

### Supporting Information

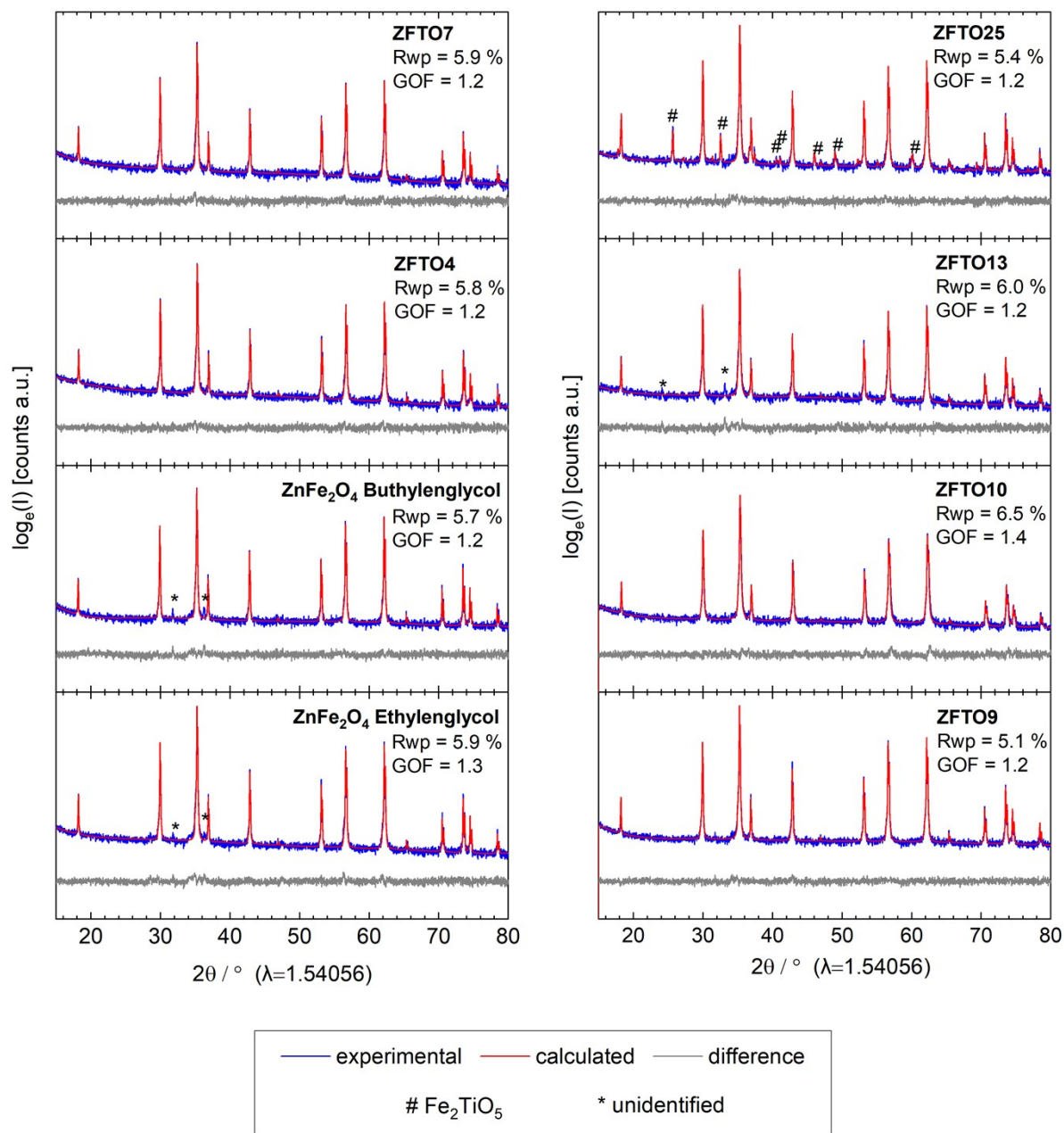


Figure SI 1: Overview of fits against PPXRD data based on the Rietveld method for all samples

Table SI 1: Overview on the results from Rietveld analysis. Spacegroup Fd-3m, structural model  $[Zn]^{8a}[Fe_2]^{16d}O^{32e}$ . Sample ZFTO25 contains 9.76(13) wt.-% of  $Fe_2TiO_5$

