

Binary ionic liquid hybrid electrolyte based supercapacitors with high energy & power density

Zheng Bo¹, Xu Zhang¹, Zhesong Huang¹, Yuhui Huang^{2,3}, Jianhua Yan¹, Kefa Cen¹ and Huachao Yang^{1,3*}

¹State Key Laboratory of Clean Energy Utilization, Institute for Thermal Power Engineering, College of Energy Engineering, Zhejiang University, Hangzhou 310027, Zhejiang, China.

²College of Materials Science and Engineering, Zhejiang University, Hangzhou, 310027, Zhejiang, China.

³Research Institute of Zhejiang University-Taizhou, Taizhou, Zhejiang Province 318000, China.

E-mail: huachao@zju.edu.cn

Supplementary section

Table S1 display the results of replicate experiments in working voltage tests. As shown in the Table S1, the coulombic efficiency of 1M [TMPA]_{0.5}[Pyr₁₄]_{0.5}[TFSI]/ACN decreases below 95% when the applied voltage is over 3.1 V.

Table S1 Coulombic efficiency of 1 M [TMPA]_{0.5}[Pyr₁₄]_{0.5}[TFSI]/ACN in different voltage window (the scan rate is 20 mV S⁻¹)

Voltage window (V)	Coulombic efficiency (%)		
	Group 1	Group 2	Group 3
0 - 2.7	99.51	98.94	99.49
0 - 2.9	98.89	98.85	98.83
0 - 3.1	97.70	97.66	97.67
0 - 3.3	94.79	94.86	94.43
0 - 3.5	94.32	94.49	94.27

In order to calculate the theoretical value of operating voltage of 1 M [TMPA]_{0.5}[Pyr₁₄]_{0.5}[TFSI]/ACN, LSV tests were performed at the scan rate of 42 μV s⁻¹ in a three-electrode system in negative range (from 0 to -3 V) and positive range (from 0 to 3 V), respectively.¹ In the three-electrode system, the working electrode and counter electrode were activated carbon with a diameter of 8 mm, and a silver wire was used as pseudo-reference electrode.

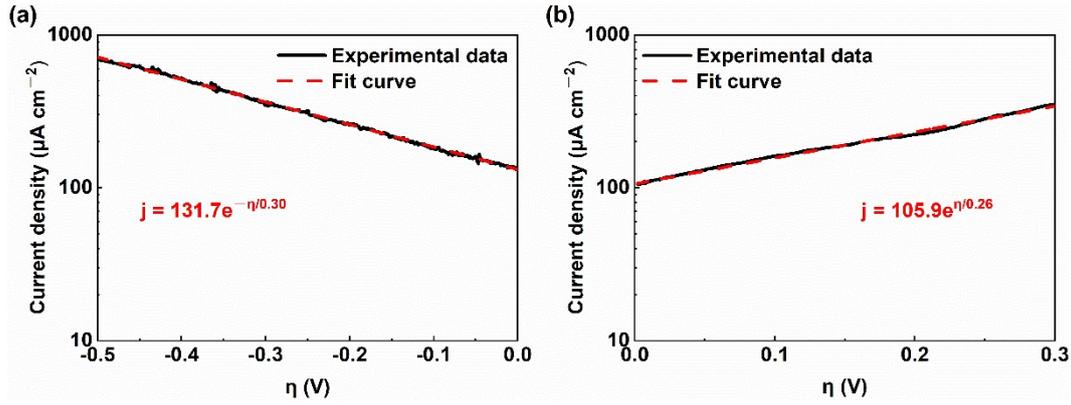


Fig. S1 (a) cathodic and (b) anodic Tafel plots (linear regions – i.e., where the current density varies exponentially with the overpotential) for 1 M [TMPA]_{0.5}[Pyr₁₄]_{0.5}[TFSI]/ACN. In (a), η is the difference between the applied potential and -1.6 V and in (b), η is the difference between the applied potential and 1.2 V.

As shown in Fig. S1, the Tafel curves show that the current density increases exponentially with the overpotential. Therefore, the current density can be modelled with the equation below,

$$j = j_0 e^{\eta/\beta} \quad (9)$$

where j and j_0 are, respectively, the current density and the exchange current density; β is the Tafel coefficient and η is the overpotential at which the anodic/cathodic reaction is taking place. And the operational voltage ΔV_{max} can be calculated by the equation below,

$$\Delta V_{max} = \Delta V_0 + \frac{\beta_a}{\log e} \log\left(\frac{j_{max}}{j_{0,a}}\right) + \frac{\beta_c}{\log e} \log\left(\frac{j_{max}}{j_{0,c}}\right) \quad (10)$$

where ΔV_0 is the theoretical cell voltage at equilibrium; j_{max} is the maximum current density considered acceptable (which will be the same for the positive and negative electrodes, unless they have different areas or mass loadings); $j_{0,a}$ and $j_{0,c}$ are, respectively, the exchange current density for the anodic and for the cathodic parasitic reactions; and β_a and β_c are, respectively, the Tafel coefficients for the anodic and for the cathodic parasitic reactions.

Because the BILs electrolyte is non-aqueous system, the acceptable parasitic current is selected as 0.2 mA cm^{-2} , and the theoretical value of maximum working voltage is 3.1 V, which is consistent with the results of working voltage test.

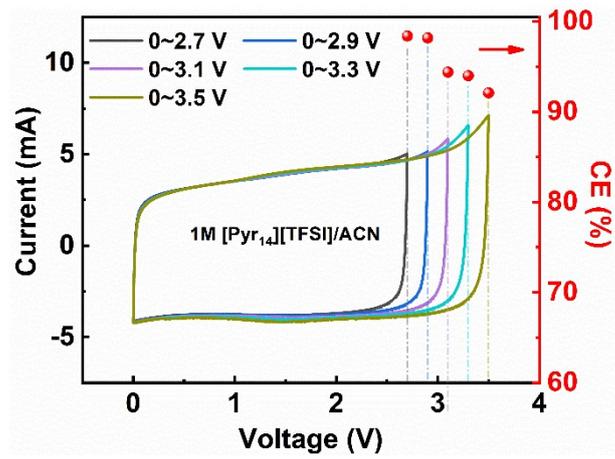


Fig. S2 CV curves and coulombic efficiency of 1 M [Pyr₁₄][TFSI]/ACN at 20 mV s⁻¹.

As shown in Fig. S2, the device using 1 M [Pyr₁₄][TFSI]/ACN displays a coulombic efficiency lower than 95% when the operation voltage is over 3.1 V.

References in Supplementary

- 1 J. M. Baptista, G. Gaspar, K. G. U. Wijayantha and K. Lobato, *J. Energy Storage*, 2022, **52**, 104927.