An Electrode Potential Regulation Strategy for Supercapacitors

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Electrolyte Preparation Procedure:

$NaClO_4$ (Aladdin) was dissolved in ultrapure water at molar ratios of 1:10, 1:7, and 1:3 to prepare electrolytes 1-10, 1-7, and 1-3, respectively. Urea (Aladdin) solutions with concentrations of 0.1 M, 1 M, 3 M, and 5 M were then added to the 1:7 electrolyte to produce mixed solutions of urea, water, and $NaClO_4$, designated as 1:7:0.1, 1:7:1, 1:7:3, and 1:7:5（1-7-0.1,1-7-1, 1-7-3, 1-7-5）. Electrolytes rich in active substances were prepared by adding 20 mM disodium anthraquinone sulfonate (Aladdin) and 40 mM, 60 mM, and 80 mM KBr (Aladdin), respectively, to the 1-7-3 electrolyte.

Preparation Electrode:

Commercial carbon nanotubes (xfnano) were selected as the electrodes, with 8-mm-square carbon nanotube sheets used for both the anode and cathode.

Material Characterization:

Nuclear magnetic resonance (NMR) analysis was performed using a Bruker AVANCE AV 400 MHz spectrometer to investigate hydrogen bonding variations among different ions in the electrolytes. Raman spectra were measured with a Horiba XploRA PLUS spectrometer equipped with a 785 nm laser line. Fourier transform infrared (FTIR) spectra were collected on a Thermo Fisher Scientific Nicolet iS50 spectrometer.

Electrochemical Characterization:

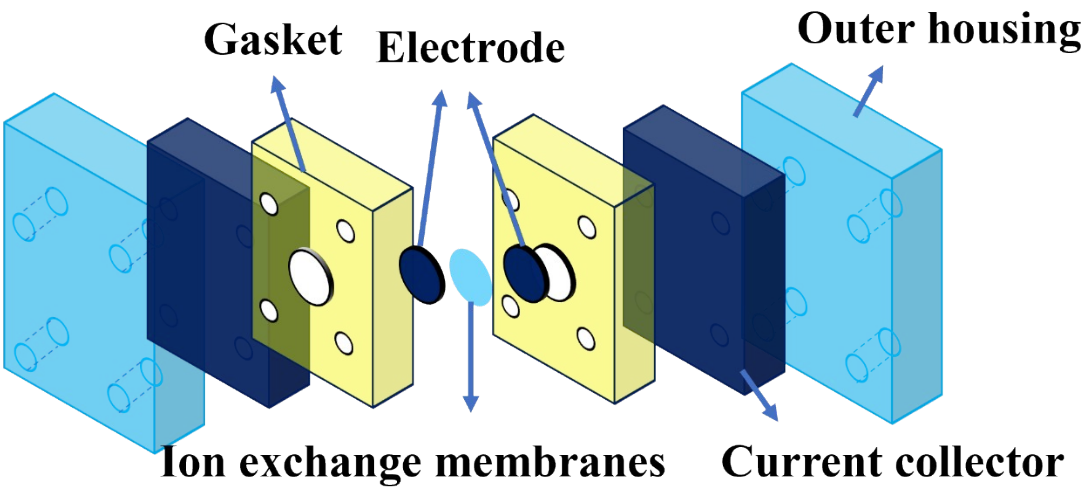
Cycling characterization of the cells was performed on a LAND battery test system (CT2001A).Cyclic voltammetry, and electrochemical impedance spectroscopy were measured using an electrochemical workstation (Autolab). Changes in electrode potential during the battery charging and discharging process were measured using an Autolab electrochemical workstation.

The formula for calculating the specific capacitance using GCD is as follows:

$$C_A = \frac{2I\int V\,dt}{A\Delta V^2}$$

Device Assembly:

To assemble a quasi-solid-state supercapacitor, carbon nanotube paper is cut into circular discs with a diameter of 8 mm and assembled into a symmetrical supercapacitor as shown in the figure:



Decomposition diagram of a supercapacitor cell used to study the electrochemical properties of electrolytes

Computational Details: Molecular dynamics simulations were performed using the Forcite module in Materials Studio, with the COMPASS II force field applied. All simulations were conducted under the NVT ensemble at 298 K. The binding energies between Na^+ and other ions, as well as the LUMO and HOMO energy levels of different molecules, were calculated using the Dmol³ module.

