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Ultrathin cellulosic gel electrolyte with gradient hydropenic interface for stable, high-energy and flexible zinc batteries

Jichao Zhai, ^{‡a} Wang Zhao, ^{‡a} Lei Wang, ^{*a} Jianbo Shuai, ^a Ruwei Chen, ^b Wenjiao Ge, ^a Yu Zong, ^a Guanjie He^{*b} and Xiaohui Wang^{*a}

^a State Key Laboratory of Pulp and Paper Engineering, School of Light Industry Science and Engineering, South China University of Technology, Guangzhou City, Guangdong Province 510640, China. E-mail: felwang@scut.edu.cn (L. Wang), fewangxh@scut.edu.cn (X. Wang)
^b Department of ChemistryUniversity College London20 Gordon Street, London WC1H 0AJ, UK. E-mail: g.he@ucl.ac.uk (G. He)

‡ These authors contributed equally to this work.

Experimental Section

Materials. Zinc trifluoromethanesulfonate (Zn(OTf)₂, 98%), 3-Amino-1-propanol (99%) and Ammonium metavanadate (AR, 99%), were purchased from Aladdin. 1.5-Diazabicyclo[4.3.0]non-5-ene and Allyl chloride were purchased from Anhui Zesheng Technology Co., Ltd. HNO₃ and acetone were purchased from Guangzhou Chemical Reagent Factory. Cellulose were purchased from Shanghai Macklin Biochemical Technology Co., Ltd. Zn foils (99.99%), Cu foils (purity 99.99%), toray hydrophilic type carbon paper, glass fibers separators and CNT were purchased from Guangdong Canrd New Energy Technology. Glass fibre fabrics were purchased from Guangzhou Chenghe Glass Fibre Co. Bacterial cellulose were purchased from Guilin Qihong Technology Co.

Preparation of deep eutectic solvent (DES). The 1,5-Diazabicyclo[4.3.0]non-5-ene (DBN, 0.5 mol, 1 equiv) was thoroughly mixed with 30 mL of acetone.¹ Subsequently, under ice-bath conditions (5°C), allyl chloride (0.65 mol, 1.3 equiv) was slowly added. Upon complete addition, the temperature was gradually raised to room temperature, and the mixture was stirred for another 2 hours. Following this, rotary evaporation was used to remove excess allyl chloride and solvent, resulting in a supercooled, viscous liquid known as 1-allyl-2,3,4,6,7,8-hexahydropyrrolo[1,2-a]pyrimidin-1-ium chloride (DBNACl). Hydrogen bond acceptors of DBNACl, along with hydrogen bond donors (3-Amino-1-propanol), were mixed in a molar ratio of 1:1. The mixture was then heated and stirred at 90°C for 1 hour. This process yielded DES, which primarily exists in liquid state at room temperature.

Preparation of CG and DCG. A transparent cellulose solution was prepared by adding 0.3g of Cellulose to 1.7g of DES and stirring for 1 hour at 90°C. The cellulose solution was subsequently spread onto a glass plate and regenerated using deionized water (DI) and replacing the DI every hour and repeating the process 3-5 times. The cellulose gel was then immersed in

a 1.5 or 3.0 mol L^{-1} Zn(OTf)₂ solution, resulting in the cellulose gel electrolyte named CG. The cellulose solution was re-prepared as previously described, spread onto a glass plate, and weighed in a ratio of cellulose solution to 1.5 or 3.0 mol L^{-1} Zn(OTf)₂ solution at a weight ratio of 1:99. It was then allowed to regenerate in the electrolyte solution for 8 hours, the cellulose gel electrolyte was named DCG.

