Electronic Supplementary Information

Synergistic effect of organic plasticizer and lepidolite filler on polymer electrolytes for all-solid high-voltage Li-metal batteries

Bo Wang, Yanchao Wu, Shuming Zhuo, Shaolong Zhu, Yuan Chen, Cheng Jiang, and Chengliang Wang*

School of Optical and Electronic Information, Wuhan National Laboratory for Optoelectronics (WNLO), Huazhong University of Science and Technology, Wuhan 430074, China. E-mail: clwang@hust.edu.cn

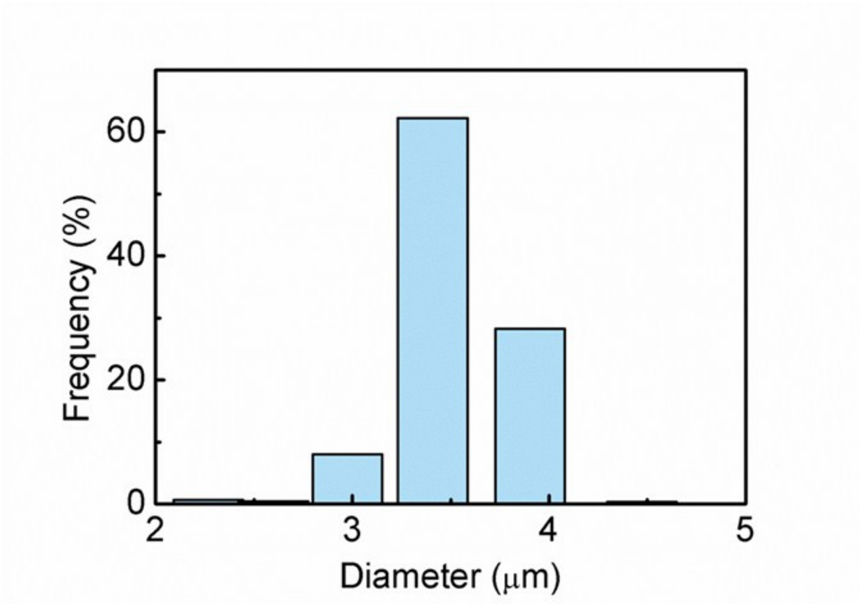

Fig. S1. Size distribution of lepidolites sheets for constructing CPEs.

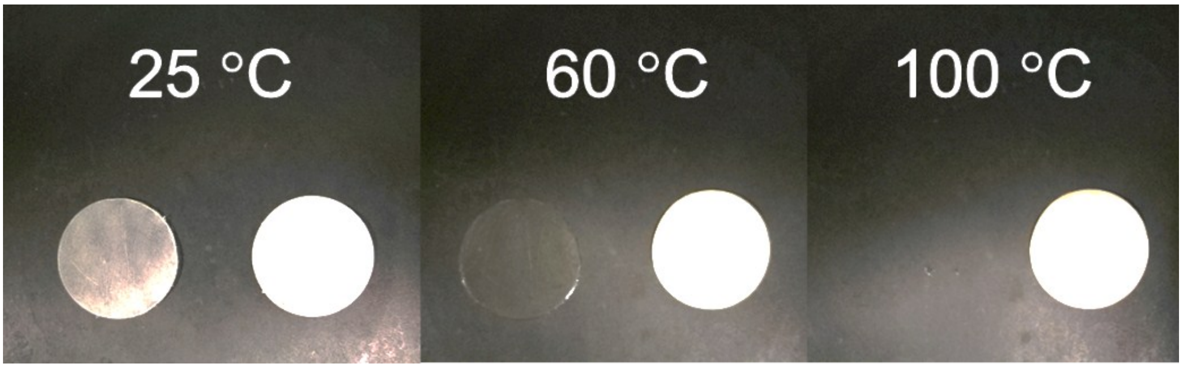

Fig. S2. Heat resistance of lepidolite-PEO-$LiClO_4$ (left) and lepidolite-PVDF-$LiClO_4$ (right) composites with lepidlite contents of 6%. It showed that the PEO-based CPEs started to soften and wrinkle at a temperature higher than 60 °C. The film was melting at a temperature at 100 °C. On the other hand, the PVDF-based CPEs kept almost intact even at a temperature higher than 100 °C.