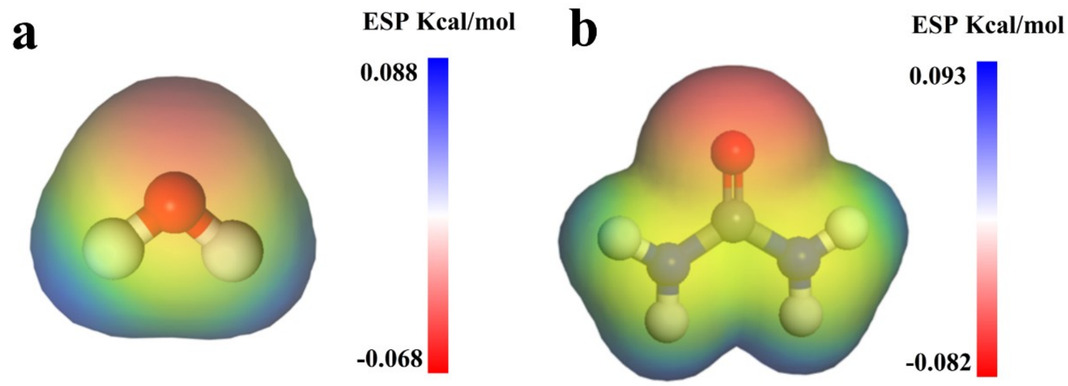


Figure S1. Electrostatic diagram of urea and water molecules.

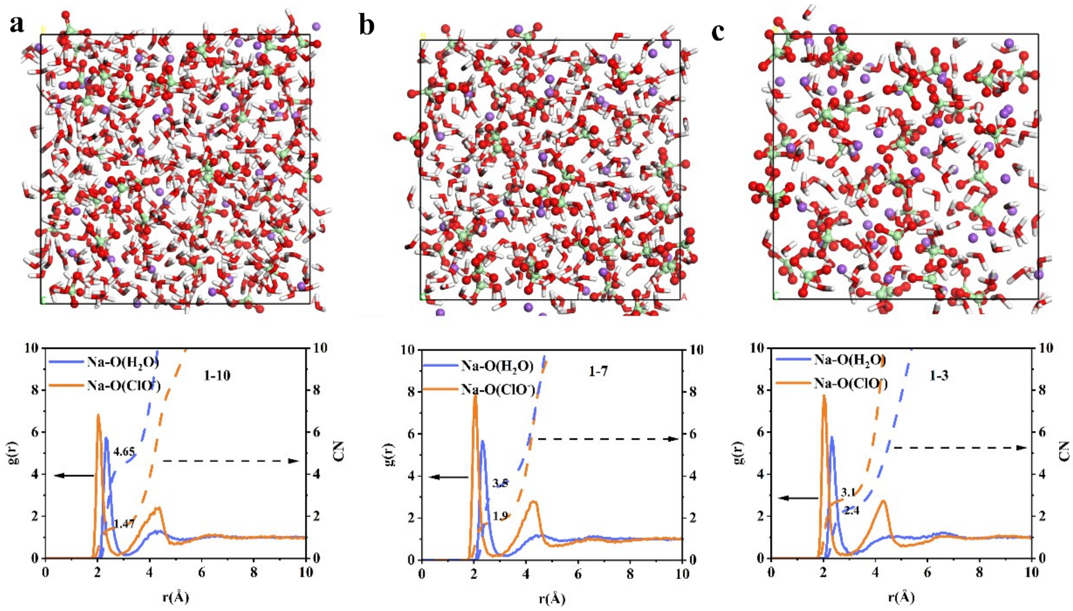


Figure S2. Radial distribution functions and coordination numbers for different ratios of $NaClO_4$ and water.