Preparation of electrodes. V_2O_5 nanowires were prepared by a hydrothermal method.² In a typical process, 0.1 g ammonium metavanadate (99%) was dispersed in 40 mL deionized water under intensive stirring. Then, HNO₃ was dropped to adjust the pH value of the solution to 2. The mixture was stirred at room temperature for 1 h and then transferred into 60 mL. Teflon-lined autoclave and heated at 180 °C for 24 h. After hydrothermal reaction, the autoclave was cooled down to room temperature naturally. The rich mahogany precipitates were washed with DI water and alcohol for several times then dried at 80 °C under vacuum for 12 h. The paper cathode was prepared by mixing V₂O₅ NWs, CNT, and Bacterial cellulose at a weight ratio of 8:1:1 using DI as the solvent, then filter and freeze-drying (with the mass loading of V₂O₅ being approximately 5 mg cm⁻²). The ZnGF flexible anodes were prepared by metal melting process with flexible substrate CuGF.³

Materials Characterization. Proton nuclear magnetic resonance (¹H NMR) spectra of DBNACl, 3-Amino-1-propanol, and DES were measured on an Avance 500 MHz spectrometer (Bruker, USA), using DMSO-d₆ as solvent. Fourier transform infrared (FTIR) spectra were recorded on a TENSOR 37 spectrometer (Bruker Corporation, German) in the wavenumber range from 4000 to 400 cm⁻¹. X-ray diffraction (XRD) measurements were performed on a X' pert Powder diffractometer (PANalytical, netherlands). The patterns of CuK α radiation (λ = 0.15418 nm) at 40 kV and 15 mA were recorded in the 2 θ region from 5 to 90° at a scanning speed of 12° min⁻¹. X-ray photoelectron spectra (XPS) were recorded on a K-Alpha

spectrometer (Thermo Scientific, USA). Field emission scanning electron microscope (FESEM) observations were performed on a SU5000 (Hitachi, Japan). Atomic force microscope (AFM) were performed on a multimode 8 (Bruker, German). The concentration of solution was measured using a UV–2600i spectrophotometer (Shimadzu, Japan) in the wavelength range from 200 to 800 nm. Optical micrographs of Zn anodes were recorded using an MSD380 upright metallurgical microscope (Murzider, China). All tensile test were operated on a 3400-material testing machine (INSTRON, USA). All freeze-drying were operated on LGJ-12 (Songyuan Huaxing, China).

Electrochemical measurements. All the electrochemical measurements were operated at 25 °C unless otherwise specified. The charge–discharge measurements of the batteries were conducted on a battery testing system (CT-4008Tn, NEWARE, China). The voltage window was 0.2-1.8 V for Zn||V₂O₅ batteries. The cyclic voltammetry (CV), linear scan voltammetry (LSV), chronoamperometry (CA), electrochemical impedance spectroscopy (EIS) and linear polarization resistance (LPR) were carried out on an electrochemical work station (CS310X, CorrTest, China). For EIS tests, the frequency range was from 10^6 Hz to 10^{-2} Hz and the voltage amplitude was 10 mV. LPR tests were conducted using a three-electrode system made up of Zn, Zn, and Ag/AgCl electrolyte in the three-electrode system, the Zn electrode was wrapped with the hydrogel and then immersed in the liquid electrolyte. LSV measurements were conducted using the same device. The ionic conductivities (σ) were obtained from EIS measurements and calculated by the following equation,

$$\sigma = \frac{l}{RA} \tag{1}$$

where l, R, and A are the thickness, bulk resistance, and area of the cellulose gel electrolyte, respectively.

The desolvation process of Zn^{2+} is usually the rate-limiting step of Zn deposition, which can be expressed by the activation energy (Ea) in the Arrhenius equation,

$$\frac{1}{R_{ct}} = Ae^{-\frac{E_a}{RT}}$$
(2)

where R_{ct} is the charge transfer resistance, A is the frequency factor, R is the gas constant, and T is the absolute temperature.

The transference numbers were measured according to the chronoamperometry (CA) measurement on Zn||Zn symmetric battery by applying a potential of 150 mV at room temperature. The current response was measured as a function time for 1000 seconds. The AC impendence measurement was conducted over the frequency range of 10^{6} Hz to 10^{-2} Hz. The transference number is calculated according to the following equation:

$$t^{+} = \frac{I_{s}(\Delta V - I_{0}R_{0})}{I_{0}(\Delta V - I_{s}R_{s})}$$
(3)

Where I_s is the steady-state current, I_0 indicates the initial current, ΔV represents the applied potential, R_0 indicates the initial resistance of the passivation layer and R_s is the steady-state resistance of the passivation layer. R_0 and R_s are determined from the AC impedance measurement immediately before and after DC polarization measurement.