Ti [mol per FU]	Sample name	Lattice parameter, a [Å]	O fractional coordinate at 32e site $x=y=z$	Atomic distances in tetrahedra (8a – 32e)	Atomic distances in octahedra (8a – 32e)
0	ZnFe <sub>2</sub> O <sub>4</sub> Butylenglycol.	8.4420(6)	0.26110(28)	1.990(4)	2.021(2)
0	ZnFe <sub>2</sub> O <sub>4</sub> Ethylenglycol	8.4421(6)	0.2612(3)	1.992(4)	2.020(1)
0.04	ZFTO4	8.4415(6)	0.26061(27)	1.983(4)	2.0248(19)
0.07	ZFTO7	8.4402(7)	0.26114(32)	1.995(4)	2.018(2)
0.09	ZFTO9	8.4435(6)	0.26043(24)	1.981(4)	2.0267(18)
0.1	ZFTO10	8.43549(13)	0.2589(3)	1.957(4)	2.036(2)
0.13	ZFTO13	8.4443(9)	0.26010(27)	1.976(4)	2.029(2)
0.25	ZFTO25	8.4497(7)	0.26022(22)	1.979(3)	2.0298(16)

Table SI 2: Molar ratios of the spinel main phase to consider site distributions based on  $[Zn]^{8a}[Fe_2]^{16d}O^{32e}$ . (\*) Sample ZFTO25 contains 9.76(13) wt.-% of  $Fe_2TiO_5$ , thus the spinel main phase stoichiometry was recalculated to  $Zn_{1.15}Fe_{1.74}Ti_{0.11}O_4$

Ti [mol per FU]	Sample name	Zn/Fe molar ratio	Zn/(Fe+Ti) molar ratio
0	ZnFe <sub>2</sub> O <sub>4</sub> Butylenglycol.	0.490	0.480
0	ZnFe <sub>2</sub> O <sub>4</sub> Ethylenglycol	0.475	0.475
0.04	ZFTO4	0.475	0.475
0.07	ZFTO7	0.659	0.619
0.09	ZFTO9	0.516	0.483
0.1	ZFTO10	0.508	0.482
0.13	ZFTO13	0.591	0.562
0.11*	ZFTO25	0.503	0.485

Table SI 3 Results from density measurements. Density measurements were performed using an Archimedean scale, the measured density was compared to the density from PXRD measurements.

Sample	Density (% of theoretical density)
ZFO	82
ZFTO4	88
ZFTO7	85
ZFTO9	90
ZFTO10	60
ZFTO13	89
ZFTO25	90

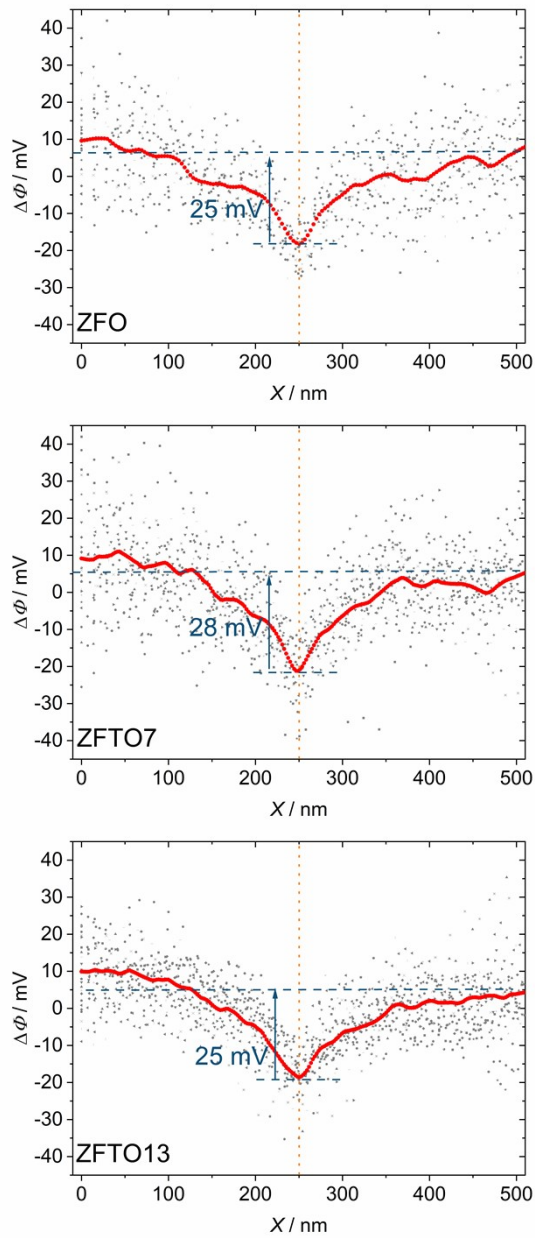


Figure SI 2 Analysis of the surface potential difference at the grain boundaries from KPFM. The crystallographic grain boundary position is indicated by orange dotted line. The average surface potential is shown in red, all measured data (25 different positions for each sample) are shown as grey dots.

