Batteries & Supercaps

Supporting Information

Gassing Behavior of High-Entropy Oxide Anode and Oxyfluoride Cathode Probed Using Differential Electrochemical Mass Spectrometry

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$$S_{\mathrm{config}} = -\mathrm{R}[(\sum_{\mathrm{i}=1}^{\mathrm{N}} x_{\mathrm{i}} \ln x_{\mathrm{i}})_{\mathrm{cation}} + (\sum_{\mathrm{j}=1}^{\mathrm{M}} x_{\mathrm{j}} \ln x_{\mathrm{j}})_{\mathrm{anion}}]$$

Equation S1. Calculation of configurational entropy (S_{config}), with x_{i} and x_{j} representing the molar fraction of cations and anions on the respective sublattice.

Table S1. Cycling protocol used for the electrochemical testing of the HEO anode material in LIB half-cells. 1/10$^{\mathrm{th}}$ of the set current was used as the termination criterion in the CV steps.

Step	Reaction	Cutoff voltage	Rate	Mode	Number of cycles
1	Lithiation	10 mV	C/20	CC-CV	2
2	Delithiation	2500 mV	C/20	CC-CV	
3	Lithiation	10 mV	C/5	CC-CV	10
4	Delithiation	2500 mV	C/5	CC-CV	
5	Lithiation	10 mV	C/5	CC-CV	2
6	Delithiation	2500 mV	C/2	CC	
7	Lithiation	10 mV	C/5	CC-CV	2
8	Delithiation	2500 mV	C/5	CC	
9	Lithiation	10 mV	C/5	CC-CV	2
10	Delithiation	2500 mV	C/10	CC	
11	Lithiation	10 mV	C/5	CC-CV	20
12	Delithiation	2500 mV	C/5	CC-CV	
13	Continue with step no. 5				

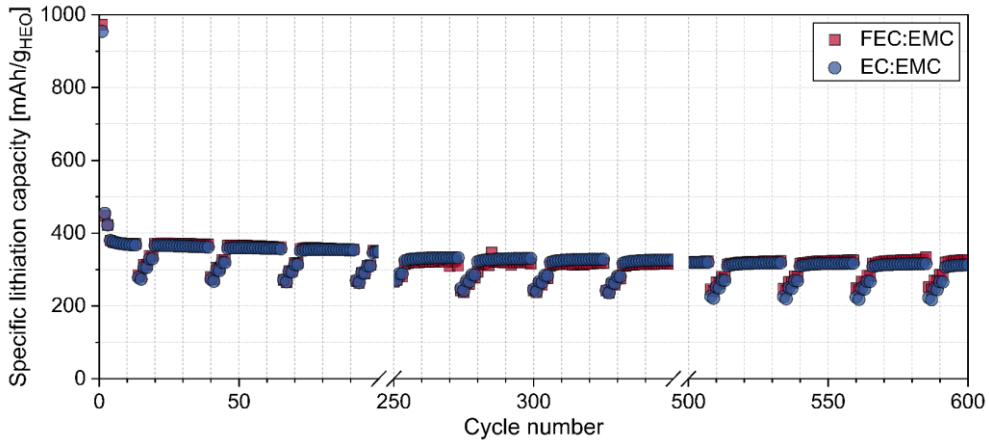

Figure S1. Long-term cycling performance of the HEO anode material in a LIB half-cell at a C/5 rate in the voltage range of 10–2500 mV versus Li^+/Li. LP57 (blue) or 1 M $LiPF_6$ in FEC:EMC (red) was used as electrolyte. Note that the first two activation or formation cycles were performed at a C/20 rate.

