100 mA. For 15 min and 30 min operations, the power supply was simply turned off when the experiment reached the designated time. For 60 min operations, the output

voltage was adjusted upward to keep the current at around 100 mA as the experiment approached the end, because the resistance increased. When the output voltage reached maximum and the current was lower than 100 mA, the power supply was turned off and the experiment ended, and the time was around 60 min. **For a reverse operation**, the setup and the procedure was the same as the forward operation, except that the connection from the electrodes to the power supply was reversed. The electrode on the Nafion 117 side was connected to the positive output and the electrode on the FAA-3-PK-75 side was connected to the negative output.

Reinjection of salt ions. Both the "same-buffer" path and the "new-buffer" path happened after a 30 min forward operation. **For the "same-buffer" path**, the connection from the electrodes to the power supply outputs were simply swapped, meaning that after the forward operation the electrode on the Nafion 117 side was connected to the positive output and the electrode on the FAA-3-PK-75 side was connected to the negative output. After the connection was completed, the output voltage of the power supply was gently turned upward until the current reached 100 mA. The output voltage was manually adjusted to keep the current at around 100 mA until it reached 30 min. **For the "new-buffer" path**, the setup and the procedure was the same as the "same-buffer" path, except that the used PAam gels were replaced with salt gels with the same dimensions.



Figure S1 A photo showing the device setup for electrodialysis. The orange components are plastic blades working as spacers and the black components are plastic place holders.



Figure S2 The circuit diagram for electrodialysis. Both voltmeters are from the oscilloscope. The current of the device was calculated by dividing the voltage between the resistor by 10 ohm.



Figure S3 Scheme of reverse operation.

Tensile tests. Tensile tests were performed on a Zwick-Roell Z010 system with a 20 N capacity load cell (X-Force HP, Zwick-Roell). The dimensions of the specimens were 80 mm X 10 mm X 1 mm. For each material sample, at least three specimens were tested. The gauge length was set to be 40 mm. The tensile tests were performed with displacement control using engineering strain. The strain rate was 0.1 /s. Engineering stress and strain were calculated from recorded force and travel during tests using the equations below.

$$\sigma_{engineering} = \frac{F}{width \times thickness}$$

 $\varepsilon_{engineering} = \frac{travel}{gauge \ length}$

Compression tests. Compression tests were performed on a Zwick-Roell Z010 system with a 20 N capacity load cell (X-Force HP, Zwick-Roell). The dimensions of the as-prepared specimens were 12 mm X 12 mm X 3 mm. A piece of FAA-3-PK-75 membrane and a piece of Nafion 117 membrane were attached to each side of the gel. Specimens after electrodialysis were tested after the PAam buffer gels were peeled off. For each material sample, at least three specimens were tested. The tests were performed with displacement control using engineering strain. The gauge length was the distance between compression plates when they were in good contact with the specimen. The strain rate was 0.01 /s. Engineering stress and strain were calculated from recorded force and travel during tests using the equations below.

 $\sigma_{engineering} = \frac{F}{length \times width}$ $\varepsilon_{engineering} = \frac{travel}{gauge \ length}$

Conductivity measurements. Residual solid from the water after the diffusion was collected in a vial with a known weight. The solid and the vial were dried in a vacuum oven at 60 °C for 24 h. After they were cooled down to room temperature, the weight of the vial (with solid inside) was measured again, thus the weight of the solid was known. 20 mL DI water was added to the vial and the solid was completely dissolved. Then 40 μ L of the newly-made solution was added to a new vial. 20 mL DI water was added to this new vial and the new solution was thoroughly mixed. A customized apparatus made with high density polyethylene and gold wires was connected to a potentiostat (Gamry Instruments Reference 3000) with a four-electrode connection setup to measure the conductivity. Distances between the wires and the depth of the cell are known. A piece of glass slide was placed on top of the cell and then the solution made previously was added into the cell until it was filled to the edge of the slide. The height of the liquid was measured. Potentiostatic electrochemical impedance spectroscopy (EIS) was run from 1 MHz to 10 kHz with an amplitude of 100 mV root mean square (RMS). The conductivity was calculated using the geometry of the cell and the magnitude of the impedance at 10kHz, where the phase angle was minimal.



Figure S4 The device used to measure conductivity of solutions.