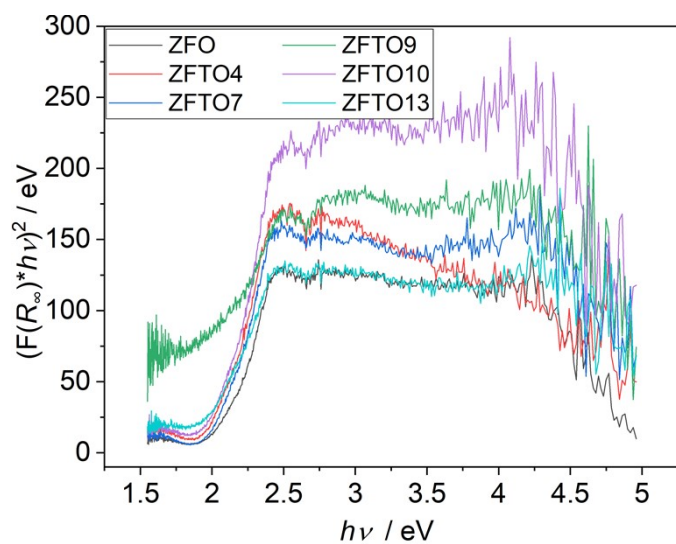


Figure 3 Tauc plot calculated from the optical data.

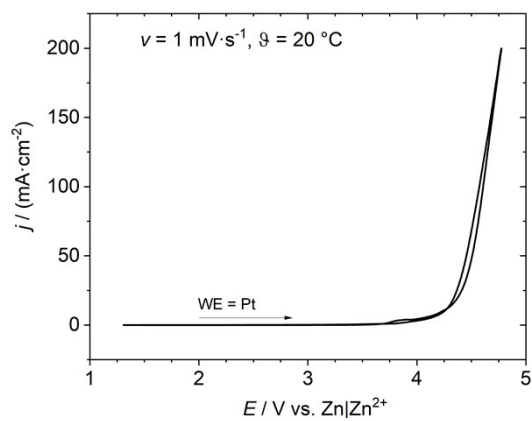
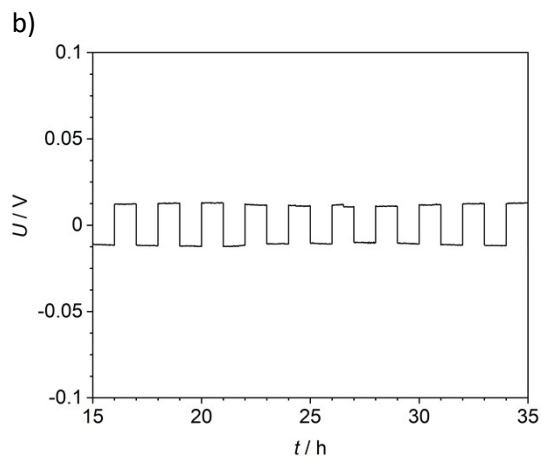
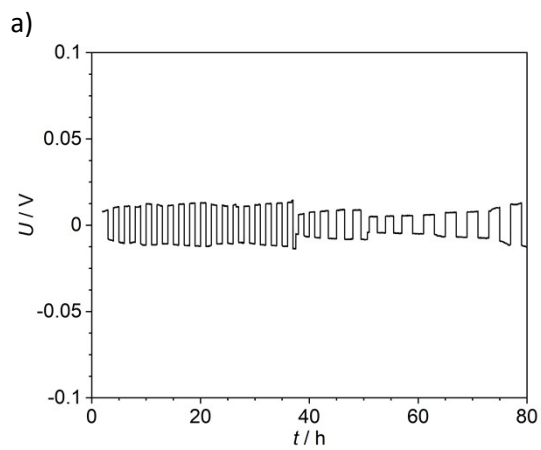


Figure SI 4 Cyclic voltammetry measurement to determine the oxidative stability of organic electrolyte 0.5 M zinc triflate in acetonitrile.



c)

d)