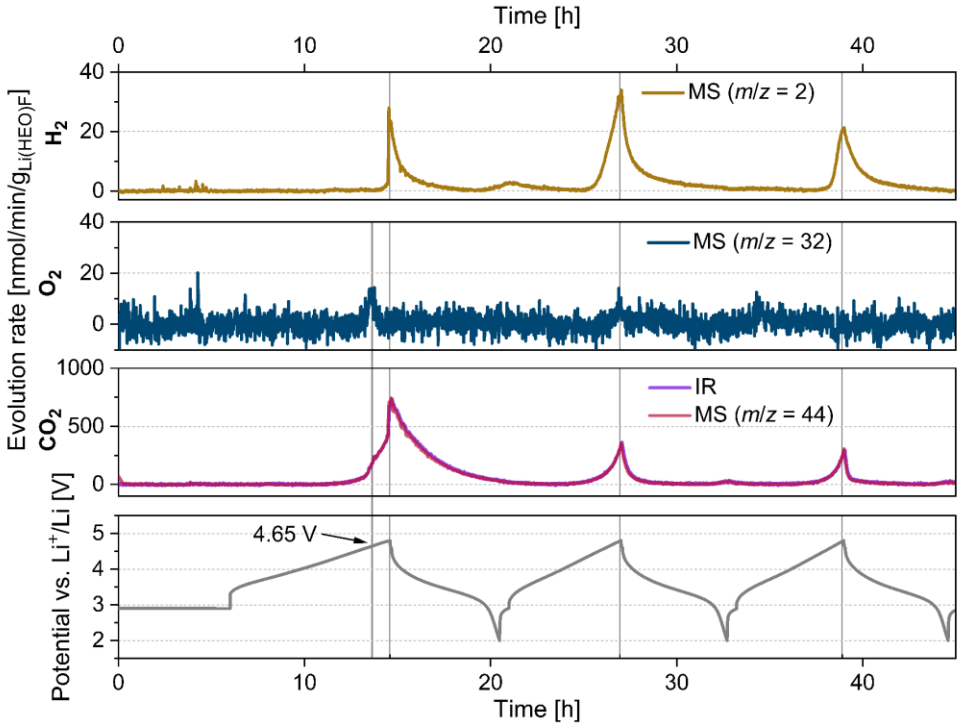

Figure S2. Gassing behavior of the Li(HEO)F cathode material in a LIB half-cell using LP57 electrolyte at a C/10 rate in the voltage range of 2.0–4.8 V versus Li^+/Li. Voltage profiles (gray) for the first three cycles and the corresponding gas evolution of H_2 (*m*/*z* = 2, gold), O_2 (*m*/*z* = 32, blue) and CO_2 (*m*/*z* = 44, pink; and via integration of IR absorbance [2315–2380 cm^{-1}], purple) are shown. No significant amount of CO was detected.

