

Supplementary Information of

Energy-Efficient Modular Water Purification System via Concurrent Freshwater
and Hydrogen Generation Using Ion Concentration Polarization

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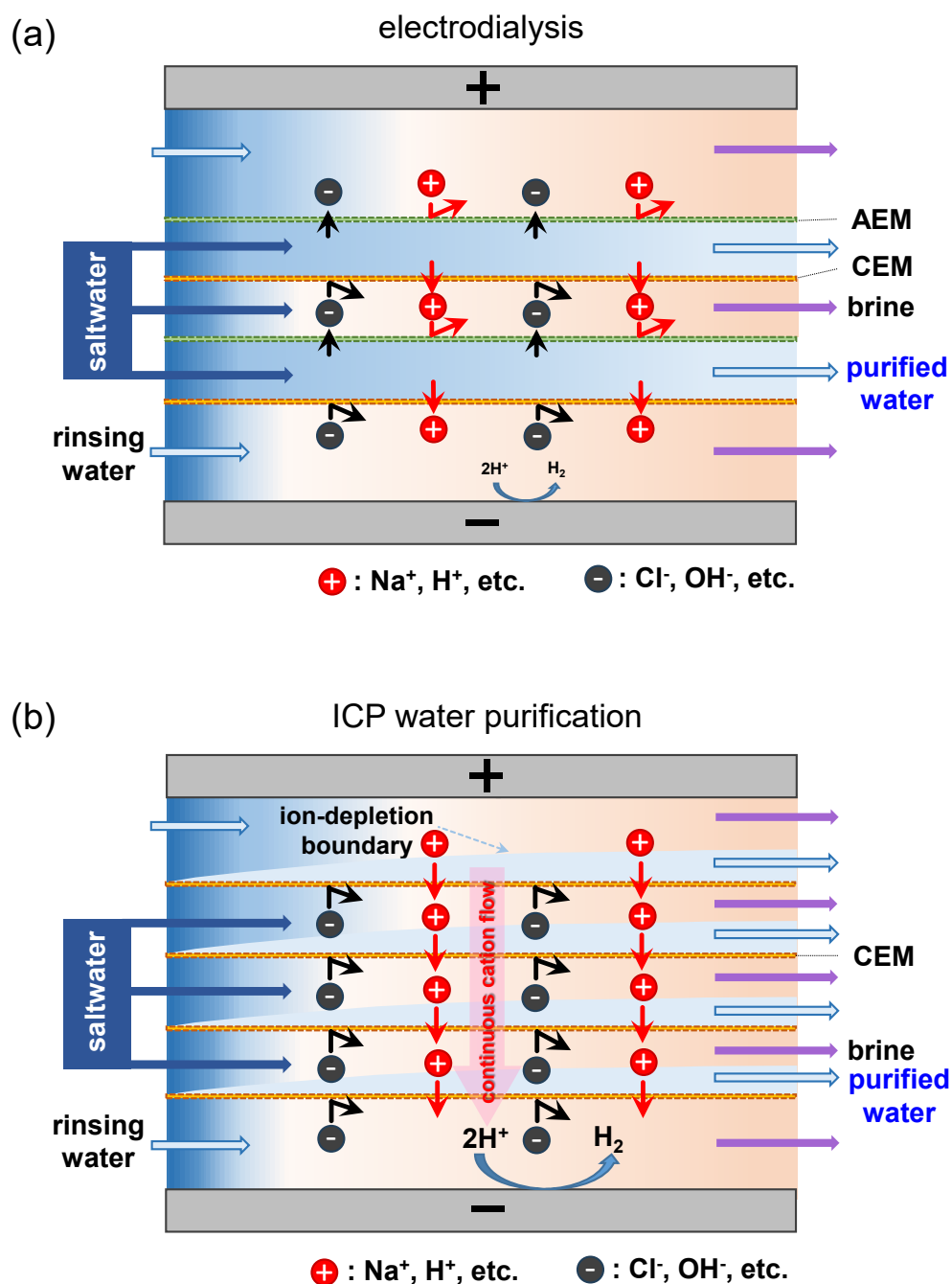
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1. Comparison Between Electrodialysis (ED) and Ion Concentration Polarization (ICP) Water Purification

