

## **Novel zwitterionic Polymer Binder with Enhanced Ionic Conductivity for Water-processable LiFePO<sub>4</sub> Cathodes**

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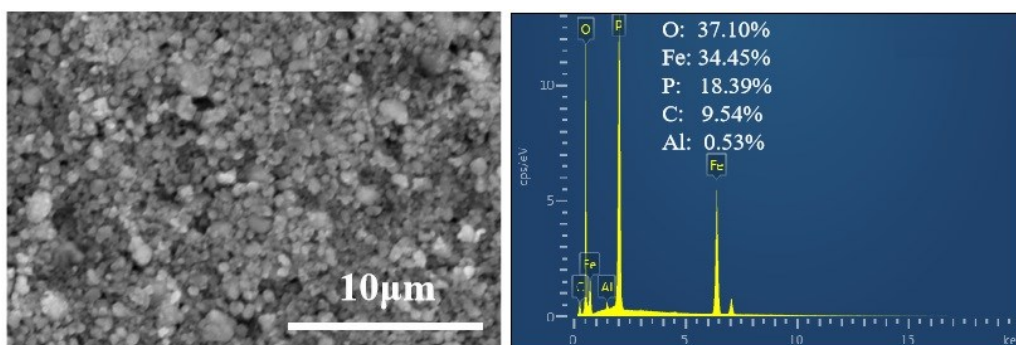
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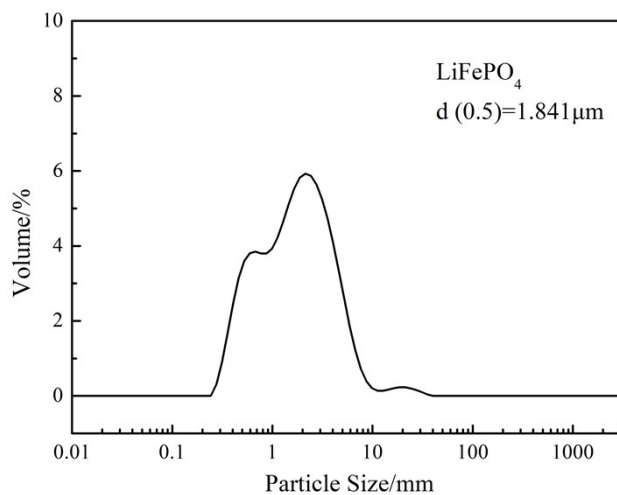
### **Experimental Section**

Energy Dispersive Spectrometer (EDS) of the electrode is characterized by X-Max (Oxford, England). The granulometry analysis is characterized by Mastersizer 3000 (Malvern, England). The rheological behavior is tested by digital viscometer (4#rotor, NDJ-5S, Shanghai, China). The particle size of the lithiated zwitterionic ionomer (LZI) binder is characterized by Nano ZS90 (Malvern, England). Thermogravimetric analysis (TGA) of pure binder films is performed with a TGA instrument (DSC 200F3, Germany). The differential scanning calorimetry (DSC) measurement is conducted using a TA Instrument (Q2, USA). The <sup>1</sup>H NMR spectrum of the LZI was characterized using a Bruker AVANCE III 400 Nuclear magnetic resonance spectrometer (Switzerland) with DMF as the solvent. Linear sweep voltammogram

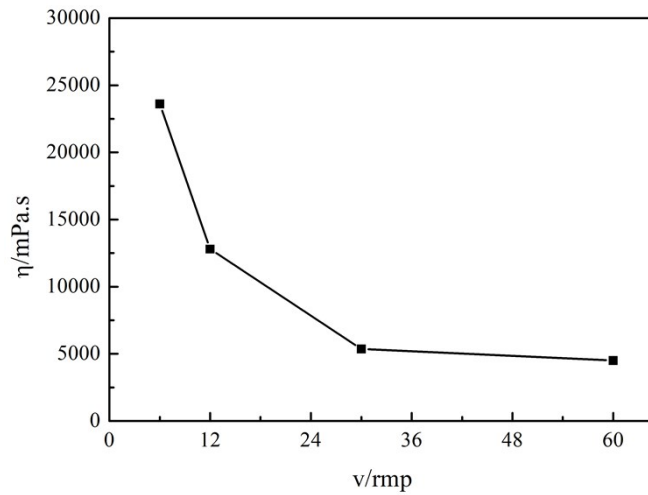
(LSV) of the LZI binder film is recorded on the SS|PSBA-Li|Li cell using a Solartron 1470 electrochemical work station from 0 V towards 6V with a scan rate of 2 mV s<sup>-1</sup>. The stainless steel (SS) acts as the working electrode, while Li foil as both the counter and reference electrodes. Peel strength test is with a tension tester (AG-X, Shimadzu, Japan).



