Supplementary Information

A 7.5 V window dual-functional planar micro-device based on biopolymer ionogel electrolyte for charge storage and neuromorphic computing

Simantini Majumdar*, Ann Mary Antony and Giridhar U. Kulkarni

Chemistry and Physics of Materials Unit, Jawaharlal Nehru Centre for Advanced Scientific Research (JNCASR), Bengaluru 560064, India

*Corresponding Author.

Email: simantini.energy@gmail.com



Figure S1. (a-e) FTIR absorption spectra in the region of 600 to 655 cm⁻¹ for GB_x ($15 \le x \le$ 50) blended biopolymer ionogel electrolytes with different BMIMBF₄ compositions. Deconvoluted FTIR absorption spectra based on Gaussian fitting function for the GB_x ionogels are represented by solid lines.



Figure S2. (a-e) FTIR absorption spectra in the region, 1200–1400 cm⁻¹ for GB_x ($15 \le x \le 50$) blended biopolymer ionogel electrolytes with different BMIMBF₄ compositions. Solid lines show the Gaussian fitting of the characteristic absorption peaks. (f) Percentage area under the peak corresponding to amide-III band is shown as a function of BMIMBF₄ doping concentrations in GB_x ($15 \le x \le 50$) ionogels.



Figure S3. Cyclic voltammetry of the device measured at a scan rate of 0.1 V/s at voltage windows of 8 V and 9 V.



Figure S4. (a) Cyclic voltammetry of the micro-devices measured at a scan rate of 0.1 V/s at different voltage windows (1-9 V) developed with ionogels (a,b) GB₁₅, (c,d) GB₂₅, (e) GB₃₅, and (f) GB₅₀.



Figure S5. Cyclic voltammetry of the device measured at a scan rate of 10 V/s at different voltage windows (1-7.5 V).



Figure S6. Galvanostatic charge-discharge for the device measured at a voltage window of 7.5 V under varying current densities, (a) 1.5, (b) 2, (c) 2.5, (d) 3, and (e) 4 mA/cm².

Current Electrode Ionic liquid-based ESW Capacitance Referenc Collector Coating material electrolytes (V) es Polyacrylonitrile **PVA-PAA** Ionogel 2 615 mF/cm^2 1 Aluminum -derived porous foil carbon fibers (PPCF) 2 Activated PAAm/ChCl/ 43.81 F/g Graphite 2.4 Carbon [EMIM][TFSI]IL paper EMImBF₄ $149 \ \mu F/cm^2$ 3 Au Vertically 4 oriented graphene ionogel (GI) Printed Carbon PVA-[DPTA][BF4] 4 Silver ink 1.2 98.8 mF/cm² Graphite PAAM-PVA/ 1.6 284.8 5 Activated mF/cm² Carbon BMIMBr IL paper PIP13FSI/ PYR14FSI/ 90 F/g 6 Activated 3 Au Carbon SiO_2 Aluminum Carbon AAm/ 2 37.35 F/g 7 foil [EMIM]⁺[NO₃]⁻/ PEGDA/ 3D **PVDF-HFP** 4 323 F/g 8 interconnected /[EMI][BF4] large mesoporous carbon 9 Ionic liquid pre-EMIMBF₄/PVDF-3 None 44 mF/cm^2 intercalated HFP ionogel MXene (PM) None CS/GL/LiClO₄/BMI 7.5 5.78 F/cm² Present Au **MBF**₄ ionogel work

 Table 1. Supercapacitive performance of CS/GL blended biopolymer ionogel electrolyte

 compared with other ionic liquid-based electrolyte materials.



Figure S7. (a) Galvanostatic charge-discharge for the device measured at a fixed current density of 2 mA/cm² under varying potential windows (1-7 V), (b) Variation of areal capacitance with the potential window at a fixed current density of 2 mA/cm².



Figure S8. (a) Galvanostatic charge-discharge for the device measured at a fixed current density of 2 mA/cm^2 for 1^{st} , 400^{th} and 500^{th} cycle, (b) Cycling stability for the device over 500 cycles.



Figure S9. Nyquist plot of the device over a frequency range of 1MHz to 1 Hz, solid line represents the modelled plots based on the equivalent circuit fitting as shown in the inset.



Figure S10. Synaptic behaviour emulated with pulsed voltage signals and fixed pulse interval $(t_i = 2 \text{ s})$ with varying number of pulses (N = 3, 5, 7, 10, 15, 20) at a pulse width (t_w) , (a) 0.2 s, (b) 0.4 s, and (c) 0.7 s.

References

¹Y. Wang, Z. Wei, T. Ji, R. Bai and H. Zhu, *Small*, 2024, **20**, 2307019.

² G. Ma, F. Pan, X. Zhou, Z. Yong, X. Wang, C. Li, W. Bai and S. Wang, *ACS Appl. Electron*. *Mater.*, 2024, **6**, **2**, 1434–1443.

³ F. Chi, Y. Hu, W. He, C. Weng, H. Cheng, C. Li and L. Qu, *Small*, 2022, **18**, 2200916.

⁴A. Satheesh, S. Sushil, A. Bharathan, P. Navaneeth, P. V. Suneesh and E. Kandasamy, *Materials Letters*, 2024, **358**, 135891.

⁵ Z. Yong, S. Wang, X. Wang, G. Liu, D. Liang, Y. Cui, F. Liu, D. Wang and Z. Wang, *Journal of Alloys and Compounds*, 2021, **893**, 162197.

⁶L. Negre, B. Daffos, V. Turq, P. Taberna and P. Simon, *Electrochimica Acta*, 2016, **206**, 490– 495.

⁷ J. Park and J-Y. Sun, ACS Appl. Mater. Interfaces, 2022, **14, 20**, 23375–23382.

⁸ K. G. Cho, H. S. Kim, S. S. Jang, H. Kyung, M. S. Kang, K. H. Lee and W. C. Yoo, *Adv. Funct. Mater.*, 2020, **30**, 2002053.

⁹ S. Zheng, C. Zhang, F. Zhou, Y. Dong, X. Shi, V. Nicolosi, Z. Wu and X. Bao, *Journal of Materials Chemistry A*, 2019, **7(16)**, 9478–9485.