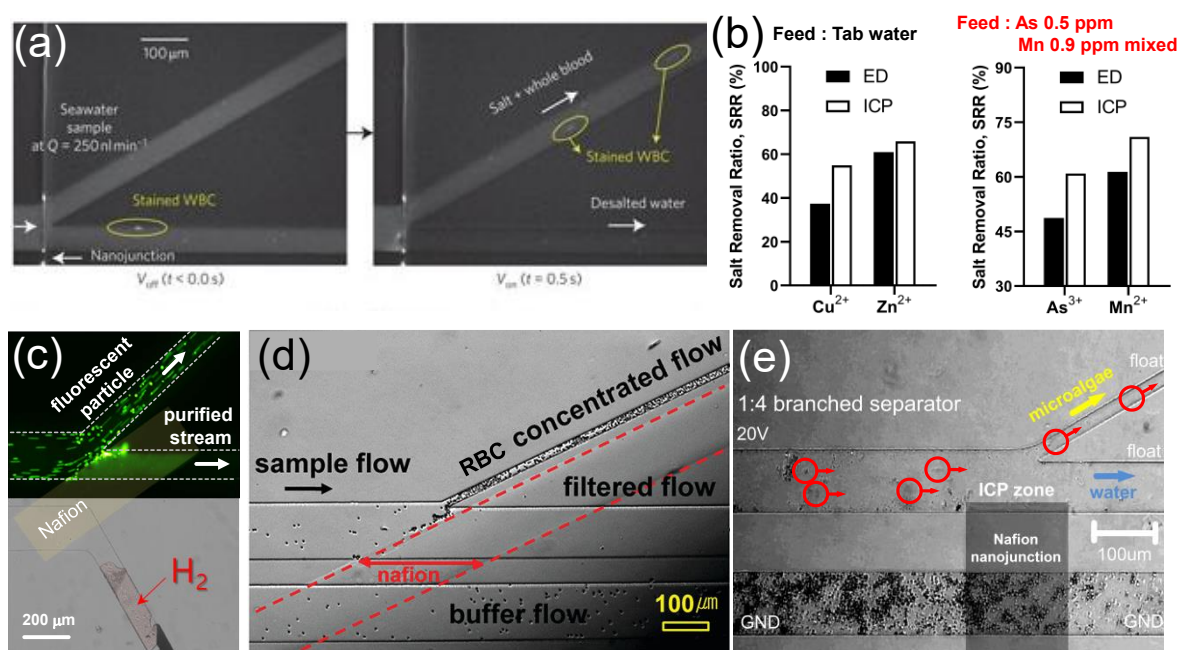


Supplementary Figure 1. (a) ED uses alternating cation-exchange membranes (CEMs) and anion-exchange membranes (AEMs) to remove ions via electric field. (b) ICP water purification utilizes only CEMs to create ion depletion zones for water purification under an electric field, allowing continuous cation flow.

Supplementary Table 1. Comparison of ICP-based and ED-based water purification systems.

Category	ICP-Based Water Purification	ED-Based Water Purification
Membrane Structure	Utilizes only a single cation exchange membrane (CEM)	Requires a combination of cation (CEM) and anion (AEM) exchange membranes
Target Contaminants	Simultaneously removes a wide range of charged pollutants including salts (ions), large ionic species (e.g., heavy metals), suspended solids, biofoulants, and oils. See Supplementary Figure 2.	Primarily removes dissolved salts (ions)
Operating Principle	Relies on a repulsive electrical force generated near the nanoporous membrane, which actively repels contaminants	Ions pass through ion-exchange membranes driven by an applied electric field; thus, only ionic species are removed
Device Size	Highly miniaturizable and modular; suitable for portable and field-deployable systems	Also miniaturizable, but typically not integrated with multi-functional capabilities
Membrane Fouling	Reduced fouling due to physical repulsion of suspended particles from the membrane interface	Severe fouling issues (e.g., scaling, biofouling) due to direct membrane filtration
Energy Consumption	RO << ICP < ED (<i>Note: While RO is more energy-efficient, it is not easily miniaturizable</i>) ^{1, 2}	Higher energy consumption compared to both RO and ICP
Hydrogen Production	Continuous H ⁺ generation allows for integration with hydrogen production systems	Cannot continuously supply protons for hydrogen generation

In summary, the ICP platform not only offers a compact and energy-efficient alternative to ED, but also enables multi-modal contaminant removal and integration with energy recovery processes. This positions ICP technology as a uniquely advantageous solution for next-generation decentralized or portable water purification systems.