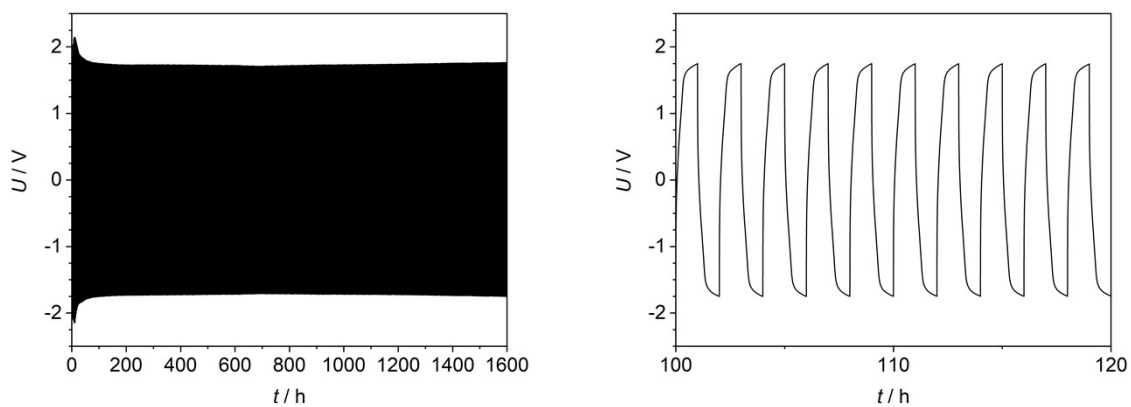


Figure SI 5 Cycling experiment in symmetric cell setup: a) ZFO | ZFO with sintered material at increased current density starting at  $0.1 \text{ mA cm}^{-2}$ ,  $0.15 \text{ mA cm}^{-2}$  to  $0.2 \text{ mA cm}^{-2}$ . b) enlarged area of a) between 15 to 35 h, current density  $0.1 \text{ mA cm}^{-2}$ . c) ZFTO9 | ZFTO9 with calcined material at  $0.1 \text{ mA cm}^{-2}$ , 800 cycles, d) time range between 100-120 h in c) to show shape of individual charge-discharge curves.

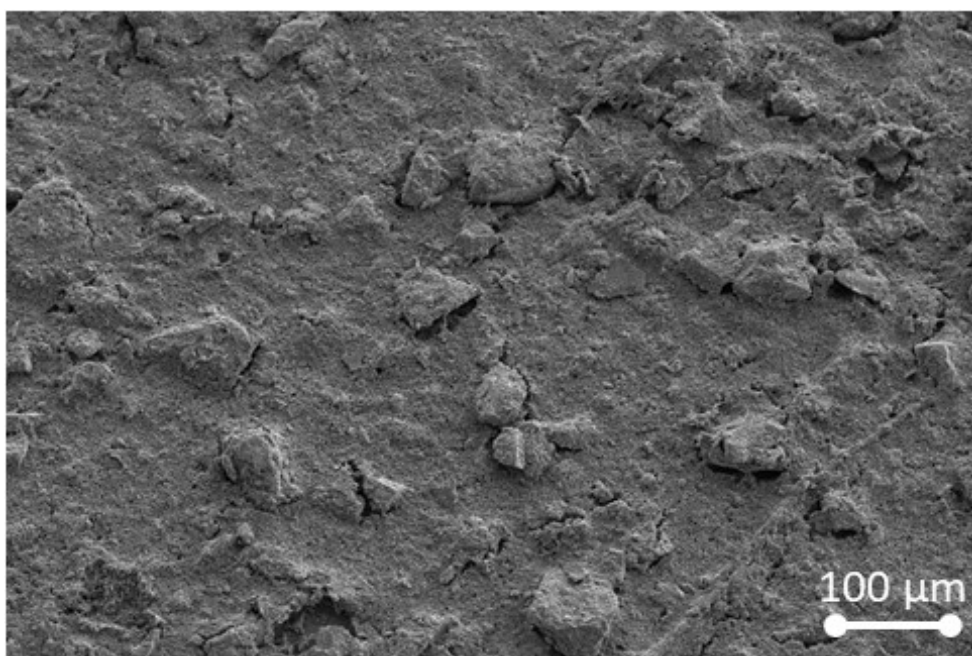


Figure SI 6 Exemplary SEM image of the electrode sheet with undoped  $\text{ZnFe}_2\text{O}_4$ , showing very large particle sizes. The other electrode sheets showed a similar particle size distribution.