

Supporting Information for

High-Voltage Flexible Aqueous Zn-Ion Battery with Extremely Low Dropout Voltage and Super-Flat Platform

Zhe Chen^{1,2,3,#}, Panpan Wang^{1,2,3,#}, Zhenyuan Ji^{1,2,3}, Hua Wang^{1,2,3}, Jie Liu^{1,2,3}, Jiaqi Wang^{1,2,3}, Mengmeng Hu^{1,2,3}, Yan Huang^{1,2,3,*}

¹State Key Laboratory of Advanced Welding and Joining, Harbin Institute of Technology, Shenzhen 518055, People's Republic of China

²Flexible Printed Electronic Technology Center, Harbin Institute of Technology, Shenzhen 518055, People's Republic of China

³School of Materials Science and Engineering, Harbin Institute of Technology, Shenzhen 518055, People's Republic of China

#Zhe Chen and Panpan Wang contribute equally to this work

*Corresponding author. E-mail: yanhuanglib@hit.edu.cn (Yan Huang)

Supplementary Table and Figures

Table S1 The ionic conductivity comparison of common zinc ion gel electrolytes

Gel types	Ionic conductivity mS cm ⁻¹	References
HPE	2.04	This work
GE	1.09	This work
PEO	0.5~0.8	[S1]
PEO	2~4	[S2]
Poly-ε-caprolactone	0.88	[S3]
Poly(4-vinylpyridine)	2×10 ⁻⁵	[S4]

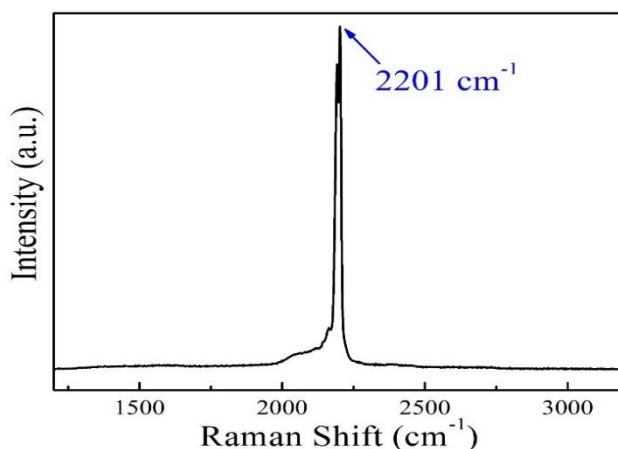


Fig. S1 Raman spectrum of ZnHCF active materials

The wavenumber peak of $\nu(\text{CN})$ located at 2201 cm^{-1} corresponds to the stretching vibration mode of the cyanide CN^- that coordinated to Fe(III) , which accords with the structure of ZnHCF . [S5].