Supplementary Figure 2. Versatility of ICP purification. (a) Selective removal of salts and particles: Fluorescent dyes (representing salts) and white blood cells (WBCs, representing micrometer-sized particles) passed only through the salted stream during ICP-based seawater purification³. (b) Heavy-metal removal: ICP yields higher salt removal ratios (SRR) than ED for Cu^{2+} , Zn^{2+} , As³⁺ and Mn²⁺ feeds. (c) Particle clearance: charged microspheres are swept into the anodic brine branch while H_2 bubbles evolve in the cathodic branch. (d) Bio-contaminant removal: red blood cells and *E. coli* exit exclusively through the brine channel⁴. (e) Oil separation: microalgae oils (red circle) partition into the brine stream, leaving the purified branch oil-free.

2. Energy Efficiency Analysis of Simultaneous ICP Purification and Hydrogen Production

This study investigates how the energy efficiency of ICP purification can be improved by recycling hydrogen gas generated during the concurrent process. In the mesoscale device (20 mM and 200 mM NaCl feeds at various applied currents; Fig. 3), the input energy required to produce 1 L of freshwater was obtained by integrating the product of the applied current and full cell voltage over the purification time. Faradaic efficiency was determined from the ratio of the charge contained in the evolved H₂ ($2F$ per mol, where F is the Faraday constant) to the total charge passed, confirming that essentially every electron contributes to hydrogen evolution. The recoverable output energy was then estimated assuming a full cell efficiency of 60 %. Detailed numerical values of each case are listed in Supplementary Table 2 and 3. It is worth noting that practical considerations such as hydrogen collection, storage, compression, and fuel cell system costs were not included in this preliminary analysis and may warrant further investigation depending on the intended application.

Supplementary Table 2. Energy analysis for producing 1 liter of freshwater from 20 mM NaCl saltwater

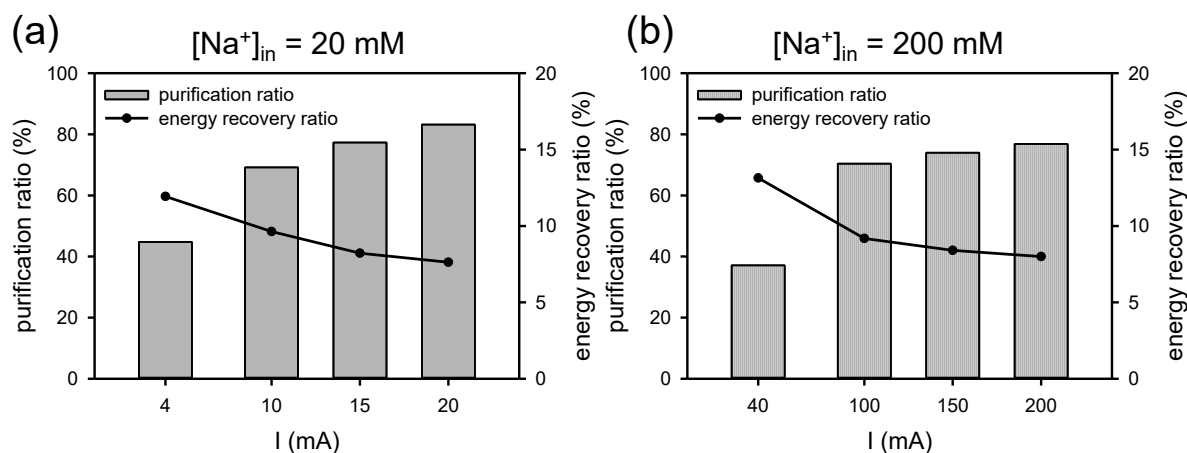
Applied Current (mA)	4	10	15	20
Applied Voltage (V)	6.20	8.74	10.75	11.68
Input Energy (Wh)	4.05	14.28	26.36	38.16
Purification Ratio (%)	44.8	69.2	77.3	83.2
H ₂ Production (g)	0.024	0.069	0.108	0.145
Faradaic Efficiency	~98	~100	~100	~100
Output Energy from H ₂ (Wh)	0.48	1.38	2.17	2.91
Energy Recovery Ratio (%)	11.94	9.63	8.22	7.63

