Electronic Supplementary Information (ESI)

for

Tunable pseudocapacitive contribution by dimension control in nanocrystalline-constructed $(Mg_{0.2}Co_{0.2}Ni_{0.2}Cu_{0.2}Zn_{0.2})O$ solid solutions to achieve superior lithium-storage properties

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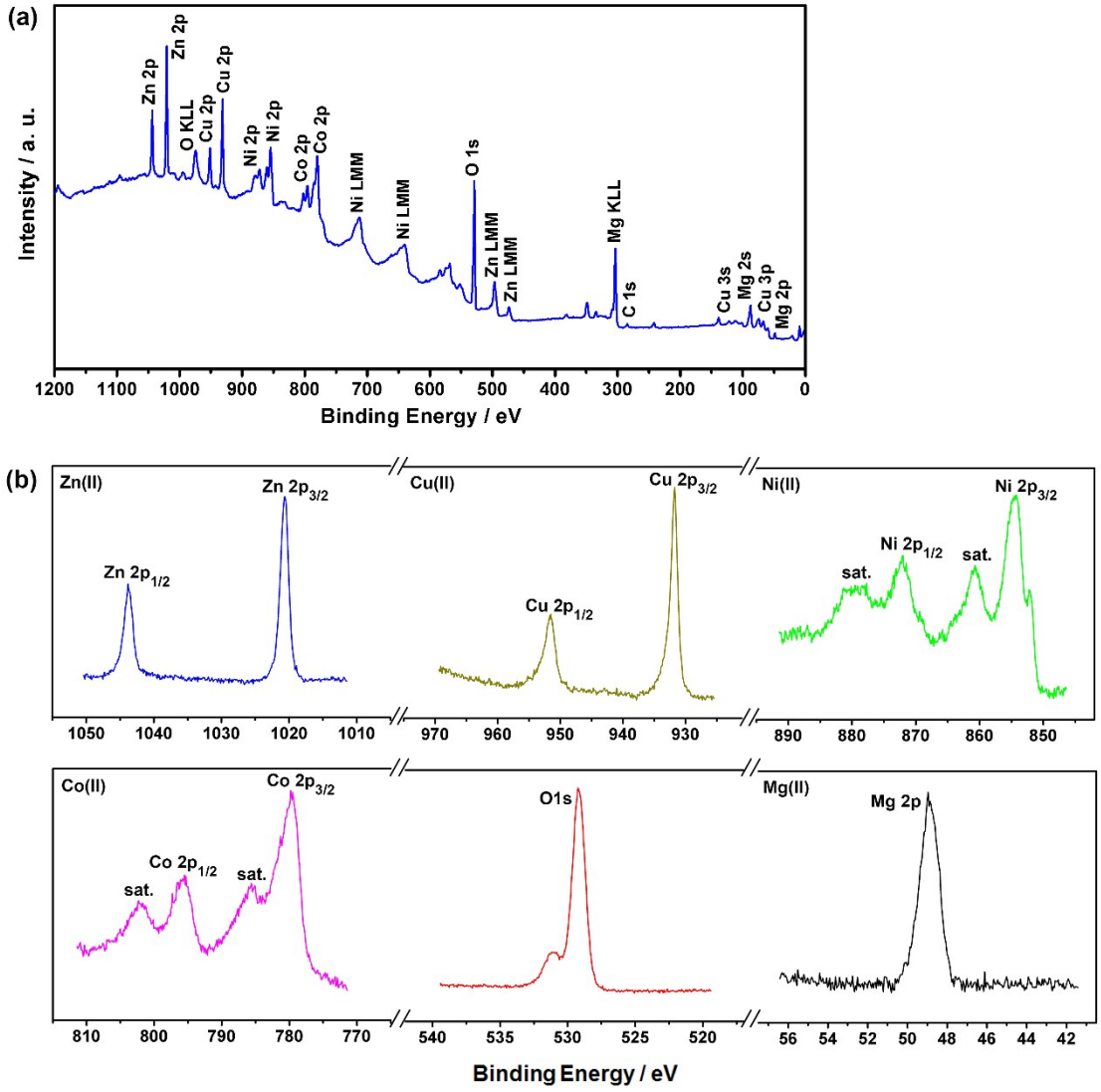


Figure S1. XPS spectrum of the as-prepared HEO-Film, (a) Survey scan; and (b) High-resolution XPS spectra of Zn 2p, Cu 2p, Ni 2p, Co 2p, O 1s, Mg 2p.