Raman spectroscopy. The Raman spectra of the materials were collected using a WITec Alpha300R Confocal Raman Microscope system. A 785 nm laser source was used for excitation at 120 mW power with 300 l/mm grating. The integration time was 20 s and there were 10 accumulations. A 50X object lens was used.

Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS).

Scanning electron microscopy (SEM) images and energy-dispersive X-ray spectroscopy (EDS) scans were acquired using a Zeiss Gemini 500 Scanning Electron Microscope equipped with an Oxford Instruments Ultim Max detector. An acceleration voltage of 8 keV was applied. Aztec control software was used for EDS mapping.

Titration of acid and base in PAam buffer hydrogels. After a forward operation, the weight of the PAam buffer hydrogels were measured. Then the PAam buffer gel on the Nafion 117 (FAA-3-PK-75) side was placed in a vial with known weight to be prepared for titration to measure the base (acid) in the gel. About 18 mL DI water was added to the vial and the gel was submerged in water for 24 h. After this, the water solution (with the weight measured) was transferred to an Erlenmeyer flask while the weight of residual liquid and the gel in the vial was measured. Two drops of 0.5% phenolphthalein solution in ethanol were added into the flask. HCl solution was prepared by adding DI water to 3.0083 g 37% HCl solution to make a 500.60 g diluted HCl solution. NaOH solution was prepared by adding DI water to 1.2151 g NaOH to make a 474.70 g NaOH solution. To titrate the base solution (Nafion 117 side), HCl solution was added to the flask dropwise until the pink color just disappeared. To titrate the acid solution (FAA-3-PK-75 side), NaOH solution was measured thus the weight of solution added was known. **Gel water content measurements**. The hydrogel sample was dried in a vacuum oven at 60 °C for 24 h. The decrease in the weight of the hydrogel was the weight of water in the original hydrogel.

Titration of base (acid) in "same-buffer" ("new-buffer") path IPN hydrogel. After the operation, the weight of the IPN hydrogel was measured. Then the IPN was placed in a vial with known weight to be prepared for titration to measure the base (acid) in the gel. About 19 mL DI water was added to the vial and the gel was submerged in water for 24 h. After this, the water solution (with the weight measured) was transferred to an Erlenmeyer flask while the weight of residual liquid and the gel in the vial was measured. Two drops of 0.5% phenolphthalein solution in ethanol were added into the flask. HCl solution was prepared by adding DI water to 2.1739 g 37% HCl solution to make a 408.94 g diluted HCl solution. NaOH solution. To titrate the base solution ("same-buffer" IPN), HCl solution was added to the flask dropwise until the pink color just disappeared. To titrate the acid solution ("new-buffer" IPN), NaOH solution was added to the flask dropwise until the pink color just disappeared. To solution added was known.

Spatial variable stiffness haptic device setup. 3D-printed parts (Figure S15) were done with the aforementioned filament and a Prusa MK4 3D printer. The injector temperature was 260 °C and the bed temperature was 95 °C. The front surface of the parts (with the windows) was sanded to be smooth. Low-salt content PAam hydrogel precursor solution was made with 3.1200 g acrylamide, 0.1169 g NaCl, 200 µL MBAA solution in water (10 mg/mL), 104 µL APS solution in water (10 wt.%), and 20 mL water. The windows were sealed with a piece of glass slide and 800 µL PAam hydrogel precursor was injected into each of the window areas with the windows facing down. The parts were then put into an oven at 50 °C for an hour. Then the parts were taken out from the oven and the glass slides were removed. One piece of Pt electrode was put into one printed part and the top of it was covered with adhesive copper tape (gray area of Figure S15). The gap between the electrode and the part was sealed with silicone. The inside of the parts (the side with Pt electrode) was filled with 0.1 M NaCl solution. The front of one piece of the window (the green area) PAam hydrogel was covered with either the Nafion 117 membrane or the FAA-3-PK-75 membrane while the other (the blue area) piece was covered with PTFE film. The semi-IPN hydrogel was then sandwiched between two parts, one with Nafion 117 and the other with FAA-3-PK-75, and the electrodes and ion-exchange membranes should be facing each other through the semi-IPN hydrogel.

