

Supporting Information

Table 1. Crystal structure of the LiFeSO_4F material obtained by Rietveld refinement.

Phase I name: LiFeSO_4F ; Space Group: P-1; fraction: 95.7 %					
Atom	Wyckoff position	x	y	z	occupancy
Fe1	1a	0.0	0.0	0.0	1
Fe2	1b	0.0	0.0	0.5	1
S	2i	0.300(0)	0.625(4)	0.246(3)	1
O1	2i	0.569(0)	0.739(8)	0.384(3)	1
O2	2i	0.081(4)	0.643(4)	0.362(8)	1
O3	2i	0.347(1)	0.354(2)	0.160(1)	1
O4	2i	0.271(3)	0.782(0)	0.110(8)	1
F	2i	0.119(6)	0.886 (5)	0.757(5)	1
a= 5.1841(4) b= 5.5058(1) c= 7.2286(7) $\alpha= 106.535(1) \beta= 107.201(7) \gamma= 97.775(3)$					
Phase II name: LiF ; Space Group: Fm-3m; fraction: 4.3 %					
Li	4a	0.0	0.0	0.0	1
F	4b	0.5	0.5	0.5	1
a= b= c= 4.0328(1) $\alpha= \beta= \gamma= 90.0$					
$R_p= 9.12, R_{wp}= 10.7, R_e=3.11$					

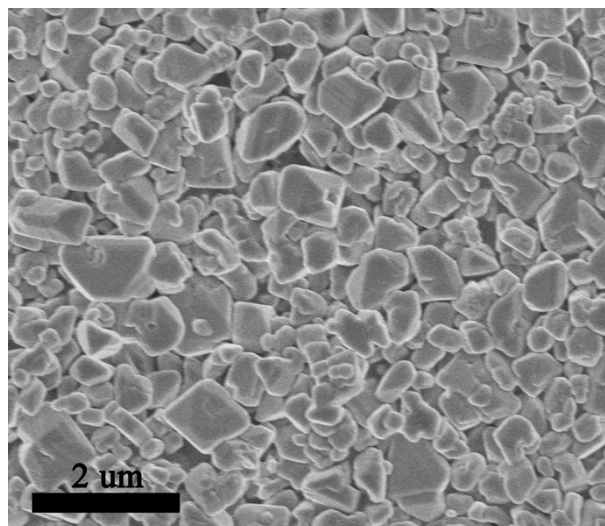


Fig. S1. SEM image of the LiFeSO_4F sample.

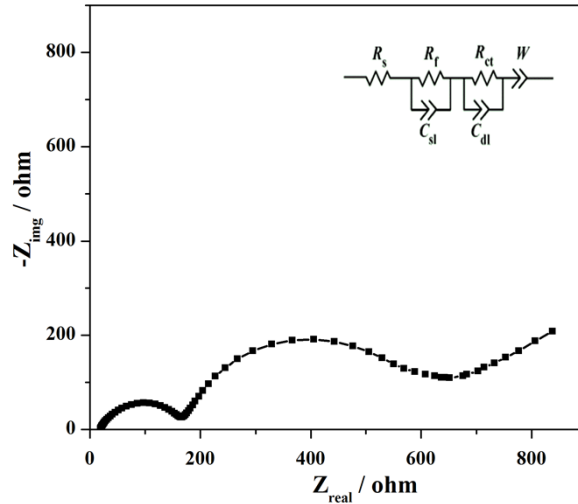


Fig. S2. Electrochemical impedance spectroscopy of the LiFeSO_4F sample at the charged state of 3.6 V. The semicircle in the high-to-medium frequency region is attributed to the solid electrolyte interface (SEI) film. The semicircle in the medium-to-low frequency region is due to the charge transfer process.

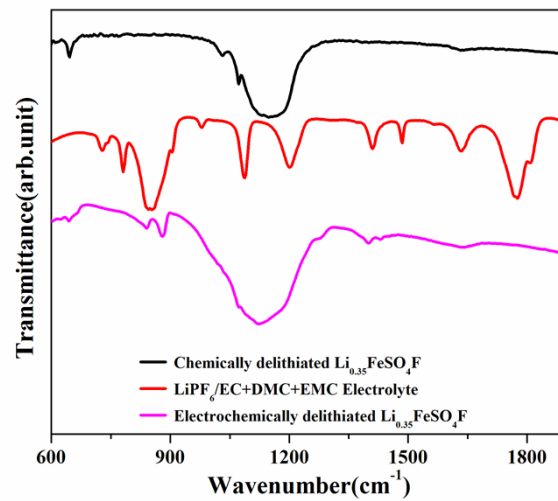


Fig. S3. FTIR curves of the electrolyte, together with those of the chemically and electrochemically de-lithiated $\text{Li}_{0.35}\text{FeSO}_4\text{F}$. The electrochemically de-lithiated sample shows some additional peaks at 840, 879, 1400 and 1633 cm^{-1} , which are due to the remaining electrolyte.

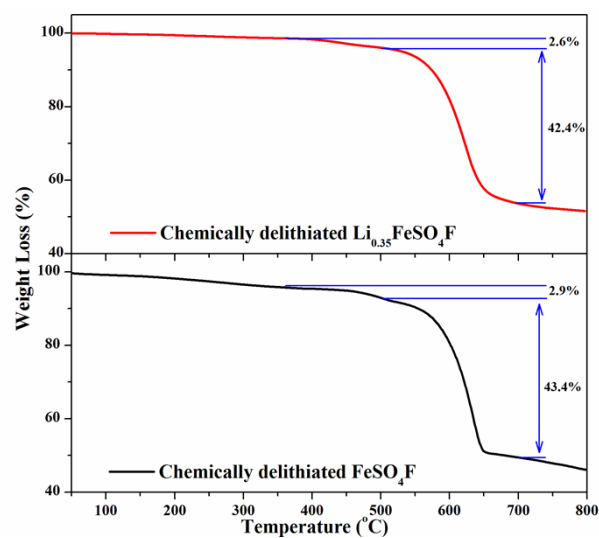


Fig. S4. TG curves of the chemically de-lithiated $\text{Li}_{0.35}\text{FeSO}_4\text{F}$ (up) and FeSO_4F (down) samples.