Density functional theory (DFT) calculation. DFT calculations are performed by the DMol3 -module of the Material Studio software (version 2023). The exchange-correlation energy was approximately described by the Perdew-Burke-Ernzerhof (PBE) functional based on the generalized gradient approximation (GGA). The convergence tolerance of energy, force and displacement convergence were set as 1×10^{-5} Ha, 2×10^{-3} Ha, and 5×10^{-3} Å, respectively. The core treatment was chosen as the effective core potential (ECP), and the electron treatment was performed by double numerical plus d-functions (DNP) basis set. The Zn (002) surface is modeled by a five-layers 5×5 surface supercell, the Zn (101) surface is modeled by a 6×4 surface supercell and the Zn (100) surface is modeled by a five-layers 3×6 surface supercell, all of which have 180 Zn atoms.

The binding energy was defined as the following equation (4):

$$E_{\text{binding}} = E_{Zn^{2+}+molecules} - E_{Zn^{2+}} - E_{\text{molecules}}$$
(4)

The adsorption energy between Zinc crystal surface and different molecules was calculated according by the formula (5):

$$E_{adsorption} = E_{Zn(x)+molecules} - E_{Zn(x)-molecules}$$
(5)

Molecular Dynamics (MD) method. The simulations were performed using Material Studio software (version 2020) with the Universal force field (UFF). The Perdew-Burke-Ernzerhof (PBE) functional in conjunction with the double numerical basis sets and polarization functional (DNP) was used in the MD simulations. The simulated solution box was constructed by Amorphous Cell (AC) Module with 200 H₂O, and 1 DES. The Zn (002) surface is modeled by a five-layers 5×5 surface supercell, which have 180 Zn atoms. The system was also used to optimize these structures with forcite Module. The box was equilibrated under the NPT ensemble at a constant temperature of 298 K and 0.1 MPa for 10 ns with an integration time-step of 2 fs.

Phase-field model. In order to simulate the dynamic Zn deposition on electrodes indifferent electrolytes, a Finite Element Analysis (FEA) model was performed using COMSOL Multiphysics 6.2 software with the "Tertiary Current Distribution" and "Phase Field" module. The size of the entire two-dimensional model for electric field distribution analysis was set to $11 \times 5 \mu m$. A transient simulation of the process was carried out in an area filled with electrolyte. The current density was set to 1 mA cm^{-2} and the plating/stripping process was simulated using

the phase field method.

The Zn^{2+} transfers by the concentration diffusion in model follow the Fick's law as shown in equation (6) and (7):

$$N_i = J_i = -D_i \nabla c_i \tag{6}$$

$$\frac{\partial c_i}{\partial t} + \nabla \cdot J_i = R_{i,tot} \tag{7}$$

Where *Ji* is the ion flux, *Di* is the diffusion coefficient of electrolytes ($DZn = 1.0 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$ for Zn(OTf)₂ electrolytes), *ci* is the ion concentration of electrolytes ($c_i = 1500 \text{ mol m}^{-3}$), ∇ci is concentration gradient.

The relation between the diffusion coefficient and electric mobility follows the Nernst-Einstein relation as shown in equation (8):

$$N_i = -D_i \nabla c_i - z_i u_{m,i} F c_i \nabla \phi_i + u c_i = J_i + u c_i$$
(8)

Where z_i is the transfer number ($z_{Zn} = 1$), $u_{m,i}$ is the electric mobility coefficient, F is the Faraday constant (96485 C mol⁻¹), ϕ is the electrolyte potential.

