

## **Supporting Information**

### **Ternary PEO/PVDF-HFP-based polymer electrolytes for Li-ion batteries**

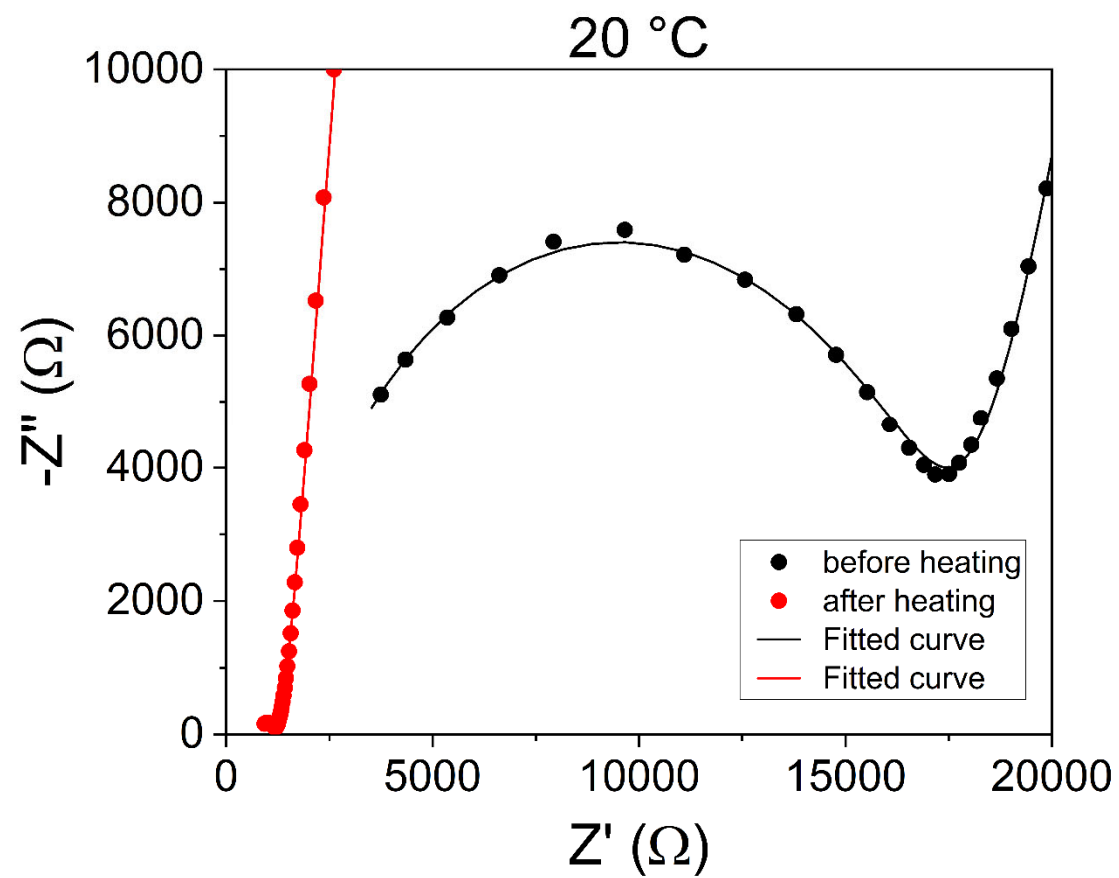
Hoang Bao Tran Nguyen<sup>1\*</sup>, Ling Ding<sup>1</sup>, Björn Pohle<sup>1</sup>, Toni Schmeida<sup>1</sup>, Hoang Bao An Nguyen<sup>1</sup>, Daria Mikhailova<sup>1\*</sup>

<sup>1</sup> *Leibniz Institute for Solid State and Materials Research (IFW) Dresden e.V., Helmholtzstraße 20, 01069 Dresden, Germany*

