

Supplementary Materials: Acoustic Emission Monitoring of High-Entropy Oxyfluoride Rock-Salt Cathodes during Battery Operation

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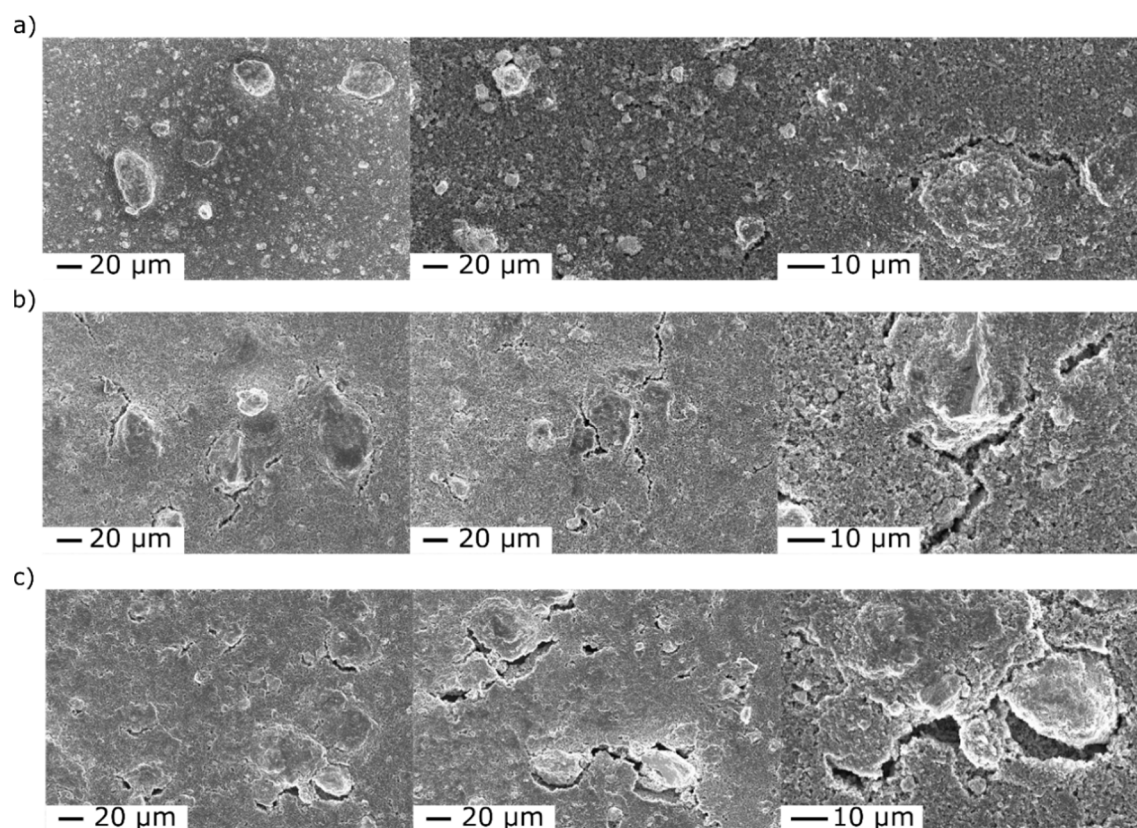


Figure S1. Top-view SEM images at different magnifications of the LiHEOF cathode charged to different cutoff voltages of a) 4.6 V, (b) 4.8 V and (c) 5.0 V vs Li⁺/Li in the fifth cycle.