The equilibrium potential of the electrode surface follows the Nernst equation as shown in equation (9) and (10):

$$E_{eq} = -\frac{\Delta G}{nF} \tag{9}$$

$$E_{eq} = E_{eq,ref} - \frac{RT}{nF} ln \prod_{i} \left(\frac{a_i}{a_{i,ref}}\right)^{\nu_i}$$
(10)

Where E_{eq} is the electrode potential, $E_{eq,ref}$ is the standard electrode potential, ΔG is the Gibbs free energy, R is the ideal gas constant, T is the temperature (T = 298 K), n is the transfer electron number of the reaction, α_i is the (electrode reactive ion concentration), $\alpha_{i,ref}$ is the (standard electrode reactive ion concentration), v_i is the reaction stoichiometric number. The electrode reaction for the electrode surface follow the Butler-Volmer kinetics expression, as shown in equation (11):

$$i_{loc} = i_0 \left(\exp\left(\frac{\alpha_a F \eta}{RT}\right) - \exp\left(\frac{-\alpha_c F \eta}{RT}\right) \right)$$
(11)

Where i_{loc} is the local current density at the electrode/electrolyte interface, i_0 is the exchange current density ($i_0 = 1.0 \text{ A m}^{-2}$), and is the cathodic and anodic charge transfer coefficients ($\alpha_a = 0.5$, $\alpha_c = 1.5$), η is the activation overpotential.

Supporting Figures



	H ₁	H ₂	H ₃	H ₄	H₅	H ₆	H ₇
DBNACI	2.02	3.03	3.38	3.69	4.09	5.30	5.88
DES	2.03	3.02	3.38	3.69	4.08	5.30	5.88
DES+H ₂ O	2.03	3.01	3.38	3.69	4.07	5.30	5.88
	H _a	H _b	H _c				
3-Amino-1-propanol	1.48	2.59	3.45				
DES	1.52	2.62	3.45				
DES+H ₂ O	1.53	2.62	3.45				

Fig. S1 ¹H NMR spectra of DBNACl, 3-Amino-1-propanol, DES and DES in water.

Following the formation of DES, the majority of the spectral signals represent a superposition of acceptor and donor signals. This process also induces the ionic liquid to exhibit a weak peak adjacent to the original signal peak. This occurs due to the partial shift of the allyl group on DBNACl to the donor's amino group, resulting in the formation of a small quantity of allylated propanolamine derivatives. After DES formation, the displacement of DBNACl signal peaks remains largely unchanged. However, the two signals (H_a , H_b) associated with the donor's amino group shift to a lower field (increased chemical shift), while the signal peak (H_c) linked to the hydroxyl group remains unchanged. This suggests that the donor amino group forms additional hydrogen bonds with the acceptor, leading to a reduced electron cloud density around

the attached carbon atoms and an enhanced deshielding effect. Based on the DES structure, the addition of a small amount of water molecules does not significantly change the signal region shifts, indicating that the addition of water does not affect the interactions between donors and acceptors.



Fig. S2 The performance of Zn||Zn battery with different DES concentration.

We evaluated the performance of Zn||Zn batteries using gel electrolytes with varying concentrations of DES and discovered that the electrolyte containing 1% DES exhibited the lowest polarization voltage and the longest cycle life.



Fig. S3 (a) Photographs of a 1.5 M Zn(OTf)₂ solution containing 1% 3-Amino-1-propanol. (b) The performance of a Zn||Zn battery using a 1.5 M Zn(OTf)₂ solution containing 1% 3-Amino-1-propanol and 1% DBNACl.

The two components of DES were individually introduced into $1.5 \text{ M Zn}(\text{OTf})_2$ to confirm DES's role in the electrolyte. The addition of 3-amino-1-propanol resulted in the formation of a white precipitate, as illustrated in Fig. S3a. Additionally, Fig. S3b demonstrates that the addition of these two components does not enhance battery performance.



Fig. S4 The XRD patterns of Cellulose, CG and DCG.

Following cellulose dissolution-regeneration process, we observed a transformation in the cellulose crystal structure from type I to type II. This observation aligns with the characteristics commonly associated with the cellulose dissolution-regeneration process.⁴



Fig. S5 Determination of DES content in DCG gel electrolytes. (a) UV spectra of the solidification bath following DCG regeneration, and $Zn(OTf)_2$ electrolytes with different DES concentrations. (b) Concentration curves derived from absorbance measurements at 222 nm.

The absorbance of the solidification bath at 222 nm was measured at 2.01. The DES content was quantified as 1% through curve fitting, thereby confirming that the DCG contained 1% DES.