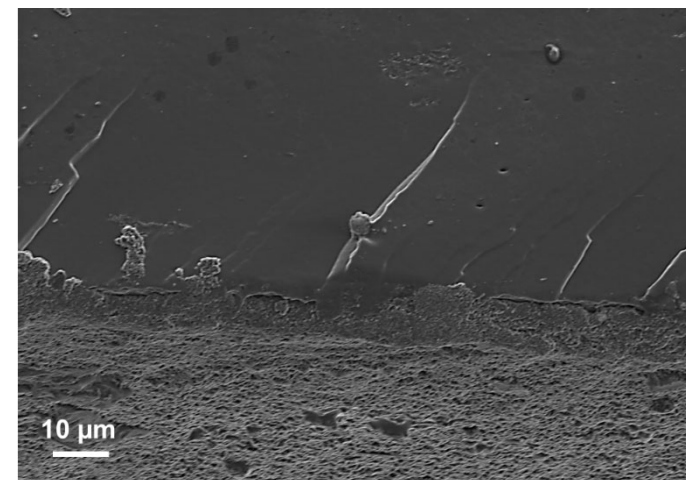
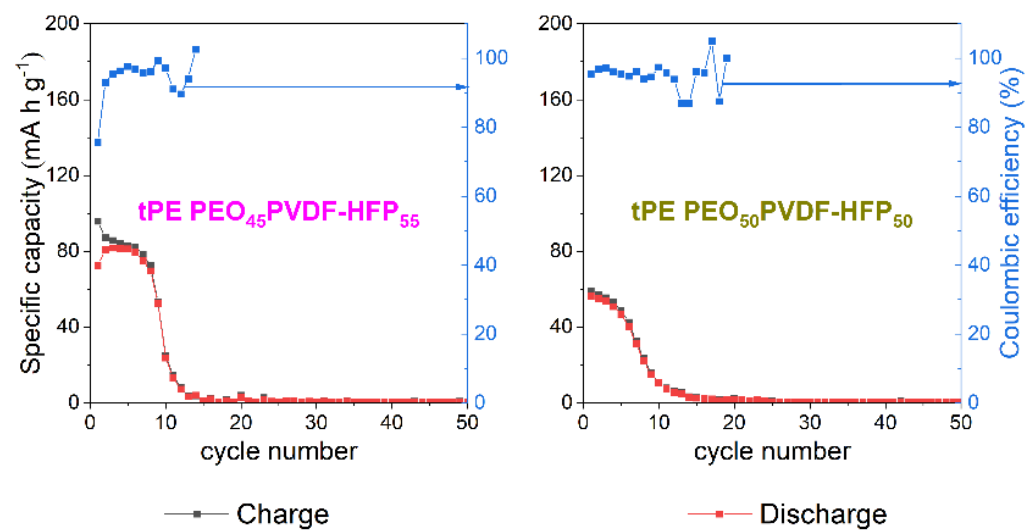
**Table S1.** Vibrations assigned in IR spectra.

Pure PVDF-HFP	Pure PEO	PEO/PVDF-HFP	Pure LiTFSI and pure EMITFSI	tPE PEO <sub>y</sub> PVDF-HFP <sub>z</sub> y : z						Vibration	Reference
				25:75	30:70	35:65	40:60	45:55	50:50		
Wavenumber [cm <sup>-1</sup> ]											
	2886	2882		2920	2922	2925	2927	2933	2927	C-H stretching vibration of PEO	[45]
	1359	1352		absent in ternary polymer electrolytes						CH <sub>2</sub> wagging vibration of PEO	[29,46]
	1342	1342		absent in ternary polymer electrolytes						CH <sub>2</sub> bending vibration of PEO	[29,46]
	1100	1097		absent in ternary polymer electrolytes						symmetric and asymmetric C-O-C stretching of PEO	[29,46]
	952	953		955	955	956	956	953	956	C-O stretching vibration of PEO	[45]
	841	842		838	838	838	837	836	836	CH <sub>2</sub> rocking vibration of PEO	[45,46]
3024, 2985				absent in ternary polymer electrolytes						asymmetric and symmetric C-H stretching vibration of PVDF-HFP	[43]
1400		1403		absent in ternary polymer electrolytes						deformation vibration of the C-H bond of PVDF-HFP	[16]
974		975		absent in ternary polymer electrolytes, at 972 weak peak in 45:55						crystalline phase (alpha phase) of PVDF-HFP, C-F stretching	[43]
795		795		absent in ternary polymer electrolytes						crystalline phase (alpha phase) of PVDF-HFP, CF <sub>3</sub> stretching vibration	[43]
762		762		absent in ternary polymer electrolytes						crystalline phase (alpha phase) of PVDF-HFP, CH <sub>2</sub> rocking vibration	[43]
613		613		absent in ternary polymer electrolytes						crystalline phase (alpha phase) of PVDF-HFP, mixed mode of CF <sub>2</sub> bending and CCC skeletal vibration	[43]
532		532		absent in ternary polymer electrolytes						crystalline phase (alpha phase) of PVDF-HFP, wagging vibrations of CF <sub>2</sub> group	[43]
871		872		879	879	879	879	879	879	amorphous phase (beta phase) of PVDF-HFP	[43]