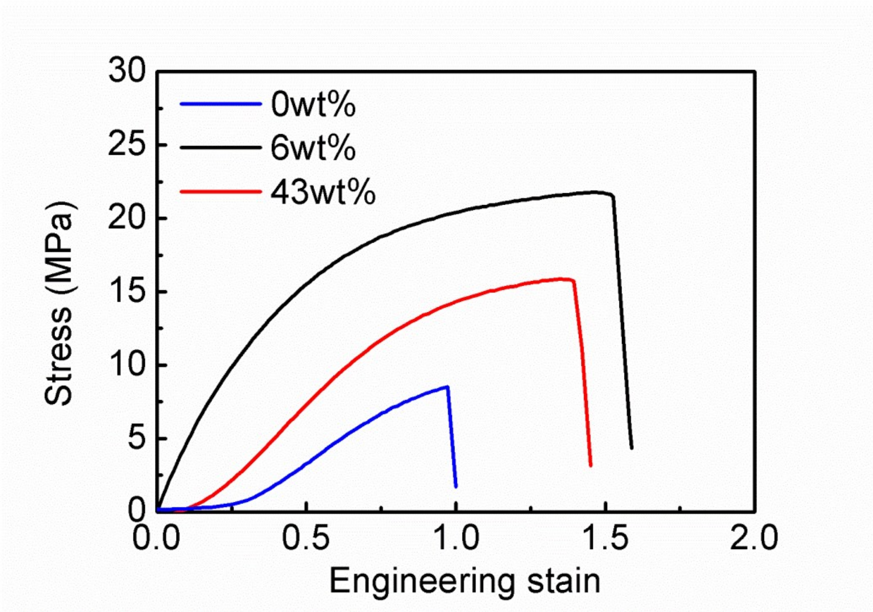

Fig. S3. Tensile stress/strain curve for lepidolite-PVDF-$LiClO_4$ composites sample. Without addition of lepidolite, the tensile strength of PVDF-$LiClO_4$ matrix is only 7.5 MPa. After addition of lepidolite, the tensile strength increased to 22 and 15 MPa for the CPEs with lepidolite content of 6 wt% and 43 wt%, respectively.

Table S1. Relationship between dried temperature and DMF contents.

Dried temperature (°C)	DMF contents (%) in CPEs with lepidolite content of 6%	DMF contents (%) in CPEs without lepidolite
80	3.9	3
100	0.9	1
120	0.3	0.3

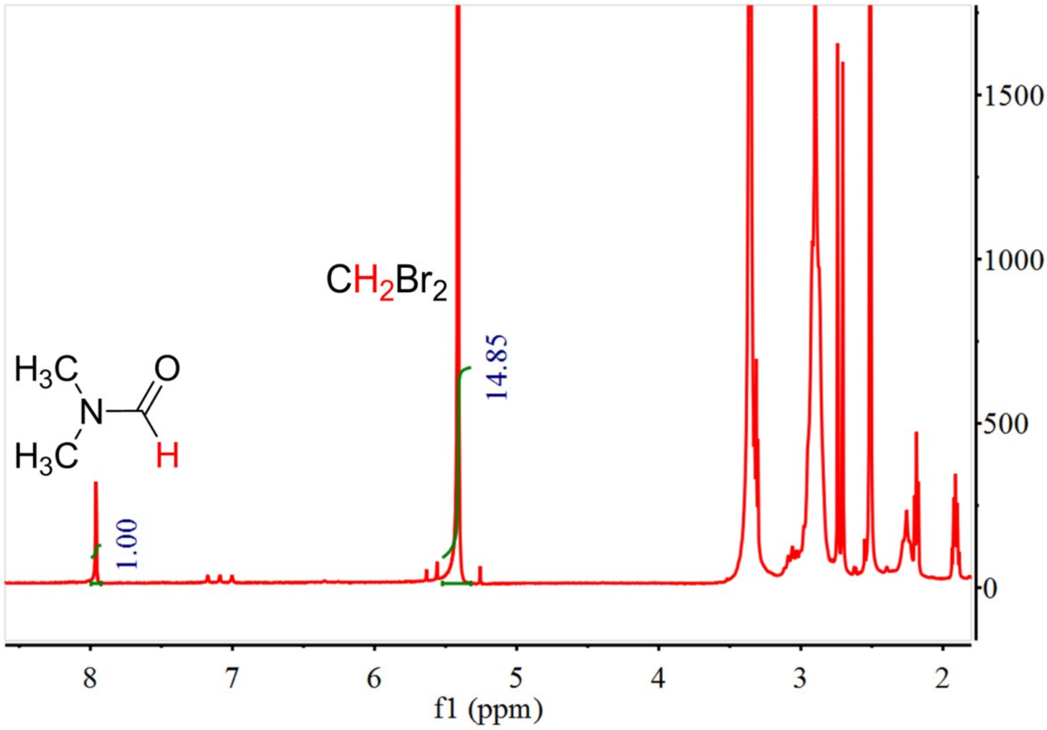

Fig. S4. [1]H NMR spectra of CPEs with lepidolite content of 6% dried at 80 °C. The solvent was DMSO. CH_2Br_2 was added as calibration agent. The content of DMF was therefore calculated accordingly.