Device operation. The electrode on the Nafion 117 side was connected to the counter electrode of the Gamry Reference 3000 Potentiostat while the electrode on the FAA-3-PK-75 side was connected to the working electrode. For a forward operation (to stiffen the gel), a 25 V voltage was applied for 5 min. For a reverse operation (to soften the gel), a -25 V voltage was applied until the integral of current vs. time was about the same as the previous forward operation.

Indentation tests. The 3D printed part with the FAA-3-PK-75 ion-exchange membrane was removed from the semi-IPN hydrogel. The hydrogel and the other 3D printed part was placed on top of a compression test plate, with the hydrogel up. Cyclic indentation tests were performed on a Zwick-Roell Z010 system with a 20 N capacity load cell (X-Force HP, Zwick-Roell). A 10 mm diameter steel ball was used as the indenter. The indentation depth was 1 mm and the travel speed was 5 mm/min. For each test there were two cycles of loading and unloading. The indentation was centered on the IPN on top of the center of the windows.

S2. Calculations.

Removal of salt ions (NaCl) via path 1 diffusion. The conductivity of the solution was calculated with the following equation:

 $\sigma = \frac{width}{thickness \times height \times |Z|}$

where width is 35 mm, thickness is 1.95 mm, and the height was measured for every experiment. From the conductivity σ (S/m), we calculated the removal ratio of salt ions in the semi-IPN hydrogels. The molar conductivity Λ of NaCl is necessary for the calculation. We used $\Lambda = 123.68 \times 10^{-4} m^2 \cdot S \cdot mol^{-1}$ (0.001 mol/L) for low-NaCl samples and $\Lambda = 124.44 \times 10^{-4} m^2 \cdot S \cdot mol^{-1}$ (0.0005 mol/L) for medium-NaCl samples ^[1]. These numbers were chosen based on our estimation of NaCl concentrations in the second vial. The estimation was also verified from our actual calculations, the concentrations of low-NaCl samples being in the 0.00083 ~ 0.00097 mol/L range and the concentrations of medium-NaCl samples being in the 0.00017 ~ 0.00031 mol/L range. The equation for removal ratio is

 $Removal \ ratio \ = \ \frac{58.44 \ \sigma \cdot (m_{2s} + m_{2w}) \cdot m_{1w}}{1000000 \times 0.15078 \ A \cdot m_{2s} \cdot m_{q}}$

where m_{2s} (g) is the weight of solution added to the second vial, m_{2w} (g) is the weight of the DI water added to the second vial, m_{1w} (g) is the weight of the DI water added to the first vial, and m_g (g) is the weight of the gel samples. 0.15078 is the calculated weight ratio of NaCl in the asprepared semi-IPN hydrogel based on added ingredients.

Salt ion weight in the semi-IPN hydrogel. For the as-prepared semI-IPN hydrogel, the salt ion weight is 0.15078 times the weight of the hydrogel. For the hydrogel after an operation, the salt ion content is

 $m_{salt} = m_{dried} - 0.15262m_{gel}$

where m_{dried} is the dried weight of the semi-IPN hydrogel after the operation and m_{gel} is the original weight of the hydrogel. 0.15262 is the calculated weight ratio of polymers in the hydrogel.

Acid and base content in the PAam buffer gels. Based on how the HCl and NaOH solutions were made, the concentration of the HCl solution was 6.0984 X 10⁻⁵ mol/g (mol per gram of

solution), and the concentration of the NaOH solution was 6.3998×10^{-5} mol/g (mol per gram of solution). The calculation for the amount of H⁺ is

$$n(H^{+}) = \frac{6.3998 \times 10^{-5} m_{NaOH} \cdot (m_w + a_{HCl} \cdot m_{HClgel})}{m_{solution} - 36.46 \times 6.3998 \times 10^{-5} m_{NaOH}}$$

where m_{NaOH} (g) is the weight of the NaOH solution used, m_w (g) is the weight of DI water added to the vial, m_{HClgel} (g) is the weight of the PAam buffer gel after the forward operation, $m_{solution}$ (g) is the weight of the solution transferred to the flask, and a_{HCl} is the ratio of water in the PAam buffer gel after the forward operation, which was measured by parallel experiments in the same conditions. The a_{HCl} for 15 min is 0.81990, for 30 min is 0.80605, and for 60 min is 0.79698. The calculation for the amount of OH⁻ is