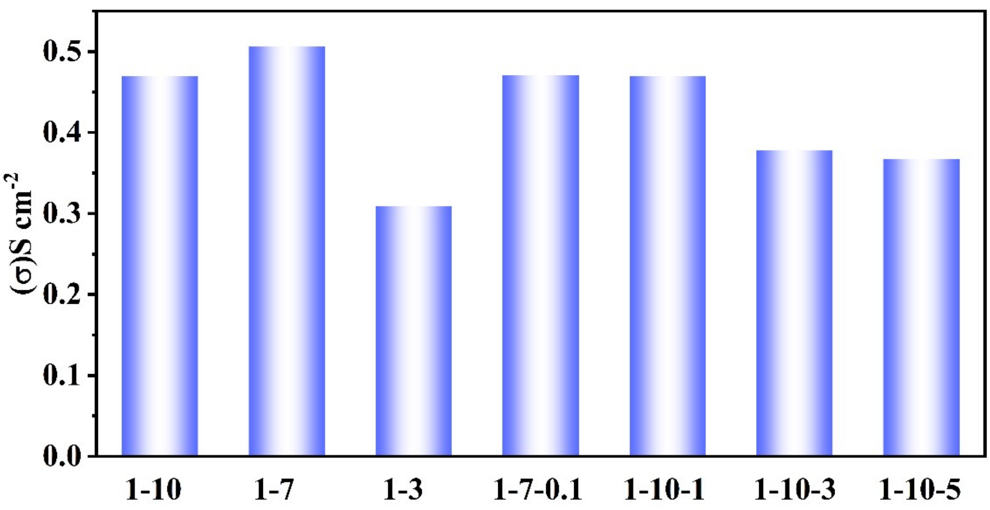


Figure S3. Graphs showing the conductivity of solutions of water, $NaClO_4$, and water at different concentrations.

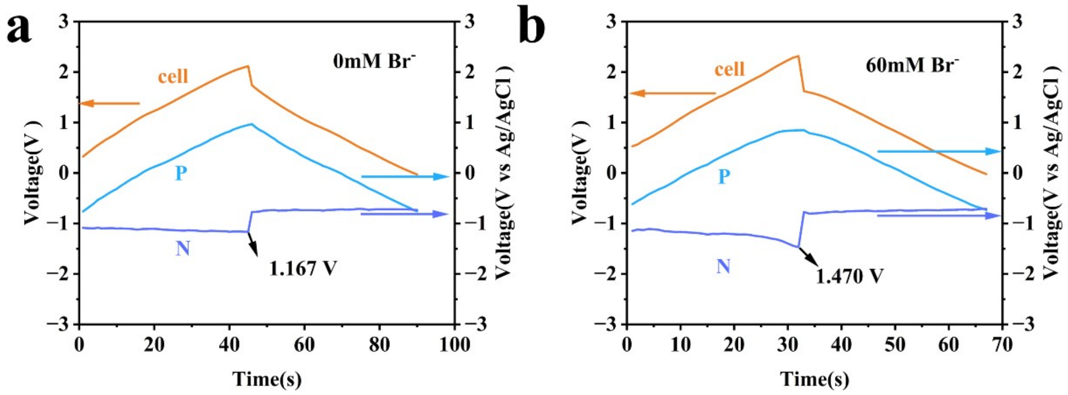


Figure S4. Zn hybrid supercapacitors assembled by introducing 20 mM AQ at the anode and different concentrations of Br^- at the cathode in 1-7-3 electrolyte. (The anode material is a Zn foil.) (a) 0 mM Br^-, (b) 60 mM Br^-.