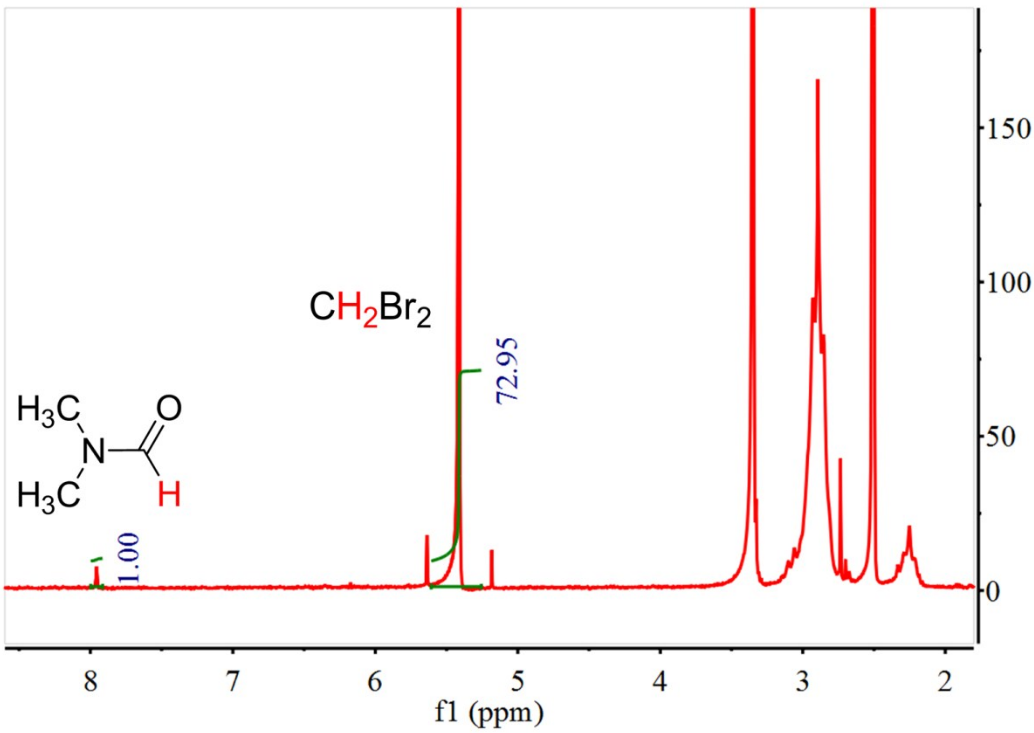

Fig. S5. ^{1}H NMR spectra of CPEs with lepidolite content of 6% dried at 100 °C. The solvent was DMSO. CH_2Br_2 was added as calibration agent. The content of DMF was therefore calculated accordingly.