$$n(OH^{-}) = \frac{6.0984 \times 10^{-5} m_{HCl} \cdot (m_w + a_{NaOH} \cdot m_{NaOHgel})}{m_{solution} - 40.00 \times 6.0984 \times 10^{-5} m_{HCl}}$$

where m_{HCl} (g) is the weight of the HCl solution used, m_w (g) is the weight of DI water added to the vial, $m_{NaOHgel}$ (g) is the weight of the PAam buffer gel after the forward operation, $m_{solution}$ (g) is the weight of the solution transferred to the flask, and a_{NaOH} is the ratio of water in the PAam buffer gel after the forward operation, which was measured by parallel experiments in the same conditions. The a_{NaOH} for 15 min is 0.81255, for 30 min is 0.79132, and for 60 min is 0.79039.

Acid and base content in the IPN hydrogels. Based on how the HCl and NaOH solutions were made, the concentration of the HCl solution was 5.3947×10^{-5} mol/g (mol per gram of solution), and the concentration of the NaOH solution was 4.8169×10^{-5} mol/g (mol per gram of solution). The calculation for the amount of H⁺ is

$$n(H^+) = \frac{4.8169 \times 10^{-5} \, m_{NaOH} \cdot (m_w + a_{HCl} \cdot m_{HClgel})}{m_{solution} - 36.46 \times 4.8169 \times 10^{-5} \, m_{NaOH}}$$

where m_{NaOH} (g) is the weight of the NaOH solution used, m_w (g) is the weight of DI water added to the vial, m_{HClgel} (g) is the weight of the IPN hydrogel, $m_{solution}$ (g) is the weight of the solution transferred to the flask, and a_{HCl} is the ratio of water in the IPN after the "new-buffer" path reverse operation, which was 0.75192. The calculation for the amount of OH⁻ is

$$n(OH^{-}) = \frac{5.3947 \times 10^{-5} m_{HCl} \cdot (m_w + a_{NaOH} \cdot m_{NaOHgel})}{m_{solution} - 40.00 \times 5.3947 \times 10^{-5} m_{HCl}}$$

where m_{HCI} (g) is the weight of the HCI solution used, m_w (g) is the weight of DI water added to the vial, $m_{NaOHgel}$ (g) is the weight of the IPN hydrogel, $m_{solution}$ (g) is the weight of the solution transferred to the flask, and a_{NaOH} is the ratio of water in the IPN after the "same-buffer" path reverse operation, which was 0.69205. The final pH in the IPN is estimated based on the acid and base content and the dimension of the IPN which is estimated to be 12 mm x 12 mm x 3 mm.

Calculation of change in the modulus of the hydrogel when compression tests were performed with membranes. For a stacking structure, the modulus of the overall structure is

$$E' = \frac{L_1 + L_2 + L_3}{\frac{L_1}{E_1} + \frac{L_2}{E_2} + \frac{L_3}{E_3}}$$

For Nafion 117, the thickness is 0.183 mm (L₁) and the modulus is 0.25 GPa (E₁). For FAA-3-PK-75, the thickness is 0.075 mm (L₃) and the modulus is 2 GPa (E₃). L₂ is the thickness of the hydrogel and E₂ is the modulus of the hydrogel. We can calculate E_2

$$E_2 = \frac{E'E_1E_3L_2}{E_1E_3(L_1 + L_2 + L_3) - E'(L_1E_3 + L_3E_1)}$$

It is important to note that this calculation is most relevant for very small strains. The ion exchange membranes impose a boundary constraint of effectively no in-plane deformation on the two faces of the IPN. As axial deformation increases, this boundary condition (changing the stress mode away from uniaxial) strongly influences an increasing fraction of the gel. The full stress-strain curve therefore shows an additional stiffening effect at large strains not present for an IPN under pure slip boundary conditions or under uniaxial tensile tests (even if accounting for area changes).

Fitting stress-strain curves with the Neo-Hookean model. To calculate the modulus of hydrogels, we employed the Neo-Hookean model to fit the stress-strain curves.

$$\sigma = 2(\lambda^2 - \lambda^{-1})C_{10}$$
$$\lambda = 1 + \varepsilon_{engineering}$$
$$\sigma = \lambda \sigma_{engineering}$$

Engineering stress and strain are negative for compression tests and they are positive for tensile tests. The modulus is

$$E = 6C_{10}$$

We selected the stress-strain curve in the 15~25% engineering strain range for fitting. The moduli from the compression tests already account for the thickness according to the equation above.