Supplementary Table 3. Energy analysis for producing 1 liter of freshwater from 200 mM NaCl saltwater

Applied Current (mA)	40	100	150	200
Applied Voltage (V)	7.13	10.07	11.61	12.17
Input Energy (Wh)	46.64	169.22	284.59	397.71
Purification Ratio (%)	37.1	70.41	73.99	76.85
H ₂ Production (g)	0.307	0.777	1.196	1.590
Faradaic Efficiency	~100	~100	~100	~100
Output Energy from H ₂ (Wh)	6.13	15.54	23.91	31.80
Energy Recovery Ratio (%)	13.14	9.18	8.40	8.00

For the 20 mM NaCl feed, the purified ratio improved significantly with increasing current, from 44.8% at 4 mA to 83.2% at 20 mA, indicating effective ion removal. However, as shown in Supplementary Figure 3(a), the energy recovery rate decreased from 11.94% to 7.63%, reflecting that the energy input significantly increases as purification performance is enhanced. For the 200 mM NaCl feed, a similar trend was observed. The purified ratio increased from 37.1% at 40 mA to 76.9% at 200 mA. However, the energy recovery rate, shown in Supplementary Figure 3(b), decreased from 13.1% at 40 mA to 8.00% at 200 mA. The purified ratio and energy recovery efficiency trends of Supplementary Figure 3 emphasize the importance of balancing applied current to optimize overall system performance. By carefully selecting operating conditions, it is possible to achieve effective purification while considerable energy recovery from hydrogen production, positioning this approach as a viable alternative to

conventional purification technologies.



Supplementary Figure 3. (a) Purification ratio (left y -axis) and energy recovery ratio (right y -axis) as a function of applied current for 20 mM NaCl feed saltwater. Higher applied currents improve the purified ratio but reduce the energy recovery rate. (b) Same analysis for 200 mM NaCl feed saltwater.