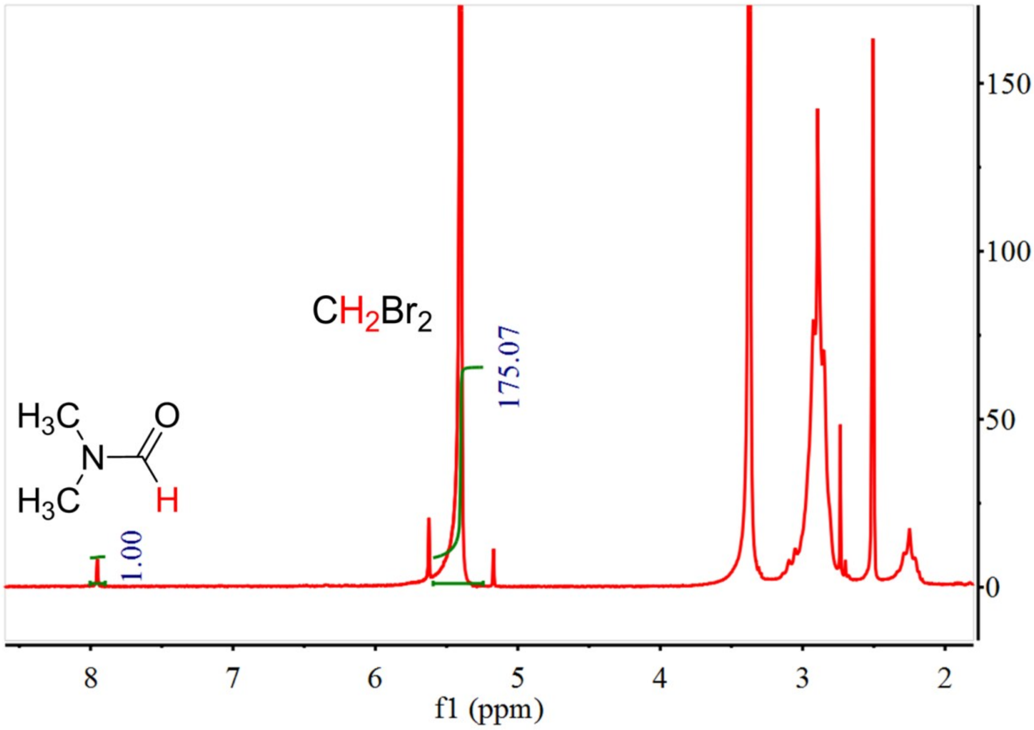

Fig. S6. [1]H NMR spectra of CPEs with lepidolite content of 6% dried at 120 °C. The solvent was DMSO. CH_2Br_2 was added as calibration agent. The content of DMF was therefore calculated accordingly.