Fig. S6 Illustration of coordination effect of Zn cations with cellulosic chains.

The cellulose gel electrolyte regenerated in $Zn(OTf)_2$ electrolyte allows part of the hydroxyl group to form polar coordination with Zn^{2+} , creating a rapid Zn^{2+} transport channel and enhancing tensile strength.



Fig. S7 Ionic conductivities of LE, CG and DCG.

According to the EIS test, the ionic conductivity of the DCG gel electrolyte was determined to be 29.5 mS cm⁻², significantly higher than that of CG (19.4 mS cm⁻²) and LE (14.7 mS cm⁻²).



Fig. S8 The Raman spectrum of Cellulose, CG and DCG.



Fig. S9 Thickness measurement of the DCG electrolyte.



Fig. S10 The gel electrolytes performance comparison between our developed DCG with previously reported gel electrolyte technologies in terms of thickness, ionic conductivity, and tensile strength.

Figure S10 provides a comparison of the properties of various gel electrolytes, focusing on thickness, ionic conductivity, and tensile strength. The DCG exhibits significantly superior performance, characterized by its ultra-thin thickness (10 μ m), high tensile strength (0.82 MPa), and enhanced ionic conductivity (29.5 mS cm⁻¹), outperforming previous gel electrolytes.



Fig. S11 Zeta potential of DES.



Fig. S12 The adsorption energy of -NH₂ and H₂O on the Zn^{2+} .

It has been observed that the adsorption energy between DES and Zn^{2+} is higher than that between H₂O and Zn²⁺. The DES residues in the gel electrolyte can bind Zn²⁺, disrupting the solvation structure, thereby reducing the activation energy and facilitating the rapid transfer of Zn²⁺ at the electrode-electrolyte interface.



Fig. S13 The EIS curves of the Zn||Zn battery with a LE, b CG and c DCG from 20 to 60°C.



Fig. S14 (a), (c) and (e) CA curves of Zn||Zn symmetric batteries with different electrolytes. (b), (d) and (f) the corresponding Nyquist plots before and after polarization tests.



Fig. S15 The corresponding t_{Zn}^{2+} for different electrolytes.



Fig. S16 The COMSOL simulation results of the Zn dendrite growth in CG.



Fig. S17 The XPS F1s spectra of Zn anode with CG and DCG in the Zn||Zn batteries after 20 cycles.

The presence of element F suggests that a side reaction occurs, forming by-products $(Zn_x(OTf)_y(OH)_{2x-y} \cdot nH_2O)$ on the Zn anode of the Zn||Zn battery with CG after 20 cycles.



Fig. S18 Surface SEM images of Zn foils at deposited state after cycling (1 mA cm⁻² 1 mAh cm⁻²) for different numbers of cycles (5, 10 and 15) in the Zn||Zn batteries with DCG.

The formation of hexagonal zinc plates indicates that the use of DCG facilitates the deposition of Zn^{2+} in a (002) crystalline structure, leading to a more uniform electrodeposited Zn surface.



Fig. S19 Surface SEM images of Zn foils at deposited state after cycling (1 mA cm^{-2} 1 mAh

 cm^{-2}) for different numbers of cycles (5, 10 and 15) in the Zn||Zn batteries with LE.



Fig. S20 Surface SEM images of Zn foils at deposited state after cycling (1 mA cm^{-2} 1 mAh

 cm^{-2}) for different numbers of cycles (5, 10 and 15) in the Zn||Zn batteries with CG.



Fig. S21 The cross-sectional SEM images of the Zn anode after plating 20 cycles with (a, b) DCG, (c, d) LE and (e, f) CG.

The cross-sectional SEM images of the Zn anode after 20 cycles further show that the use of DCG results in a uniform deposition of Zn^{2+} .



Fig. S22 AFM images of the zinc anode after 20 cycles using (a) DCG, (b)LE and (c)CG at a current density of 1 mA cm⁻² and a discharge capacity of 1 mAh cm⁻².



Fig. S23 The SEM images of the Zn metal after plating for 20 cycles with different electrolytes.