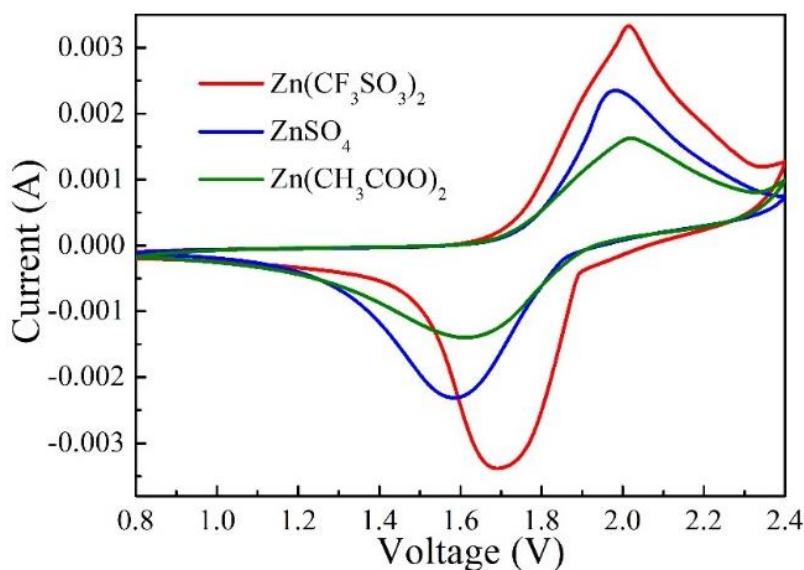


Fig. S2 CV curves of three zinc salts ($\text{Zn}(\text{CF}_3\text{SO}_3)_2$, ZnSO_4 , $\text{Zn}(\text{CH}_3\text{COO})_2$) at 5 mV s^{-1} scan rate

In the picture, the charging/discharging voltage $2.01/1.69\text{ V}$ of $\text{Zn}(\text{CF}_3\text{SO}_3)_2$, $1.98/1.58\text{ V}$ of ZnSO_4 , $2.01/1.62\text{ V}$ of $\text{Zn}(\text{CH}_3\text{COO})_2$ give a distinct result that $\text{Zn}(\text{CF}_3\text{SO}_3)_2$ is more suitable for electrolytes of ZIBs to give full play to their high voltage performance.

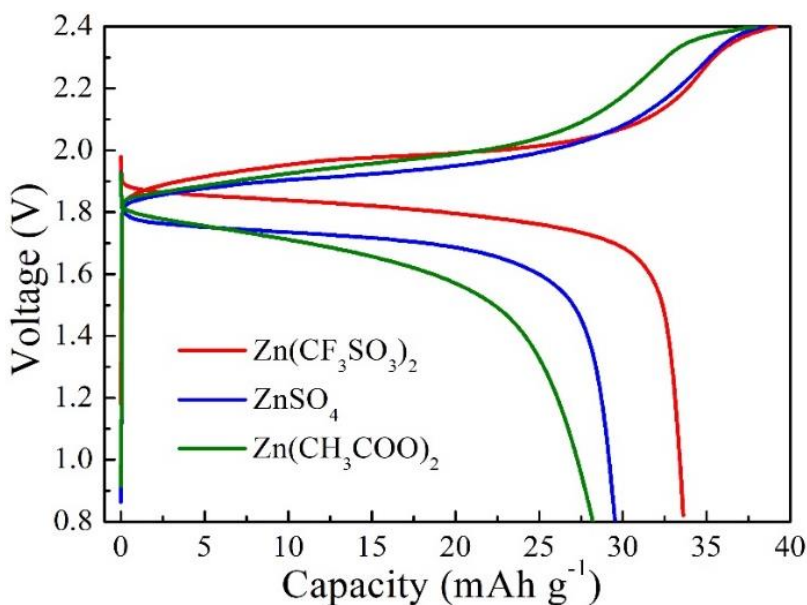


Fig. S3 GCD plots of three zinc salts ($\text{Zn}(\text{CF}_3\text{SO}_3)_2$, ZnSO_4 , $\text{Zn}(\text{CH}_3\text{COO})_2$) at 12.5 C

$\text{Zn}(\text{CF}_3\text{SO}_3)_2$ shows the highest capacity and excellent voltage platform, as well as the smallest dropout voltage among the three salts.