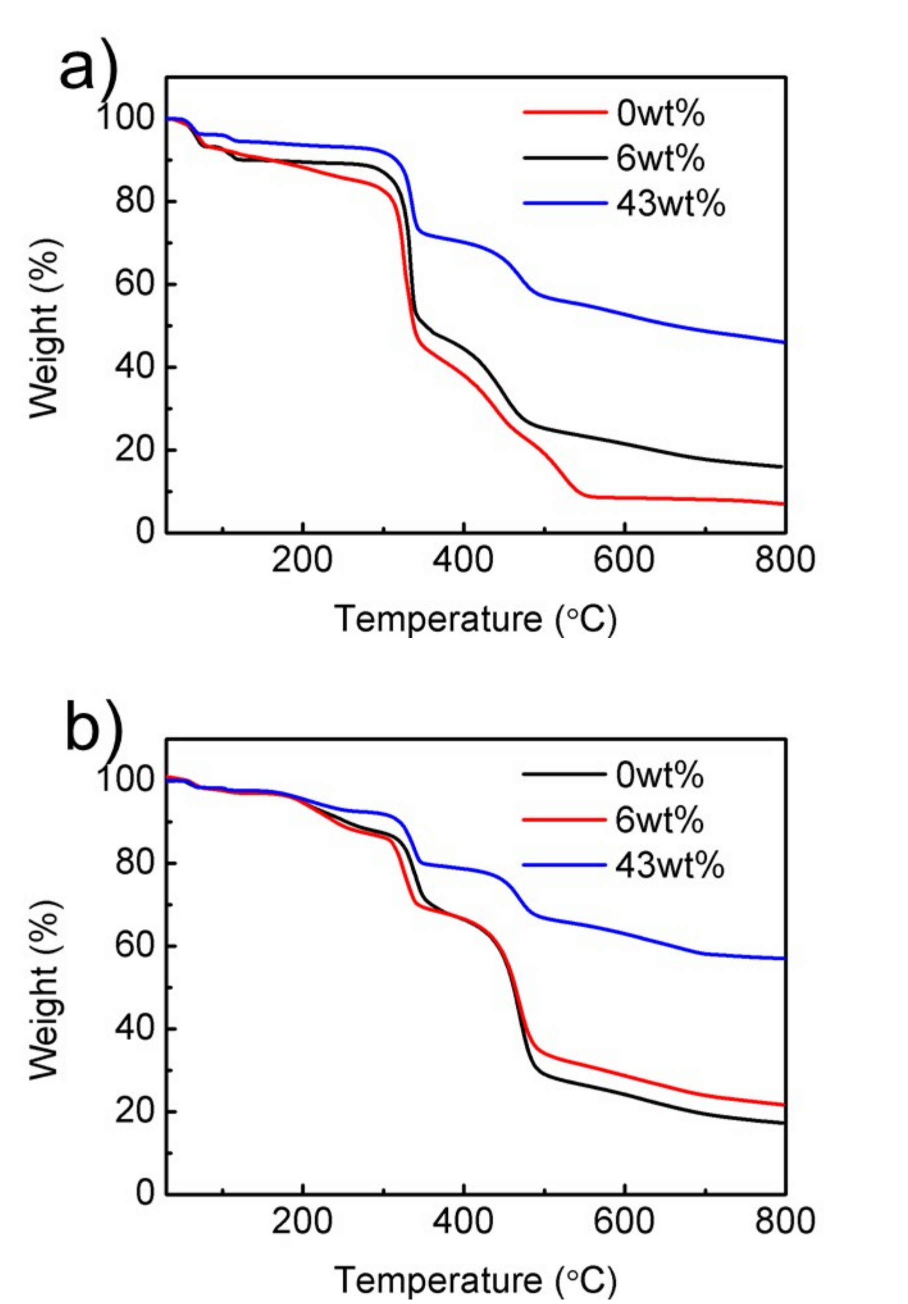

Fig. S7. TGA curves of PVDF-$LiClO_4$-lepidolite CPEs after dried at a) 80 and b) 100 °C with different ratio. The weight loss at temperature of about 80 °C should be contributed to the adsorbed water in ambient air. The weight loss at temperature of about 120 °C should be contributed to the DMF solvent. The content of DMF is about 5% and 1% for the samples after dried at 80 and 100 °C, respectively, which are coincident with the NMR measurements (3.9% and 0.9% respectively). It is clear that the content of DMF decreased after bing dried at higher temperatures.

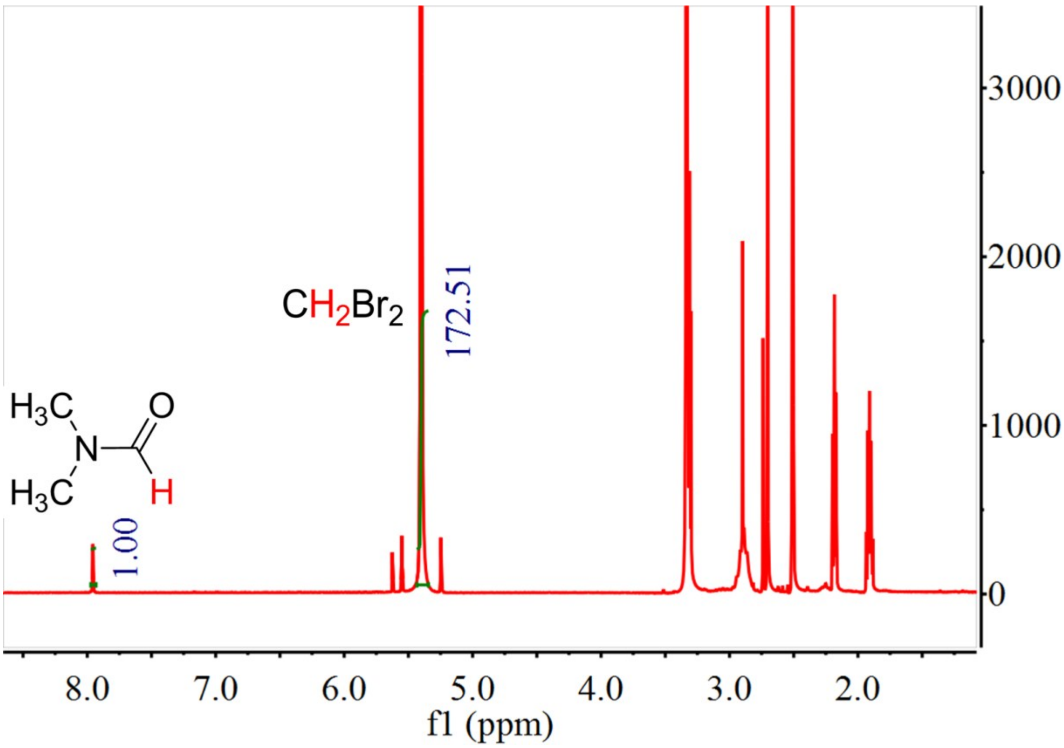

Fig. S8. ^1H NMR spectra of CPEs without lepidolite dried at 80 °C. The solvent was DMSO. CH_2Br_2 was added as calibration agent. The content of DMF was therefore calculated accordingly.