Fig. S24 The photographs of **a** DCG and **b** CG after cycling (4 mA cm⁻² 4 mAh cm⁻²) for different numbers of cycles

The DCG gel electrolyte remains intact without dendrite residues after 500 cycles in a Zn||Zn battery. In contrast, the CG breaks down, and dendrites puncture the gel after only 200 cycles.



Fig. S25 The performance of Zn||Zn battery with LE, CG and DCG under 30 mA cm⁻², 30 mAh cm⁻².



Fig. S26 The photographs of ZnGF.

The ZnGF flexible anodes, prepared through a metal melting process using a flexible substrate, exhibit excellent flexibility and have a thickness of only 30µm.



Fig. S27 The XRD patterns of V₂O₅ NWs.

The pattern of the prepared V_2O_5 is in well accordance with the standard diffaraction peaks (JCPDS: PDF #77-241). The synthesized V_2O_5 displays peaks at 15.4° (200), 20.3° (001), and 31.0° (301).



Fig. S28 The SEM images of V_2O_5 NWs.

The SEM revealed that the synthesized V_2O_5 possesses a nanowire structure.



Fig. S29 The photographs of flexible V_2O_5 paper electrode

The thickness of the paper cathode can be adjusted by controlling the amount of active material used, and it exhibits excellent flexibility.



Fig. S30 The photograph of flexible $Zn \parallel V_2O_5$ pouch cell with DCG.



Fig. S31 Resistance changes of flexible electrodes and metal foils as a function of bending cycles at a bending radius of 2 mm.

Compared to Zn and Cu foils, the prepared flexible electrodes exhibit superior flexibility, and their resistance remains stable after 5000 bending cycles, which is advantageous for the development of flexible pouch cells.



Fig. S32 CV profiles of the Zn $\|V_2O_5$ battery with LE, CG and DCG at 1 mV s⁻¹



Fig. S33 The GCD curves of the $Zn||V_2O_5$ battery with (a) LE and (b) CG at 1 A g⁻¹



Fig. S34 The GCD curves of the $Zn||V_2O_5$ battery with (a) LE, (b) CG and (c) DCG at different current densities



Fig. S35 The EIS curves for the $Zn \| V_2 O_5$ batteries with LE, CG and DCG.

Sample		Thickness	Ionic	Tensile	
	Material	(µm)	conductivity	strength	Reference
			$(mS cm^{-1})$	(MPa)	
DCG	Cellulose fiber	10	29.5	0.82	This work
PZIL	Carboxymethyl chitosan	30	26.3	0.56	5
COOH-f- CellPZ-gel	Carboxylic acid				
	functionalized cellulose /	239	19.6	0.7	6
	PVA				
SFPAM-Zr	Silk fibroin / PAM	302	22.9	0.45	7
HE-PAM	PAM	500	22.5	0.16	8
HE-RS	PVA	680	4	0.02	9

Table. S1. Comparisons gel electrolytes performance with recently reported literatures (Fig.S10)

Sample	Gravimetric energy	Volumetric energy	Reference	
	density	density density		
	$(Wh kg^{-1})$	$(Wh L^{-1})$		
DCG Zn V2O5	222.2	214.3	This work	
Se-in-CuCo(CN) ₆ Zn Se	60.6	137.8	10	
12-C-4 Zn Zn _{0.25} V ₂ O ₅ -	51.1	104	11	
nH ₂ O				
$T\text{-}ZnSe \; Zn \ I_2$	32	101	12	
TN-20-ZS Zn VO ₂	42.3	79.5	13	
$RME \ Zn \ Zn_{0.25}V_2O_5\text{-}nH_2O$	31	70	14	
PHE Zn LNMO	25	54	15	
$Zn(DFTFSI)_2 \ Zn-Br_2$	23.3	43	16	
SiBE Zn ZnVO	18.9	43	17	
PAZPM Zn NVO	21.6	28.7	18	
$GFA-5 \ Zn \ I_2$	19.3	134	19	
HT-Zn Zn $\ I_2$	15.9	16.5	20	
Zn@MCFs Zn a-MnO2	10.8	13.4	21	

Table. S2. Comparisons energy density with recently reported literatures for Zn pouch cells

 (Fig. 5f)

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