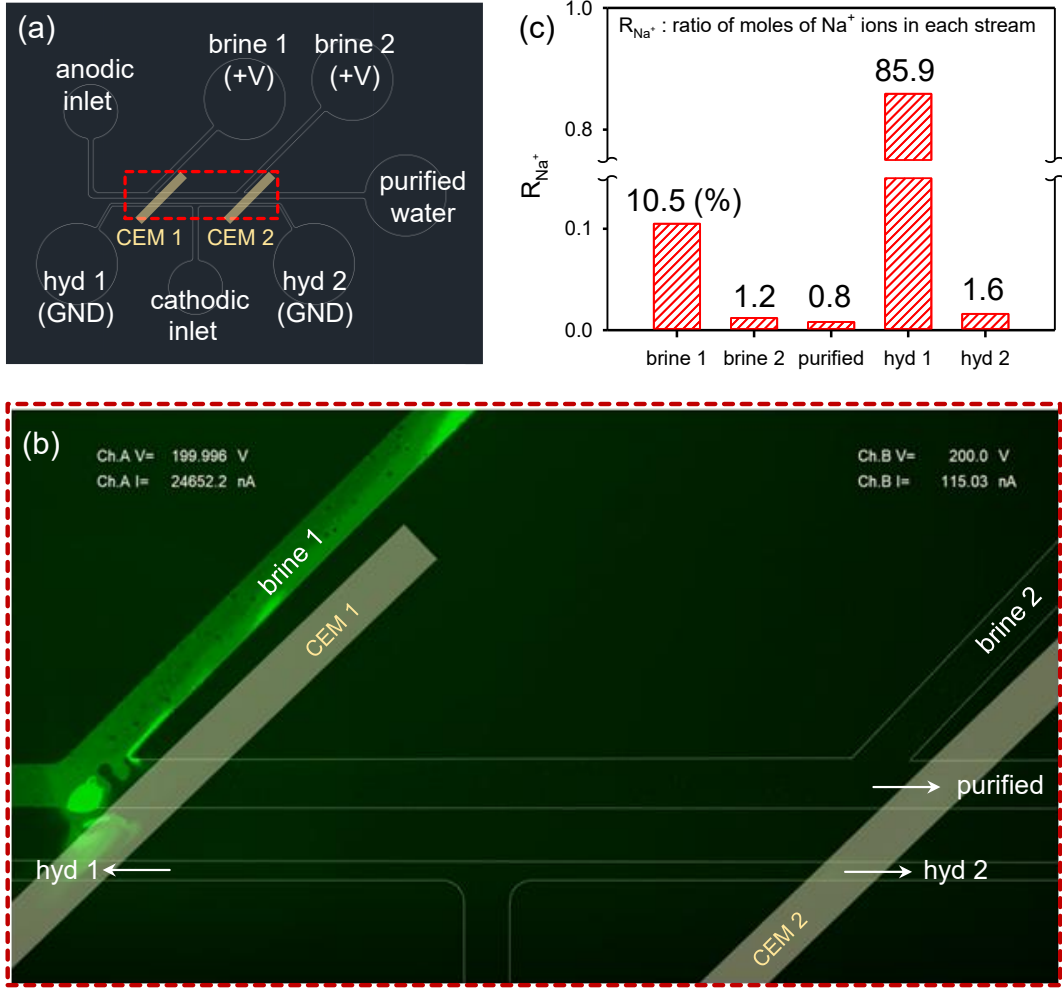
While a full life-cycle assessment of the ICP system itself lies outside the present scope, our group has already completed system level techno-economic and life-cycle assessment studies for three representative decentralized, small scale energy–water platforms that could incorporate the proposed ICP module in future deployments: (i) highway solar EV-charging stations that self-supply panel cleaning water and supplemental electricity; (ii) off-grid data centers constrained by both cooling-water availability and power budget; and (iii) solar-powered rail systems required to recycle grey water onboard^{5, 6, 7}. In each scenario we figured out the value of routing the cogenerated hydrogen through a fuel cell and confirmed clear economic and ecological advantages over conventional technologies. These findings suggest that integrating the ICP purifier could further lower the effective cost of water and energy in portable micro-utility settings, reinforcing its practical relevance beyond laboratory performance.

3. Two-stage serial ICP microfluidic device experiments and numerical simulations

To evaluate whether a serial configuration could enhance desalination performance beyond that of a single-stage device, we carried out both experimental and numerical investigations using a two-stage serially connected ICP microfluidic system. This approach allowed us not only to examine the practical effectiveness of sequential ion removal, but also to explore the feasibility and possible energy implications of multi-stage operation.

3.1. Experimental Demonstration

A two-stage serial ICP microfluidic device (Supplementary Figure 4(a)) was fabricated to evaluate whether sequential operation improves purification beyond a single unit. The feed solution enters through the anodic inlet; most cations migrate across the CEM1 into the hyd1 or leave with the brine1. The partially purified stream then encounters the second bifurcation, where the residual cations are removed through CEM2 or brine2 before the effluent reaches the purified water outlet. Fluorescence image (Supplementary Figure 4b) visualizes IDZ in front of CEM1 and shows that the fluorescent dye (representing cation) is almost entirely diverted into brine1, confirming effective first-stage removal. IC quantification (Supplementary Figure 4c) reveals that 85.9 % of the Na^+ exits via hyd1 (through CEM1) and 10.5 % via brine1; less than 4 % of the original Na^+ reaches the second stage, and the Na^+ content in the final purified water stream is under 1 % of the feed.



Supplementary Figure 4. (a) Schematic of the two-stage serial ICP purification microfluidic device with two bifurcation units. (b) Fluorescent image showing IDZ near CEM1. (c) Molar distribution of Na⁺ across all outlets.