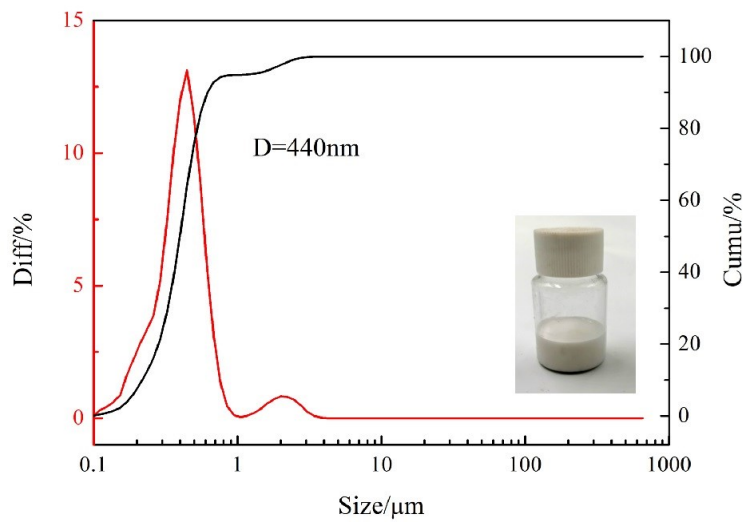
**Figure S1.** Energy Dispersive Spectrometer (EDS) test of LiFePO<sub>4</sub> polycrystalline.



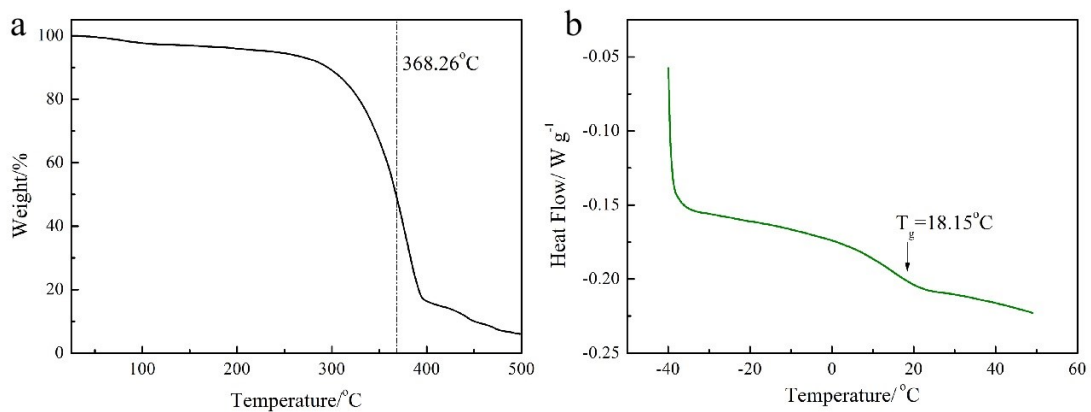
**Figure S2.** The granulometry of LiFePO<sub>4</sub> polycrystalline used in this paper.



**Figure S3.** The rheological behavior curve of the slurry with 1.5 wt.% LZI.

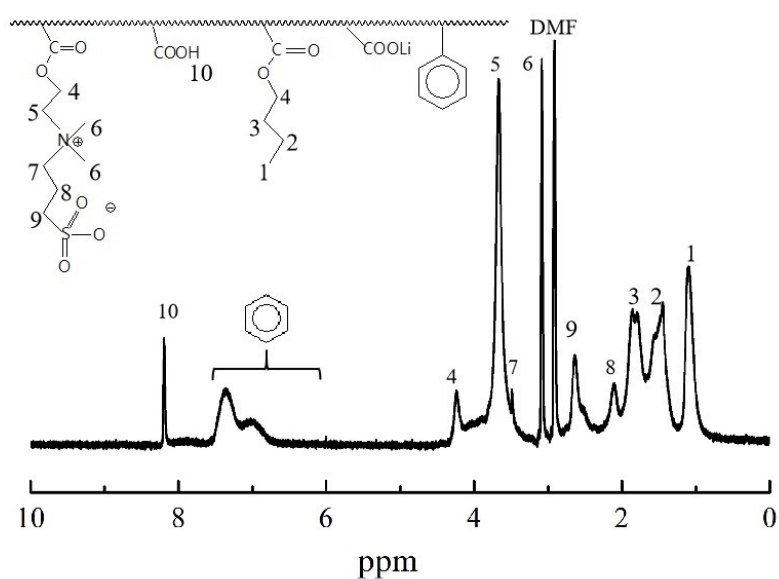


**Figure S4.** Emulsion (a) and particle size (b) of LZI binder.



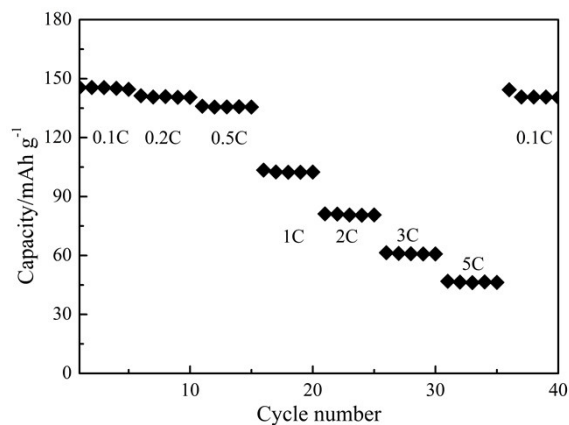
**Figure S5.** Thermal properties of LZI. (a) The TGA curve of LZG films in air; (b) The second DSC heating curve of LZI. The  $T_g$  of the LZI is 18.15°C. The results show that LZG is thermally

stable up to 368.26 °C in air atmosphere. Thereafter, the LZG presents a rapid mass loss due to the thermal decomposition of ester groups and reaction with oxygen which form molecule chains. Remains are most likely lithium groups [1]. These results explained sufficiently that the ionomer binder is thermal stable below 300°C in air atmosphere, which are superior during fabrication process such as pre-dry and vacuum [2].