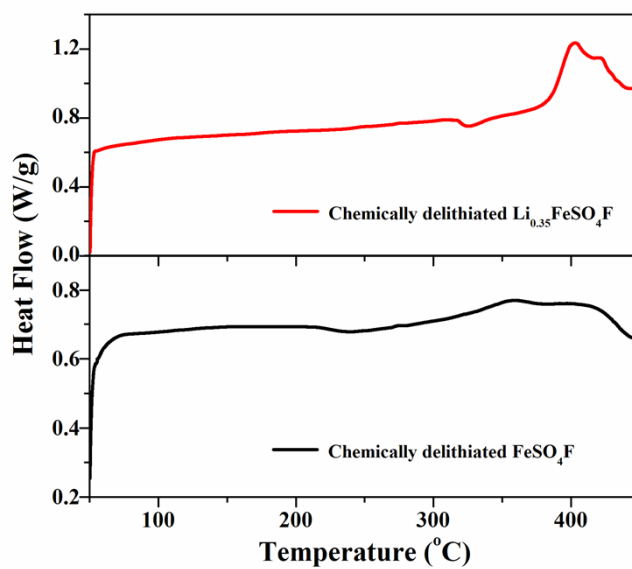


Fig. S5. DSC curves of the chemically de-lithiated $\text{Li}_{0.35}\text{FeSO}_4\text{F}$ (up) and FeSO_4F (down) samples.

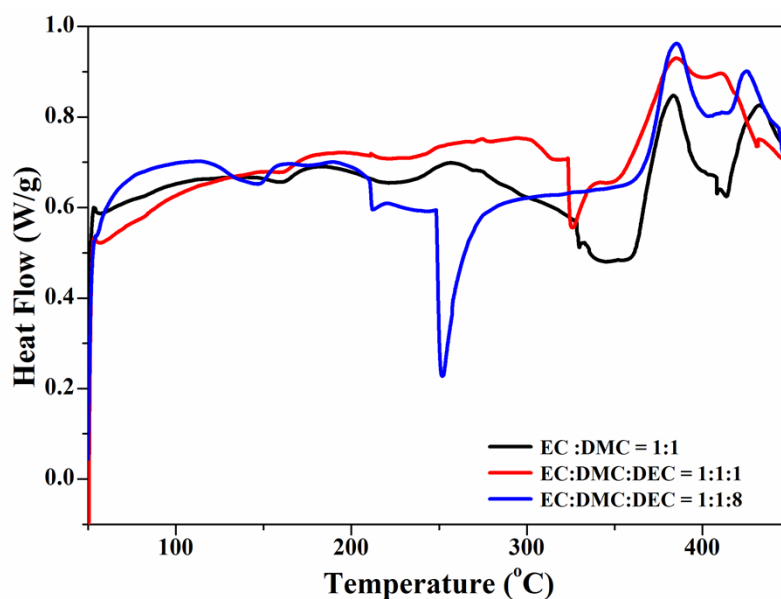


Fig. S6. DSC curves of the electrochemically de-lithiated $\text{Li}_{0.35}\text{FeSO}_4\text{F}$ using different LiPF_6 based electrolyte. The endothermic temperatures are different which is due to the use of different electrolyte solvents. But the exothermic processes of the samples lie in the same temperature range of 350-450 °C. In addition, the involved exothermic heat is similar for different electrolytes. This indicates that the material has good thermal stability in conventional LiPF_6 based electrolytes.

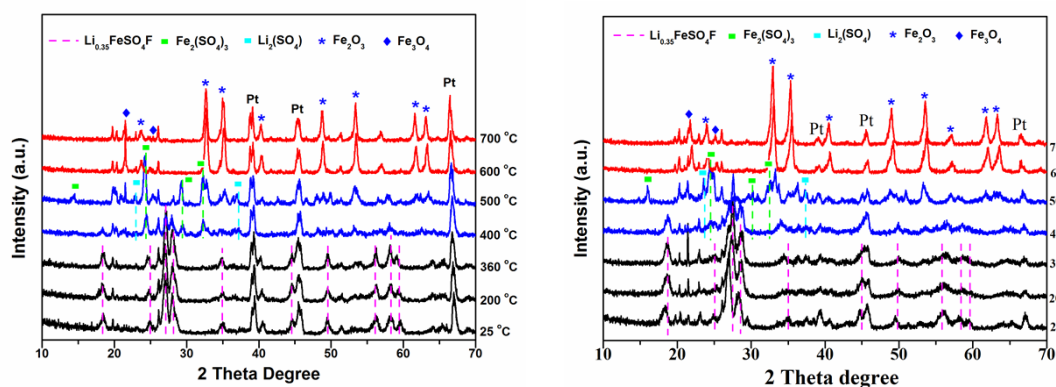


Fig. S7. Temperature dependent XRD patterns of the electrochemically (left) and chemically (right) de-lithiated $\text{Li}_{0.35}\text{FeSO}_4\text{F}$ samples. Both samples show similar XRD patterns indicating that the phase transformation is only correlated with the structure properties of $\text{Li}_{0.35}\text{FeSO}_4\text{F}$ itself.