3.2. Numerical Simulation

To investigate the ion transport in multi-branched ICP device, the transport phenomena were formulated based on charge conservation, mass conservation and momentum conservation. The charge conservation was described by the Poisson equation.

$$-\tilde{\nabla}^2 \tilde{\phi} = \frac{1}{2\tilde{\lambda}_D^2} (\tilde{c}_+ - \tilde{c}_-) \quad (1)$$

where the variables with tilde ($\sim$) means the dimensionless variables of which characteristic

scales were summarized in Supplementary Table 4, $\tilde{\phi}$ is the electrical potential, $\tilde{\lambda}_D$ is the Debye length, $\tilde{c}_+$ is the cation concentration, and $\tilde{c}_-$ is the anion concentration. In this work, 1:1 electrolyte such as KCl, NaCl, *etc* was considered. The mass conservations of ionic species were described by the Nernst-Planck equations for steady-state.

$$0 = -\tilde{\nabla} \cdot (-\tilde{D}_\pm \tilde{\nabla} \tilde{c}_\pm \mp \tilde{D}_\pm \tilde{c}_\pm \tilde{\nabla} \tilde{\phi} + \tilde{c}_\pm \tilde{\mathbf{v}}) \quad (2)$$

where $\tilde{D}_\pm$ is the diffusivity of cation or anion and $\tilde{\mathbf{v}}$ is the flow field. The mass conservation and the momentum conservation of fluid were described by the continuity equation and the Stokes equations with external electrical force term.

$$\tilde{\nabla} \cdot \tilde{\mathbf{v}} = 0 \quad (3)$$

$$0 = -\tilde{\nabla} \tilde{P} + \tilde{\nabla}^2 \tilde{\mathbf{v}} - \frac{\kappa}{2\tilde{\lambda}_D^2} (\tilde{c}_+ - \tilde{c}_-) \tilde{\nabla} \tilde{\phi} \quad (4)$$

where $\tilde{P}$ is the pressure and κ is the hydrodynamic coupling constant.

The numerical boundaries were depicted in Supplementary Figure 5(a). At inlet, ionic species were injected by convection only so that following boundary conditions were imposed.

$$\tilde{\mathbf{n}} \cdot \tilde{\nabla} \tilde{\phi} = 0, \tilde{c}_\pm = 1 \text{ and } \int_{inlet} \tilde{\mathbf{n}} \cdot \tilde{\mathbf{v}} d\tilde{S} = Pe \quad (5)$$

where $\tilde{\mathbf{n}}$ is the inward unit normal vector, $d\tilde{S}$ is the surface element and Pe is the Peclet number. Since brine 1 and brine 2 were treated as large reservoirs, diffusive transport was neglected. In addition, constant potential and pressure was imposed.

$$\tilde{\phi} = 0, \tilde{\nabla} \tilde{c}_\pm = 0 \text{ and } \tilde{P} = 0 \quad (6)$$

The effects of applied potentials at each brine reservoir were modeled using algebraic equations at the hyd 1 and hyd 2 boundaries. The Nafion was treated as an ideal permselective material so

that boundary conditions at hyd 1 were

$$\tilde{\phi} = -\ln \tilde{N} + \tilde{V}_1, \tilde{c}_+ = \tilde{N}, \mathbf{n} \cdot (-\tilde{D}_- \tilde{\nabla} \tilde{c}_- + \tilde{D}_- \tilde{c}_- \tilde{\nabla} \tilde{\phi}) = 0 \text{ and } \tilde{\mathbf{v}} = 0 \quad (7)$$

where $\tilde{N}$ is the Donnan concentration of the ideal permselective material and $\tilde{V}_1$ is the electrical potential at the interface. $\tilde{V}_1$ can be determined by following algebraic equation.

$$\tilde{i}_1 = -\int_{hyd1} \mathbf{n} \cdot [(-\tilde{D}_+ \tilde{\nabla} \tilde{c}_+ - \tilde{D}_+ \tilde{c}_+ \tilde{\nabla} \tilde{\phi}) - (-\tilde{D}_- \tilde{\nabla} \tilde{c}_- + \tilde{D}_- \tilde{c}_- \tilde{\nabla} \tilde{\phi})] dS \quad (8)$$

where $\tilde{i}_1$ is the current density pass through the hyd 1 boundary. Similary, at hyd 2