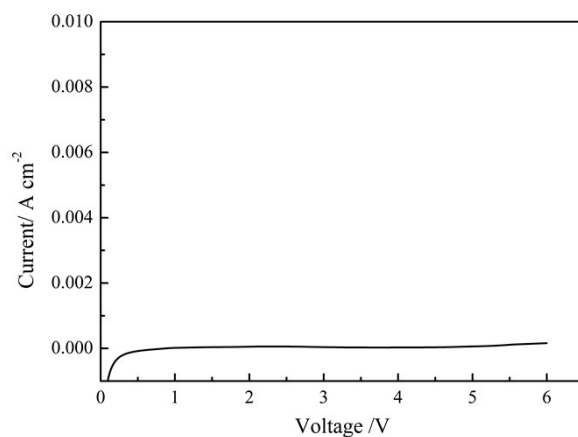


**Figure S6.** The <sup>1</sup>H NMR spectrum of the LZI. The <sup>1</sup>H NMR spectrum shows that characteristic peaks at 8.2 ppm is assigned as the chemical shifts of the H10 in the AA segment. The peaks located at 6.0-7.3 ppm belong to the chemical shifts of CH proton in the main chain of the benzene ring. The peaks at 2.1, 2.7, 3.1, 3.5, 3.7 and 4.2 ppm can be ascribed to H4-H9 in the chain of the DMAPS segments, respectively. The chemical shifts occurred at 4.2 ppm and 1.1-1.9 ppm are assigned as the chemical shifts of the CH<sub>3</sub> and CH<sub>2</sub> which attached to H1-H4, respectively, in the

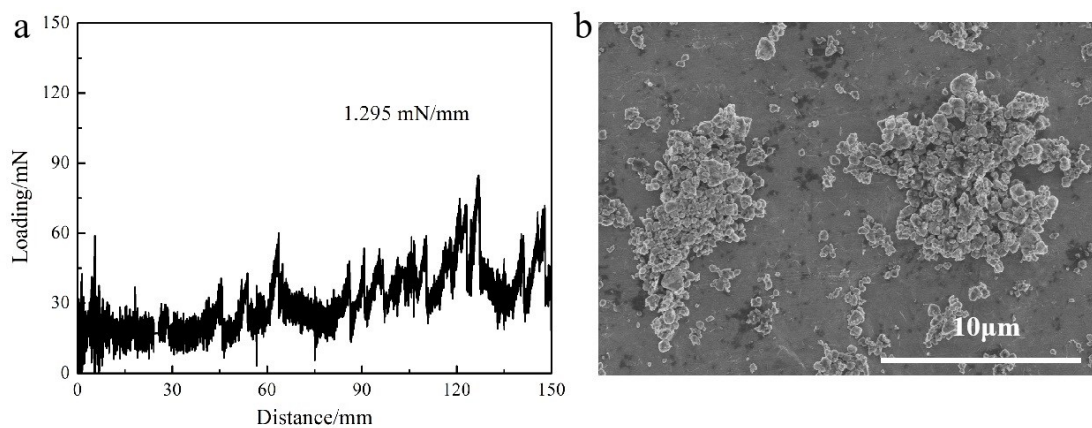
BA.



**Figure S7.** Rate performances of  $\text{LiFePO}_4$  cathodes with the LZI binder. The capacities of the LZI cathode maintain relatively stable from 0.1 C to 0.5 C. Then capacities decay obviously at the rate from 0.5C to 5C. The result shows that the zwitterion structure could not make a function of enhancing the diffusion of  $\text{Li}^+$  at higher rates apparently, which should be studied for further use.



**Figure S8.** Linear sweep voltammogram of the LZI binder film. The LSV measurement demonstrate the electrochemical stability of the LZG film.



**Figure S9.** (a) Peel strength test of electrodes prepared with the 1.5% LZG. The SEM image of

electrodes using LZG composite binder after peel strength test. The average peel strength investigated for LZG electrode is  $1.295\text{mN mm}^{-1}$ . SEM morphologies of LZG electrode indicated that plenty of  $\text{LiFePO}_4$  particles adhered to the Al foil. As a result, the LZG binder could provide enough adhesion to maintain the integrity of electrode.

## References

1. N. Loeffler, J. von Zamory, N. Laszczynski, I. Doberdo, G.T. Kim and S. Passerini, *J. Power Sources*, 2014, **248**, 915-922.
2. M. Baunach, S. Jaiser, S. Schmelzle, H. Nirschl, P. Scharfer and W. Schabel, *Drying Technology*, 2016, **34**, 462-473.