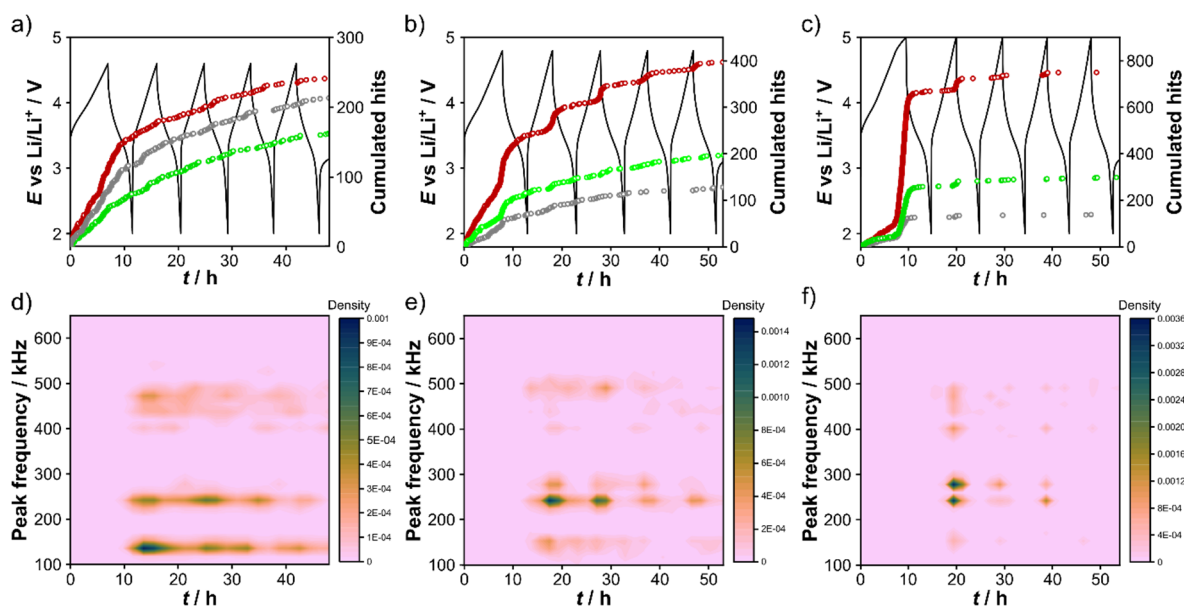


Figure S2. Voltage profile (black) of LiHEOF|Li cells cycled at 25 °C and C/10 rate between 2.0 V and either a) 4.6 V, b) 4.8 V or c) 5.0 V vs Li⁺/Li and corresponding cumulative AE hits for different peak frequency ranges (100-200 kHz in gray, 200-300 kHz in red, 300-600 kHz in green). d-f) Contour plots of hit density versus the time and peak frequency (from the second cycle onward). d) 2.0-4.6 V, e) 2.0-4.8 V and f) 2.0-5.0 V vs Li⁺/Li.

Table S1. Mean values of characteristic parameters for the AE 1 (100-200 kHz), AE 2 (200-300 kHz) and AE 3 (300-600 kHz) signals detected over the first five cycles in LiHEOF|Li cells cycled at C/10 rate and 25 °C in the voltage range of a) 2.0-4.6 V, b) 2.0-4.8 V and c) 2.0-5.0 V vs Li⁺/Li.

a)				
Parameter	AE 1	AE 2	AE 3	
Rise time / μ s	8.09	6.69	3.85	
Duration / μ s	47.99	38.88	35.05	
Amplitude / dB	35.19	34.79	36.80	
Peak frequency / kHz	137.76	244.02	454.70	
b)				
Parameter	AE 1	AE 2	AE 3	
Rise time / μ s	6.98	7.24	3.36	
Duration / μ s	35.47	38.08	32.09	
Amplitude / dB	35.19	36.72	38.78	
Peak frequency / kHz	136.21	255.12	454.98	
c)				
Parameter	AE 1	AE 2	AE 3	
Rise time / μ s	8.28	5.56	3.67	
Duration / μ s	40.42	32.26	30.82	
Amplitude / dB	35.47	35.28	36.33	
Peak frequency / kHz	141.71	252.58	456.23	

Table S2. Proportion of cumulative hits for the AE 1 (100-200 kHz), AE 2 (200-300 kHz), and AE 3 (300-600 kHz) signals detected during the first charge cycle in LiHEOF|Li cells cycled at C/10 rate and 25 °C in the voltage range of a) 2.0-4.6 V, b) 2.0-4.8 V and c) 2.0-5.0 V vs Li⁺/Li.

a)			
2.0-4.6 V	Cumulative hits	Share / %	
AE1	79	33	
AE2	110	45	
AE3	53	22	
b)			
2.0-4.8 V	Cumulative hits	Share / %	
AE1	54	17	
AE2	175	56	
AE3	81	26	
c)			
2.0-5.0 V	Cumulative hits	Share / %	
AE1	119	13	
AE2	578	64	
AE3	208	23	