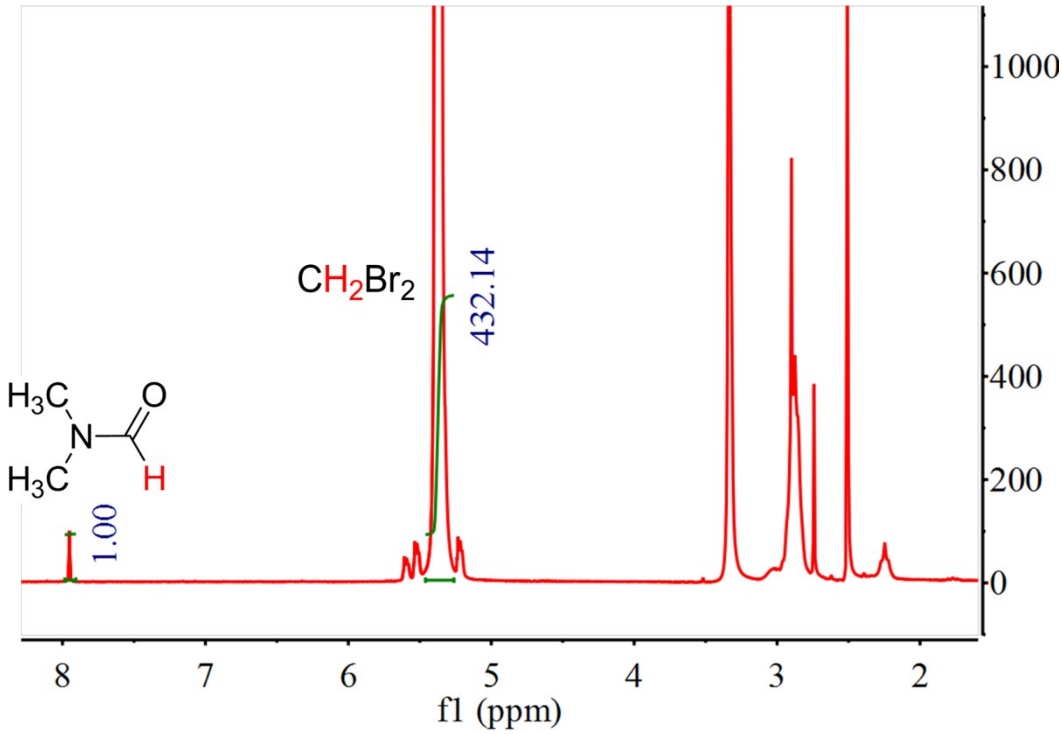

Fig. S9. ^{1}H NMR spectra of CPEs without lepidolite dried at 100 °C. The solvent was DMSO. CH_2Br_2 was added as calibration agent. The content of DMF was therefore calculated accordingly.

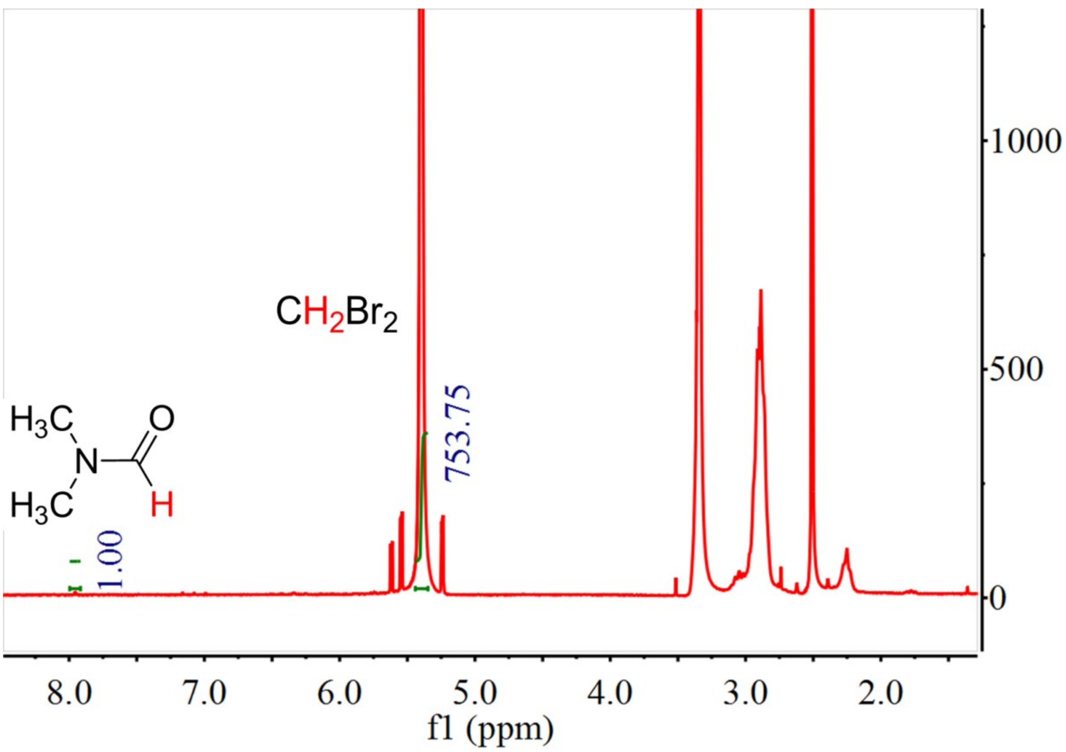

Fig. S10. [1]H NMR spectra of CPEs without lepidolite dried at 120 °C. The solvent was DMSO. CH_2Br_2 was added as calibration agent. The content of DMF was therefore calculated accordingly.

