## **Supplementary Material**

# Improving cycling performance of LiMn<sub>2</sub>O<sub>4</sub> battery by adding an ester functionalized ionic liquid to electrolyte

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#### Viscosities and Conductivities of [MMEPyr][TFSI]

The temperature dependence of viscosity is quite important because it is necessary for fluid flow to occur as the internal volume of the electrode changes due to the electrode reaction. The temperature effect on viscosity of [MMEPyr][TFSI] was displayed in the range of 293.15 to 343.15 K shown in Fig. S1. As for other viscous liquids, the viscosity of [MMEPyr][TFSI] decreased linearly with increasing temperature and governed by H - bonding and van der Waals interactions, giving an excellent fit to the Arrhenius temperature relationship and following equation (1):

$$\ln \eta = \ln \eta_{\infty} + E \eta / RT \tag{1}$$

Where, R is universal gas constant and T is measurement temperature. Activation energy  $(E_n)$  is the energy barrier, which must be overcome for ions in motion in the

IL.<sup>[S1]</sup> The viscosity at infinite temperature  $(\eta_{\infty})$  and  $E_{\eta}$  can be calculated from the slope and intercept of the Arrhenius plot, respectively. The values of activation energy and  $\eta_{\infty}$  of [MMEPyr][TFSI] shown in Fig. S1 were 25.465 kJ mol<sup>-1</sup> and 1.135 mPa·s, respectively. Compared with our previous work, [MMEPyr][TFSI] has lower  $E_{\eta}$  than that of [N<sub>2224</sub>][TFSI], which indicated that the easier it is for the ions to move past each other in the [MMEPyr][TFSI].<sup>[S2]</sup>

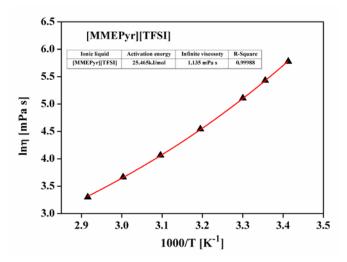
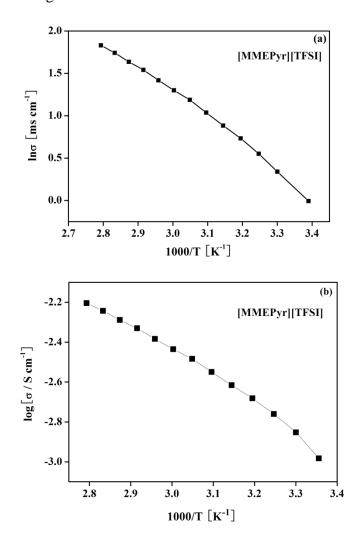


Fig. S1. Temperature dependence of viscosity of [MMEPyr][TFSI].

The ionic conductivity of IL has been regarded as a significant electrochemical property for electrolyte on the lithium-ion battery, which could be governed by its viscosity, density and formula weight. [S3,S4] The relationship between ionic conductivities of [MMEPyr][TFSI] and temperature in the range of 293.15 to 358.15 K was depicted in Fig. S2. As general trend, temperature dependence of the conductivity showed an inverse trend with that of viscosity. Elevating temperature increased the electrolyte conductivity with growing tendencies for [MMEPyr][TFSI] ionic liquid. In order to compare with the earlier data, the relationship between logσ and T was investigated, which was shown in Fig.S2b. It was found that there was a

litter lower than the results reported. It could be related to the different measurement instruments, the testing environment and so on.

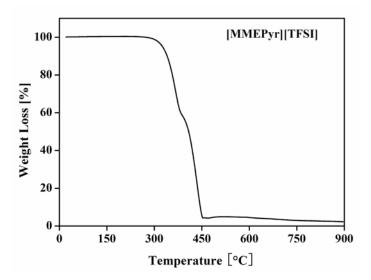


**Fig. S2.** Temperature dependence of ionic conductivity of [MMEPyr][TFSI].

### Thermal stability of [MMEPyr][TFSI]

The thermal stability of ILs strongly depends on their structure. The thermal stability of the [MMEPyr][TFSI] under nitrogen was determined by TGA shown in Fig. S3. The [MMEPyr][TFSI] exhibited excellent thermal stability which was stable at up to 300 °C. As the temperature was further increased, it rapidly decomposed between 333 and 450 °C with 100 % weight loss in two-stage decomposition process. The first decomposition stage from 333 to 372 °C was possibly to be associated with

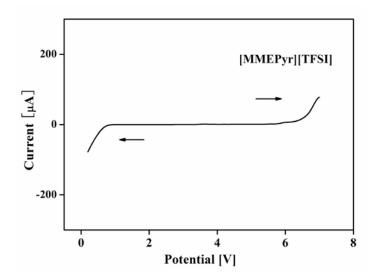
the loss of methyl acetate group. [S5] The second decomposition occurred above 375 °C, which was associated with the literature. [S5] The TGA result validated that the [MMEPyr][TFSI] had the high purity and was suitable to electrolyte of lithium-ion battery with a high thermal stability.



**Fig. S3.** The TGA curve of [MMEPyr][TFSI] at a heating rate of 10 °C min<sup>-1</sup> from 25 to 900 °C.

### **Electrochemical window of [MMEPyr][TFSI]**

Knowledge of electrochemical stability is important for assessing the suitability of ILs for electrolyte, which was considered the aspects of lithium-ion battery performance as output voltage and the charge-discharge cycling property. The electrochemical windows of [MMEPyr][TFSI] was carried out by LSV using lithium metal as reference electrode shown in Fig. S4. The cathodic limiting potential of the [MMEPyr][TFSI] was about 0.80 V versus Li/Li<sup>+</sup> and the anodic limiting potential of the IL was about 6.20 V versus Li/Li<sup>+</sup>, so its electrochemical window was about 5.40 V, which indicated that the [MMEPyr][TFSI] had a high electrochemical stability. It was a litter higher than the result in the reference, which was due to using different



**Fig. S4.** Linear sweep voltammogram of [MMEPyr][TFSI] at 25 °C with glassy carbon as working electrode, metal Li as both counter electrode and reference electrode. Scan rate: 5 mV s<sup>-1</sup>.

### References

[S1] O. O. Okoturo, T. J. VanderNoot, J. Electroanal. Chem. 2004, 568, 167.

[S2] Z. N. Wang, Y. J. Cai, T. Dong, S. M. Chen, X. M. Lu, *Ionics* 2013, 19, 887.

[S3] A. Kanazawa, O. Tsutsumi, T. Ikeda, Y. Nagase, J. Am. Chem. Soc. 1997, 119, 7670.

[S4] S. H. Fang, Y. Jin, L. Yang, S. I. Hirano, K. Tachibana, S. Katayama, *Electrochim*.
Acta 2011, 56, 4663.

[S5] J. S. Lee, N. D. Quan, J. M. Hwang, J. Y. Bae, H. Kim, B. W. Cho, H. S. Kim, H. Lee, *Electrochem. Commun.* 2006, 8, 460.