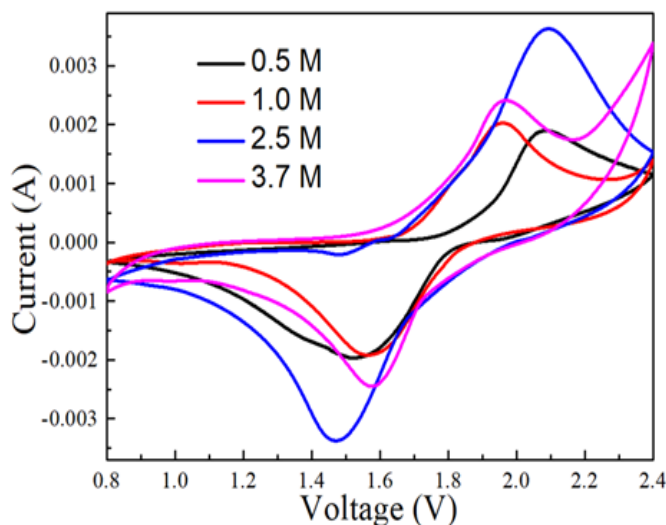


Fig. S4 CV curves of four $\text{Zn}(\text{CF}_3\text{SO}_3)_2$ concentrations

Four concentration gradients (0.5 M, 1 M, 2.5 M, 3.7 M) were selected to perform CV tests at 10 mV s^{-1} under as same battery conditions. The results show that 1 M is significantly better than other concentrations.

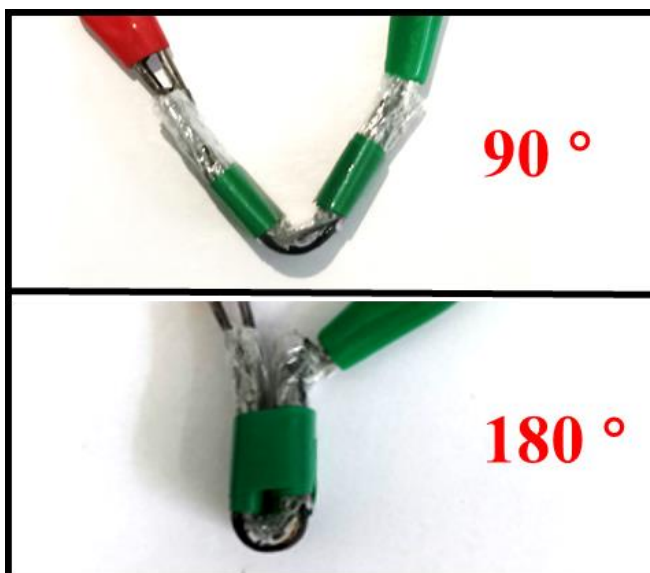


Fig. S5 Images of the battery at different bending angles

Batteries assembled in sandwich structures bent at 90° and 180° respectively, which shows the electrode is in good contact with the electrolyte. Despite being covered with a thick protective film, the battery still maintains good flexibility.

Supplementary References

- [S1] I. Pucić, A. Turković, Radiation modification of $(\text{PEO})_8\text{-ZnCl}_2$ polyelectrolyte and nanocomposite. *Solid State Ionics* **176**(19-22), 1797-1800 (2005).
<https://doi.org/10.1016/j.ssi.2005.04.042>

- [S2] A. Turković, M. Pavlović, P. Dubček, M. Lučić-Lavčević, B. Etlinger, S. Bernstorff, SAXS/DSC study of polymer electrolyte for Zn rechargeable nanostructured galvanic cells. *J. Electrochem. Soc.* **154**(6), A554-A560 (2007). <https://doi.org/10.1149/1.2724440>
- [S3] L. A. Mary, T. Senthilram, S. Suganya, L. Nagarajan, J. Venugopal, S. Ramakrishna, V. Giri Dev, Centrifugal spun ultrafine fibrous web as a potential drug delivery vehicle. *Express Polym. Lett.* **7**(3), 238-248 (2013). <https://doi.org/10.3144/expresspolymlett.2013.22>
- [S4] S.-W. Kuo, C.-H. Wu, F.-C. Chang, Thermal properties, interactions, morphologies, and conductivity behavior in blends of poly (vinylpyridine)s and zinc perchlorate. *Macromolecules* **37**(1), 192-200 (2004). <https://doi.org/10.1021/ma035655+>
- [S5] L.P. Wang, P.F. Wang, T.S. Wang, Y.X. Yin, Y.G. Guo, C.R. Wang, Prussian blue nanocubes as cathode materials for aqueous Na-Zn hybrid batteries. *J. Power Sources* **355**, 18-22 (2017). <https://doi.org/10.1016/j.jpowsour.2017.04.049>