$$\tilde{\phi} = -\ln \tilde{N} + \tilde{V}_2, \tilde{c}_+ = \tilde{N}, \mathbf{n} \cdot (-\tilde{D}_- \tilde{\nabla} \tilde{c}_- + \tilde{D}_- \tilde{c}_- \tilde{\nabla} \tilde{\phi}) = 0 \text{ and } \tilde{\mathbf{v}} = 0 \quad (9)$$

where $\tilde{V}_2$ can be determined by

$$\tilde{i}_2 = -\int_{hyd2} \mathbf{n} \cdot [(-\tilde{D}_+ \tilde{\nabla} \tilde{c}_+ - \tilde{D}_+ \tilde{c}_+ \tilde{\nabla} \tilde{\phi}) - (-\tilde{D}_- \tilde{\nabla} \tilde{c}_- + \tilde{D}_- \tilde{c}_- \tilde{\nabla} \tilde{\phi})] dS \quad (10)$$

$\tilde{i}_2$ is the current density pass through the hyd 2 boundary. The purified reservoir was mass sink so that

$$\mathbf{n} \cdot \tilde{\nabla} \tilde{\phi} = 0, \mathbf{n} \cdot \tilde{\nabla} \tilde{c}_\pm = 0 \text{ and } \tilde{P} = 0 \quad (11)$$

The remaining boundaries were impermeable walls.

$$\mathbf{n} \cdot \tilde{\nabla} \tilde{\phi} = 0, \mathbf{n} \cdot \tilde{\nabla} \tilde{c}_\pm = 0 \text{ and } \tilde{\mathbf{v}} = 0 \quad (12)$$

Above formulations were solved by COMSOL Multiphysics 6.2.

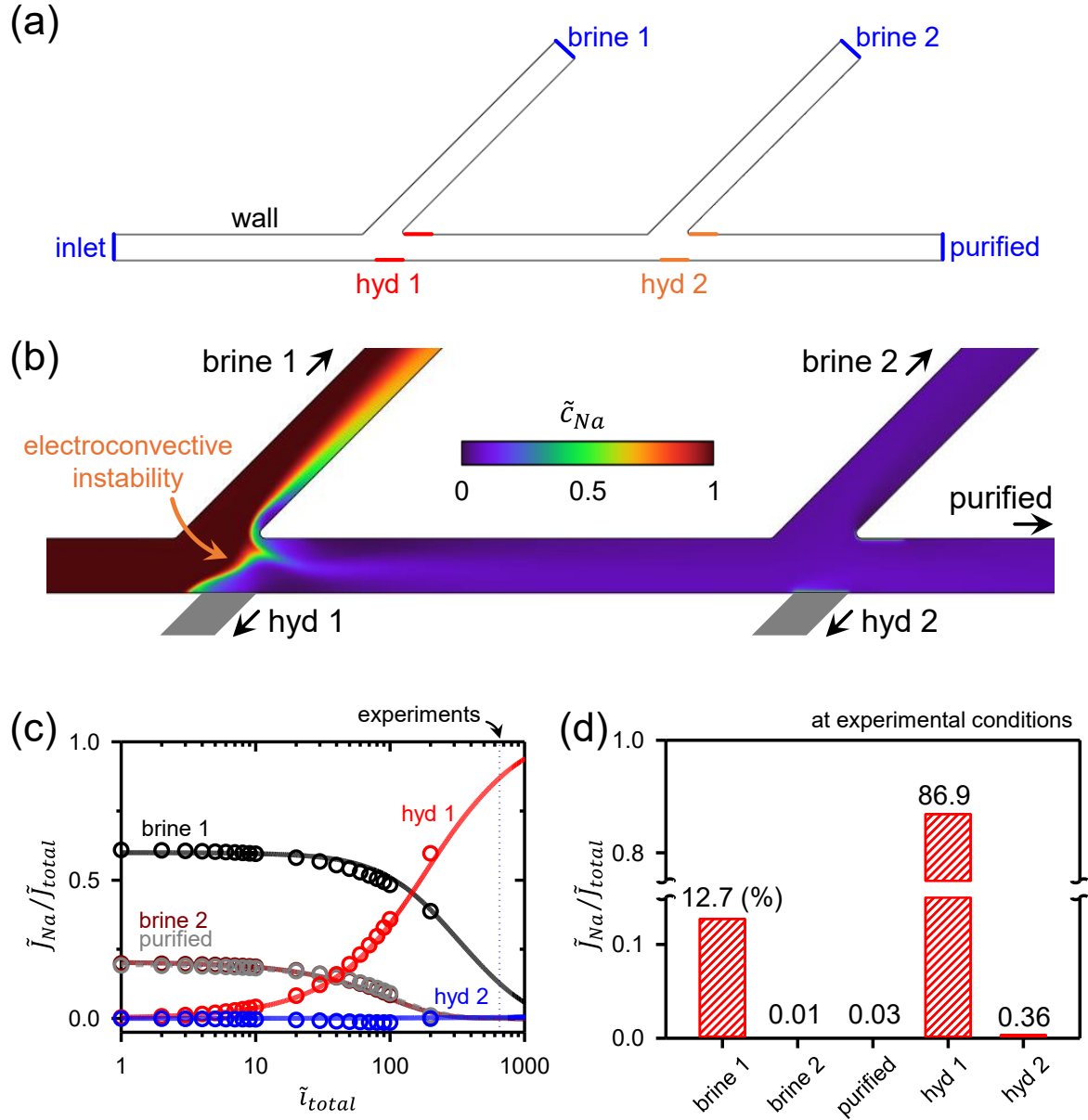
Supplementary Table 4. Scales for Non-dimensionalization