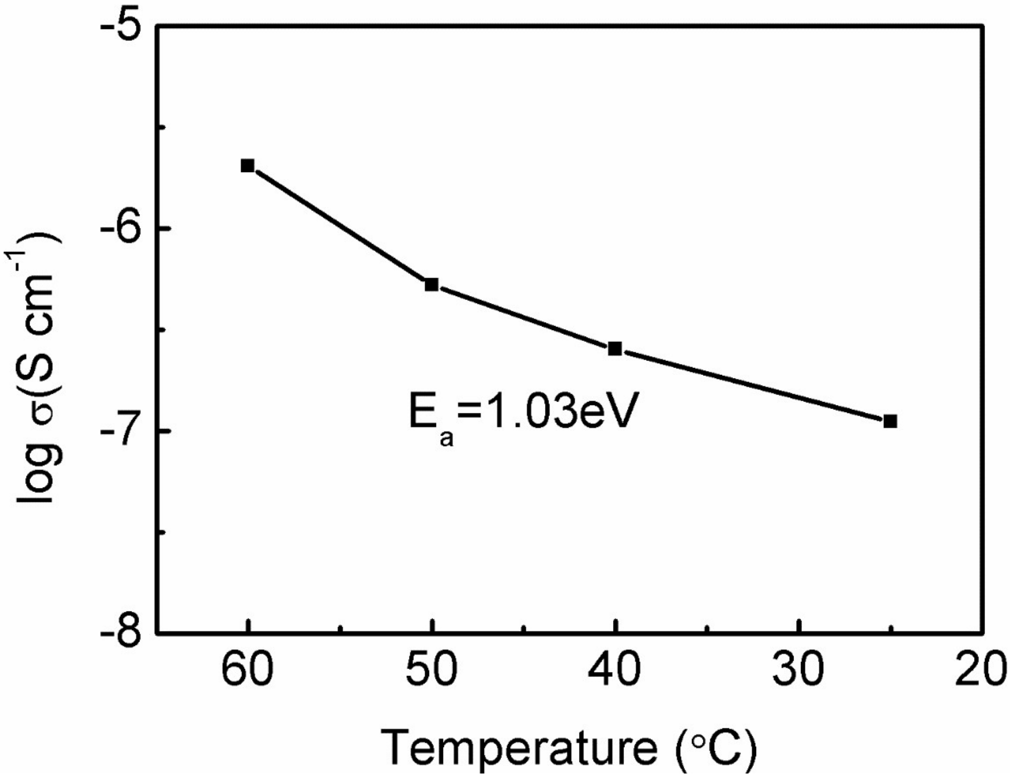

Fig. S11. Temperature dependence of the Li-ion conductivity of PVDF-based CPEs without lepidolite.