S3. Simulation.

We implement the model using our lumped element method derived in Tepermeister et al. ^[2]. In our simulation we use four mobile species: H⁺, OH⁻, Na⁺, and Cl⁻. Water is not modeled explicitly, and is instead assumed present everywhere. We use four different materials in our system, which correspond to the materials used experimentally: IPN, PAam, FAA-3-PK-75, and Nafion 117. The diffusivity that we use for each species is given in Table S3 and was chosen to fit the experimental data. We assume ideal solution mixing for all the materials.

We construct the model out of *uniform material prism*, *sharp material interface*, and *Tafel electrode* elements. The structure and connections between these elements are shown in Figure S5. We model two chemical reactions at each of the *Tafel electrode*s, the hydrogen evolution reaction and the oxygen evolution reaction. The parameters that we use for these reactions are given in Figure S6, and were matched to fit our experimental data and based on

literature ^[3,4]. The reaction rate at the electrodes is limited by a maximum reaction rate, which accounts for kinetic limitations at very large potentials not modeled by the simple Tafel equations used. For all our simulations, the electrodes saturate this limit, which was chosen to match our experimental data. The parameters for the rest of the elements are given in Table S1. Tables S2 and S3 give the simulation initial conditions and material properties respectively.

In order to model the chemical equilibrium between H⁺ and OH⁻ that occurs everywhere in this system, we use a modified volume element, which we call a *reaction volume*. The governing equation for this volume is

$$V\frac{dC_i}{dt} = j_i + \sum_j v_i^j K_j \prod_k C_k x_k^j$$

where *V* is the volume of the element, C_i is the concentration of species *i*, j_i is the flux of that species through the element's connector, v_i^j is the stoichiometry of species *i* in chemical reaction *j*, K_j is the rate coefficient for reaction *j*, and x_k^j is the rate exponent for species *k* in reaction *j*. We model two reactions using this volume: the autoionization of the implicit water into H⁺ and OH⁻ and the reaction of those two species back into implicit water. These *reaction volume* elements are used everywhere in our simulation in place of *volume* elements. For our system, K = [1e-1, 1e-9], x = [[1, 1, 0, 0], [0, 0, 0, 0]], and v = [[-1, -1, 0, 0], [1, 1, 0, 0]]. For *K* the first index is the chemical reaction, while *x* and *v* are first indexed by chemical reaction and then species, in the order [H⁺, OH⁻, Na⁺, Cl⁻].

We use a current source as the electrical input to the model. The input current over time is given in Table S4.



Figure S5 Diagram showing how elements are connected in the simulated system.

			Parameter	s	
т	20		°C V	,	
area	10e-3*10e-3		m2	,	Surface area for ions
C_reference[4]	{1e-4, 1e-4, 1, 1}		mol/m3	7	Reference molar concentration for exchange current
alpha[2]	{0.1, 0.5}		1	7	Charge transfer coefficient
nR	2				Number of chemical reactions
n[2]	{2, -2}	7			Number of electrons transferred to electrode in each reaction (positive is oxidation)
stoichiometry[2, 4]	{{2, 0, 0, 0}, {0, 2, 0, 0}}	-			Stoichiometry of each species in each reaction
rate_exponents[2, 4]	{{0, 0, 0, 0}, {0, 0, 0, 0}}				Exponent of each species in reaction's rate limiting step
V_reduction[2]	{1.23, 0}		v •	,	Standard reduction potential vs SHE for each reaction
J_0[2]	{1e-3, 1e-3}	7	A/m2	,	Exchange current density
reaction_rate_reference[2]	J_0./(Constants.F.*abs(n))		mol/(m2.s)	7	Reference reaction rate
use_reaction_rate_maximum	 Image: A start of the start of				clamp reaction rates to the maximum
reaction_rate_maximum[2]	electrode_a.reaction_rate_reference*7e1	7	mol/s	7	Maximum rate possible for each reaction
reaction_post_maximum_slope	1e-7		mol/(V.s)	,	Slope of post maximum current vs voltage