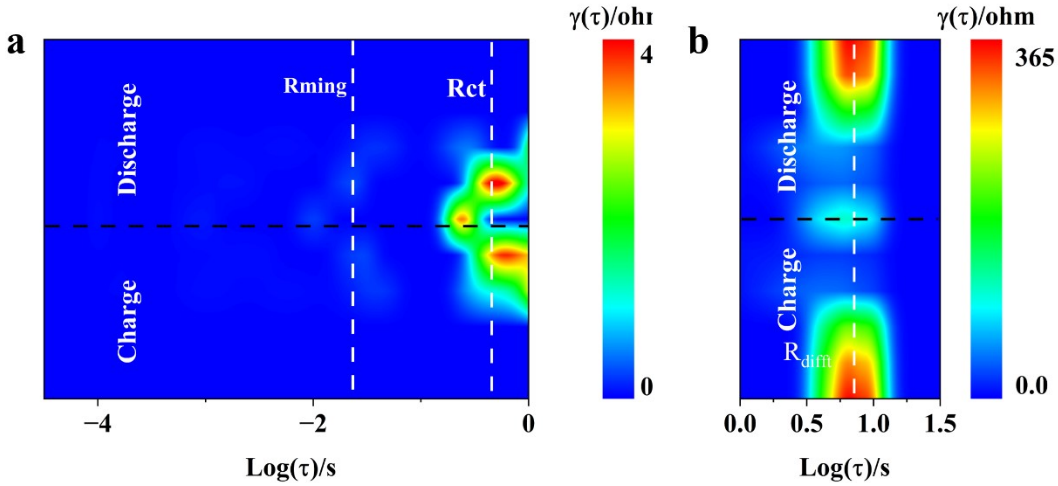


Figure S5. In-situ DRT analysis of the device's impedance during charge and discharge at a current density of 3 mA cm^{-2}.

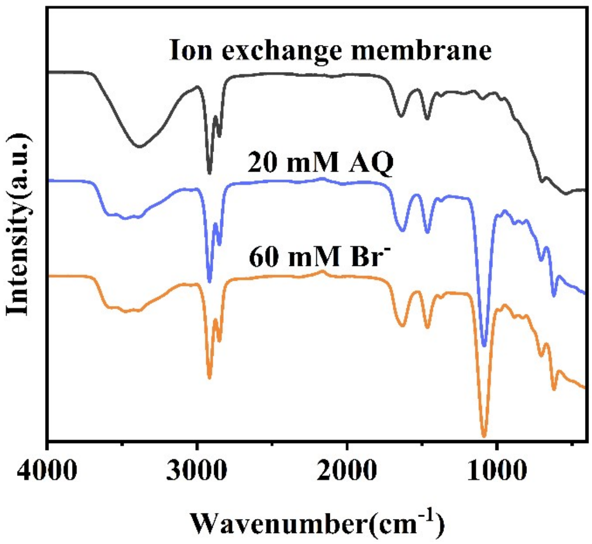


Figure S6. Infrared spectra of a blank ion-exchange membrane and one soaked in different electrolytes for 7 days.

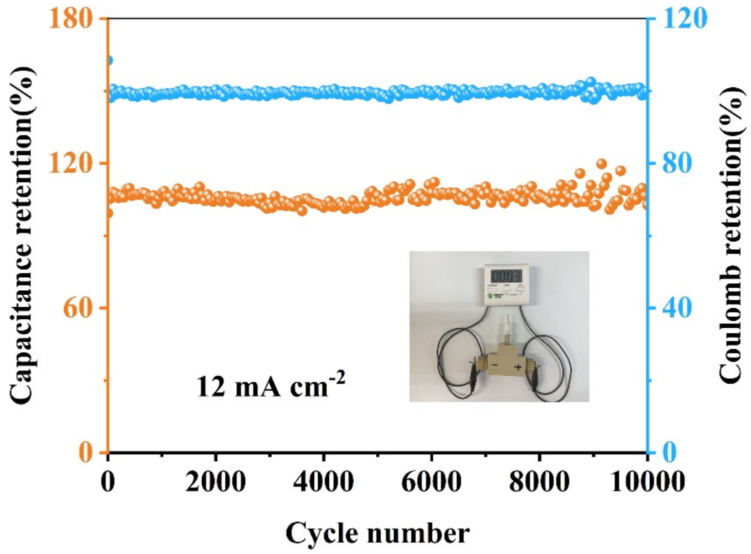


Figure S7. Cycle diagram of the device at a current density of 12 mA cm^{-2} (The accompanying figure shows an experiment in which the device lights up an alarm clock.)

Table 1 Device Performance Comparison Chart

ESW (V)	Voltage (V)	Energy density (wh cm-2)	
2.45	2.45	90	This Work
2.2	1.3	12	[1]
3.28	2.3	18	[1]
3.415	2.4	30.23	[2]
3.01	2.5	110	[3]
2.7	2.0	31.7	[4]
3.5	1.6	3.84	[5]
3.8	2.8	51.9	[6]

Reference

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