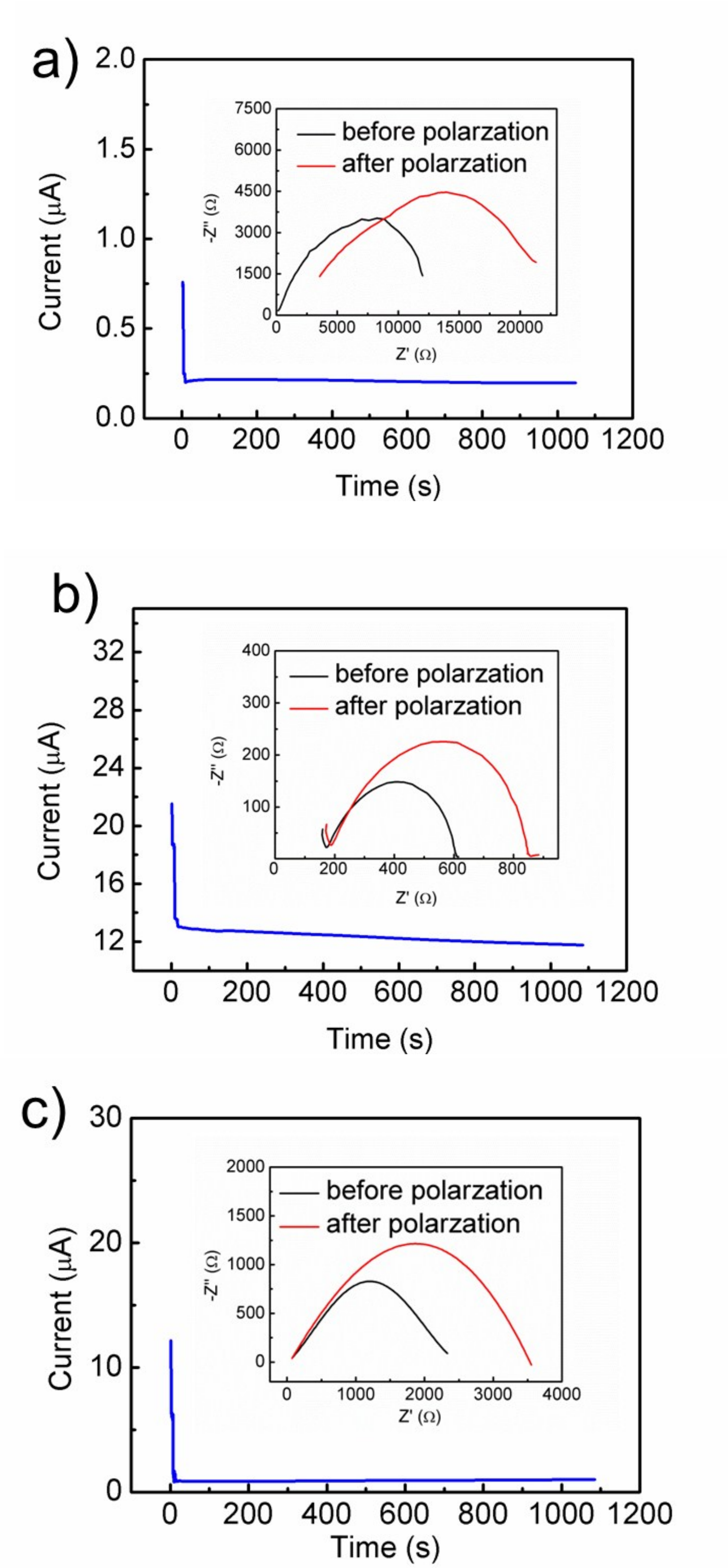

Fig. S12. a) Current−time profile of a symmetrical Li/PVDF−$LiClO_4$/Li cells after applying a dc voltage on the cell. b) Current−time profile of a symmetrical Li/lepidolite−PVDF−$LiClO_4$/Li cells with lepidolite content of 6% after applying a dc voltage on the cell. c) Current−time profile of a symmetrical Li/lepidolite−PEO−$LiClO_4$/Li cell with lepidolite content of 6% after applying a dc voltage on the cell. The curves were used for determining Li^+ transference number. The insets of (a), (b) and (c) show the Nyquist plots of the corresponding cells before and after polarization.

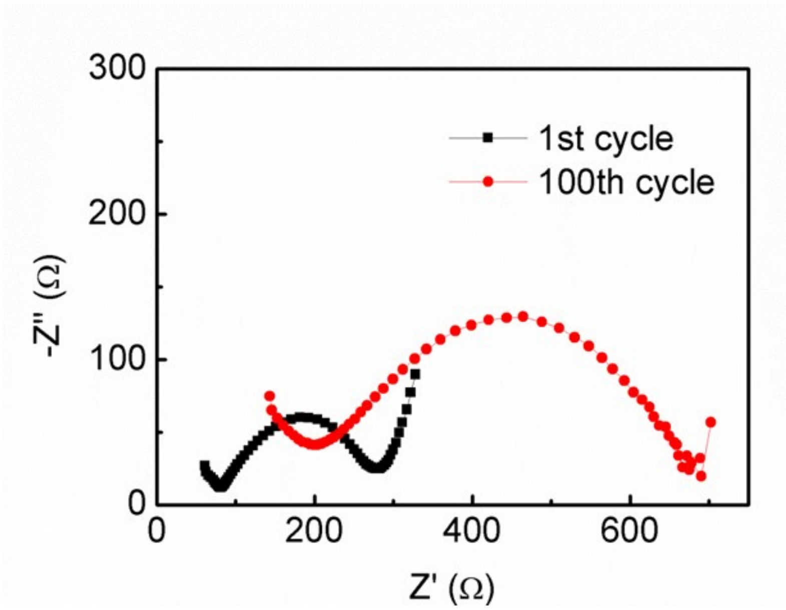

Fig. S13. Impedance spectra of $LiFePO_4$/CPEs/Li battery.

Fig. S14. LEDs lighted by a all-solid Li-metal battery cell.