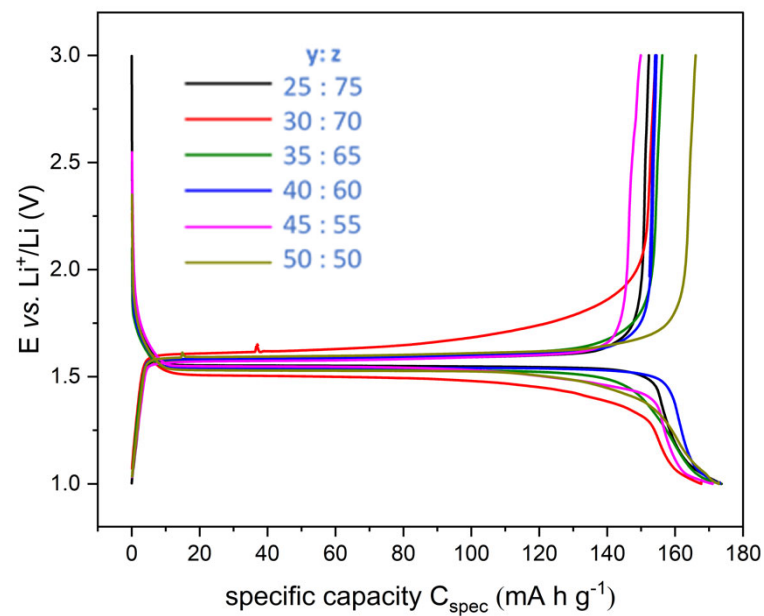
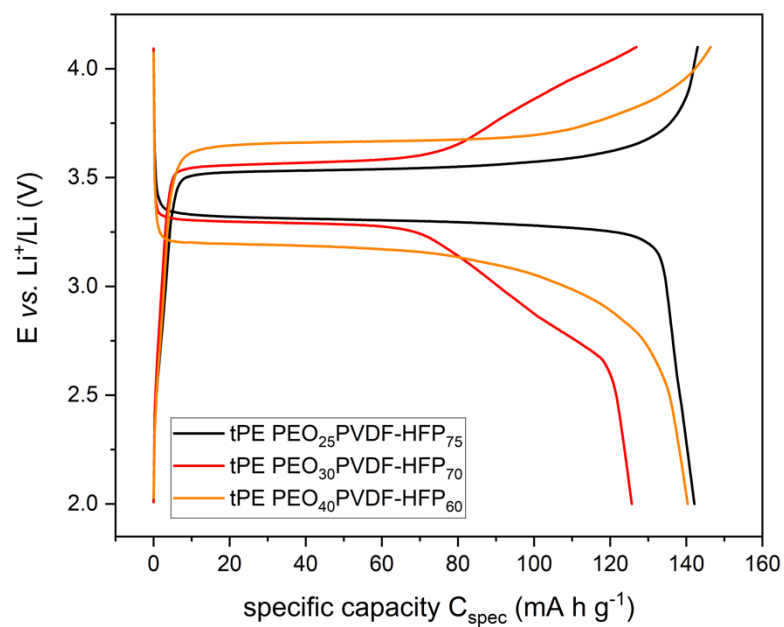
841				overlapping with CH <sub>2</sub> rocking vibration of PEO						amorphous phase (beta phase) of PVDF-HFP	[43]
			3200-3100							C-H stretching vibration of imidazolium ring	[43]
			1346	1346	1347	1347	1348	1352	1348	asymmetric SO <sub>2</sub> stretching vibration of LiTFSI and EMIMTFSI	[43]
			1326	1327	1327	1327	1328	1329	1329	C-SO <sub>2</sub> -N vibration of the LiTFSI and EMIMTFSI	[43]
			1193	1183	1183	1182	1182	1179	1181	symmetric stretching vibration of CF <sub>3</sub> group of LiTFSI and C-H vibration of imidazolium ring of IL EMIMTFSI	[43]
			1133	1131	1131	1131	1132	1132	1133	C-SO <sub>2</sub> -N vibration of LiTFSI and EMIMTFSI	[43]
			1051	1052	1052	1052	1052	1053	1052	asymmetric S-N-S stretching vibration of conducting salt and IL	[43]
			740	742	742	741	741	739	741	overlap of CF <sub>3</sub> bending vibration with the combination of C-S of the EMIMTFSI and S-N stretching vibration of LiTFSI	[43]
			600	600	600	600	600	599	600	asymmetric SO <sub>2</sub> bending vibration of LiTFSI and EMIMTFSI	[6]
			612	612	612	612	612	612	612	Ring op asymmetric bending vibration of EMIM <sup>+</sup>	[6]
			571	570	570	570	570	570	570	asymmetric bending vibration of CF <sub>3</sub> group of LiTFSI	[43]



**Figure S1.** Nyquist plots of tPE PEO<sub>35</sub>PVDF-HFP<sub>65</sub> at 20 °C recorded for materials before and after heating to 80 °C.



**Figure S2.** Left: Cycle stability of full cells LTO/tPE PEO<sub>y</sub>PVDF-HFP<sub>z</sub>/LFP with a high mass fraction of PEO at 0.1C at 25 °C. Right: SEM image of the sandwich LTO/tPE PEO<sub>50</sub>PVDF-HFP<sub>50</sub>/LFP after cycling, reflecting low mechanical integrity.



**Figure S3.** Charge-discharge curves of half cells LFP/ tPE PEO<sub>y</sub>PVDF-HFP<sub>z</sub>/Li (left) and LTO/ tPE PEO<sub>y</sub>PVDF-HFP<sub>z</sub>/Li (right) with various electrolyte compositions at 60 °C.

## Literature

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