Figure S6: Parameters for Electrode A and Electrode B Tafel electrodes

Table S1 Element Geometry

Element	Area (m ²)	Length (m)	Segments	Sharpness
PAam A	288e-6	3e-3	3	10
FAA-3-PK-75	144e-6	78e-6	3	10
IPN	144e-6	3.1e-3	3	10
Nafion 117	144e-6	210e-6	3	10
PAam B	288e-6	3e-3	3	10

 Table S2 Initial Conditions:

		P/	Aam		IPN				
Simulation	H⁺	OH⁻	Na⁺	CI⁻	H⁺	OH⁻	Na⁺	CI⁻	
forward	1e-4	1e-4	1	1	1e-4	1e-4	3100	3100	
reverse with same buffer reverse with new buffer	1e-4	1e-4	3500	3500	101.04	1.757e-9	1152.34	1253.39	

Table S3 Material Properties

		Diffusivi	Fixed charge (mol/m ³)		
Simulation	H⁺	OH⁻	Na⁺	Cl⁻	
PAam A	2.793e-9	1.581e-9	3.99e-10	6.09e-10	0
IPN	2.793e-9	1.581e-9	3.99e-10	6.09e-10	0
FAA-3-PK-75	4.3e-11	7e-11	8.8e-12	1.150e-11	1.6e3
Nafion 117	1.250e-9	4.0e-10	1.150e-10	2.0e-10	-1.3e3

Table S4. Current Input Profiles

Currents are linearly interpolated between values in the table.

Simulation	Forward 60 min				Forward 30 min Reverse 30 min with same buffer				Re	verse ne	e 30 mi w buffe	n with r	
Time (s)	0	1	60	4000	0	60	1860	1861	3720	0	1	60	1860
Current (mA)	0	0	100	100	0	100	100	-100	-100	0	0	-100	-100

S4. Additional results



Figure S7 Monotonic tensile test results of PAam hydrogel and salt PAam hydrogel vs. high NaCl content IPN and high NaCl content IPN*.

Material	Modulus (kPa)
PAam	13.2 ± 0.3
salt-PAam	14.6 ± 1.0
IPN-high-NaCl	8.0 ± 0.7
IPN-medium-NaCl	11.7 ± 1.6
IPN-low-NaCl	35.8 ± 3.7
IPN*-high-NaCl	6.6 ± 0.5
IPN*-medium-NaCl	5.1 ± 0.9
IPN*-low-NaCl	11.8 ± 1.2

 Table S5 Modulus of hydrogels from the tensile tests.

 Table S6 Modulus of hydrogels from compression tests.

Material	Modulus (kPa)
PAam	32.7 ± 3.1
IPN	19.7 ± 3.1
IPN-less-water	21.2 ± 1.6
IPN-more-water	8.6 ± 2.1
IPN-15 min	30.9 ± 5.2
IPN-30 min	60.8 ± 11.0
IPN-reverse	8.4 ± 2.2
IPN-same-buffer	12.2 ± 1.8
IPN-new-buffer	9.0 ± 2.5
IPN-60 min	96.9 ± 21.1



Figure S8 Compression test results of initial IPN, initial IPN with 26% of water content removed by evaporation, and initial IPN with 33% of water content added by blowing moisture to gels.



Figure S9 Compression test results of initial IPN, IPN after 30 min forward operation, and PAam buffer hydrogel.



Figure S10 Amount of water remaining in the IPN after a 30 min reinjection operation, shown as fractions of the initial composition.



Figure S11 Average pH change of PAam buffer gels during the "same-buffer" reinjection operation.



Figure S12 Simulated flux of OH⁻ and Cl⁻ (left) into and (right) out of the IPN.



Figure S13 Simulated flux of H⁺ and Na⁺ (left) into and (right) out of the IPN.



Figure S14 Picture of an IPN gel after a reverse operation.



Figure S15 A screenshot of the 3D printed part. The dimension of one window is 7 mm X 17 mm.



Figure S16 Cyclic indentation test results of IPN hydrogel before and after a forward voltage was applied. The legend indicates first versus second cycle.



Figure S17 Cyclic indentation test result of IPN hydrogel after a reverse voltage was applied.

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