Quantity	Definition	Description
Length	W	Microchannel width
Velocity	$U_0 = \frac{D_+}{W}$	Velocity scaled by cation diffusivity
Pressure	$\frac{\mu D_+}{W^2}$	Pressure scaled by U_0
Concentration	c_0	Bulk concentration
Electrical potential	$\frac{RT}{F}$	Thermal voltage
Diffusivity	D_+	Cation diffusivity
Current density	$i_0 = \frac{FD_+c_0}{W}$	Diffusion-limited current density
λ_D	$\sqrt{\frac{\varepsilon RT}{2F^2 c_0}}$	Debye length
Pe	$\frac{U}{U_0}$	Peclet number
κ	$\frac{\varepsilon R^2 T^2}{2\mu F^2 D_+}$	Hydrodynamic coupling constant

In above table, μ is the viscosity of water, R is the gas constant, T is the absolute temperature, F is the Faraday constant, ε is the electrical permittivity of fluid and U is the cross-sectional averaged flow velocity.

For numerical convergence the model was run in constant current mode, fixing the stage-current split at $i_{hyd1} : i_{hyd2} = 250 : 1$ to match the experimental results. The dimensionless total current ($\tilde{i}_{total}$) was swept from 1 to 1000. As $\tilde{i}_{total}$ increased, a larger fraction of Na^+ crossed CEM1 into the hyd1 channel, so that stage 1 removed nearly all salt and only a highly diluted feed reached stage 2; beyond a threshold current the model also showed electroconvective

instability near CEM1 (see Supplementary Figure 5(b)). Under the experimental operating point ($\tilde{i}_{total} = 650$) the simulation predicted that 87 % of the incoming Na^+ in hyd1, 11 % in brine1, and less than 1 % reaches the purified-water outlet, in close agreement with the experimental measurements.



Supplementary Figure 5. (a) Domain boundary for numerical simulation of Na^+ transport in a two-stage serial ICP device with $i_{hyd1} : i_{hyd2} = 250 : 1$. (b) Concentration contour at $\tilde{i}_{total} = 200$.

(c) Fraction of Na^+ reaching each outlet as $\tilde{i}_{total}$ varies from 1 to 1000. (d) Molar distribution among brine1, brine2, purified, hyd1, and hyd2 channels at the experimental condition $\tilde{i}_{total} = 650$.

Both the experimental observations and computational simulations revealed that the majority of cations were already removed in the first stage, resulting in a substantially diluted feed concentration entering the second stage. Under such low-concentration conditions, the solution resistance increased significantly, leading to additional voltage drop across the device without appreciable gains in salt removal. These findings indicate that the serial configuration offers limited benefit relative to the additional energy input. Therefore, for improved purification performance, it is more effective to apply higher current to a single ICP module, as experimentally and numerically demonstrated in this study.

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