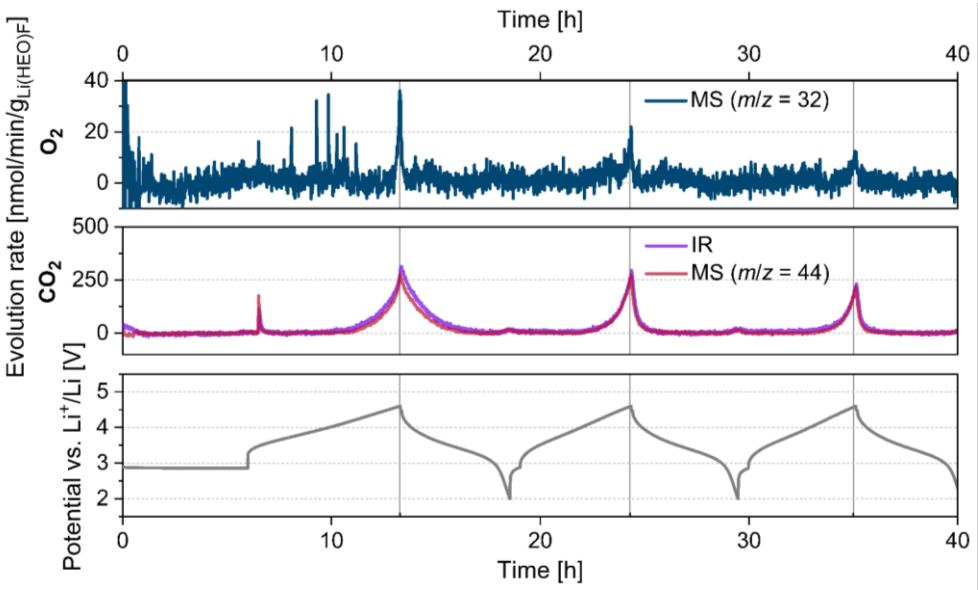

Figure S3. Gassing behavior of the Li(HEO)F cathode material in a LIB half-cell using LP57 electrolyte at a C/10 rate in the voltage range of 2.0–4.6 V versus Li^+/Li. Voltage profiles (gray) for the first three cycles and the corresponding gas evolution of O_2 (*m*/*z* = 32, blue) and CO_2 (*m*/*z* = 44, pink; and via integration of IR absorbance [2315–2380 cm^{-1}], purple) are shown. No significant amounts of H_2 and CO were detected.

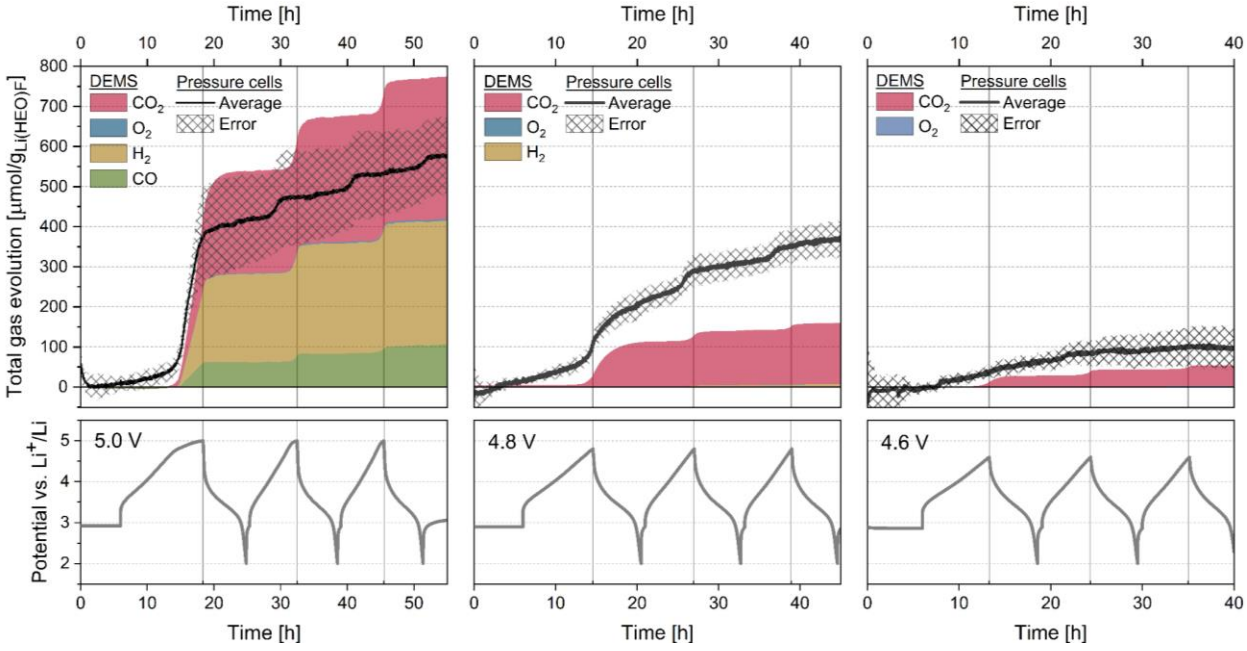

Figure S4. Quantitative comparison of the results obtained from *in situ* pressure and DEMS measurements (integrated amounts of evolved gases).