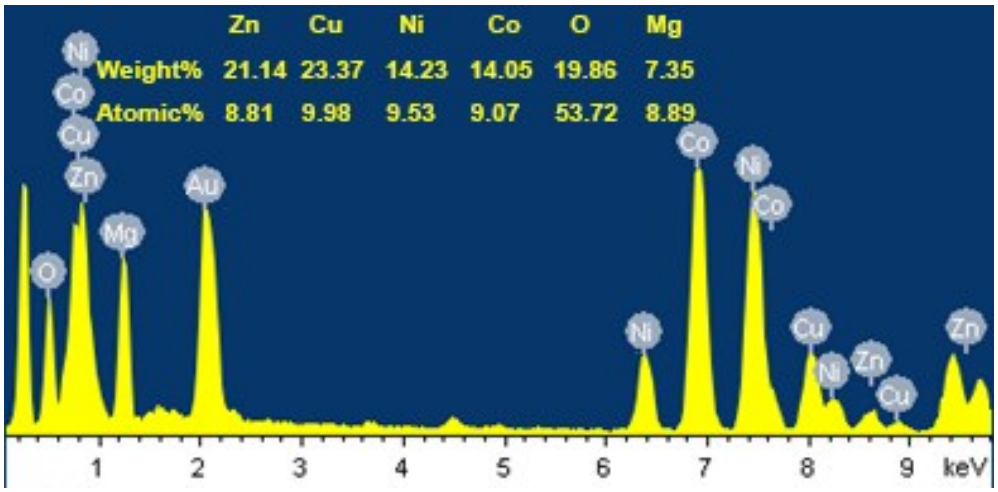


Figure S2. STEM-EDS analysis of the as-prepared HEO-Film on a gold grid. The corresponding element atomic ratio indicate the homogeneous spatial distributions of each element.

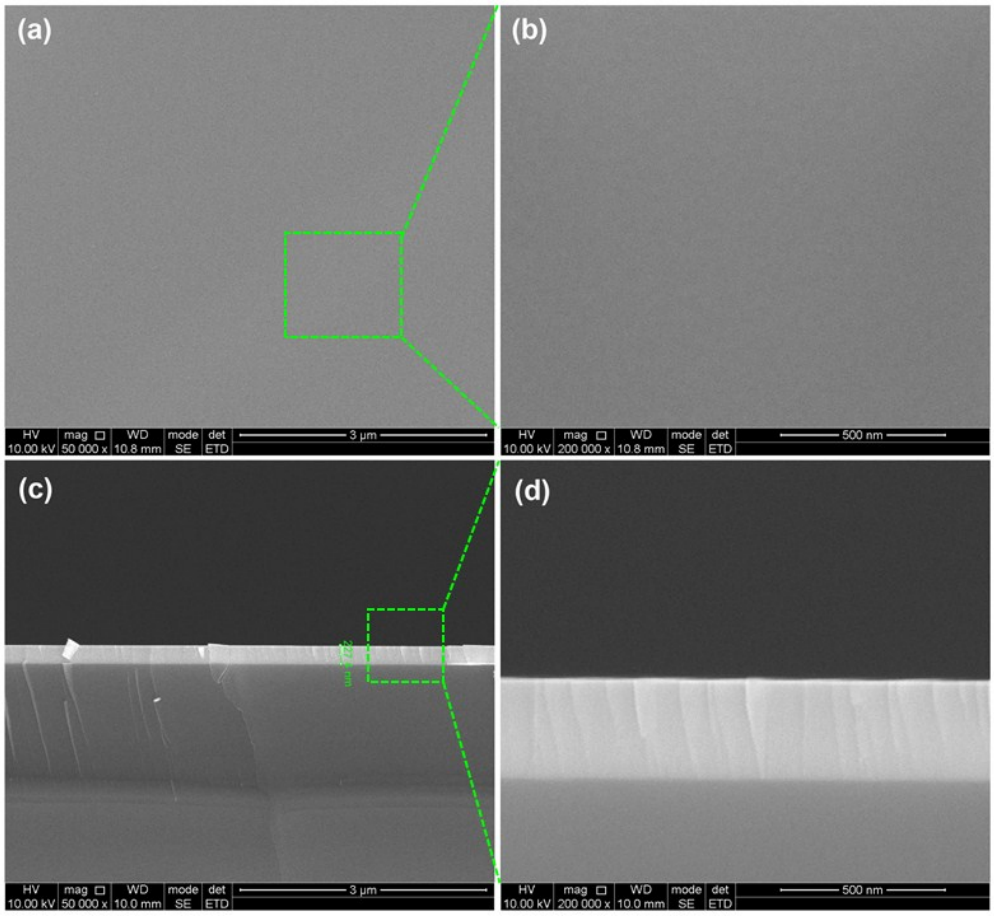


Figure S3. (a-b) Surface and (c-d) cross-section SEM images of the as-deposited HEO thin film.

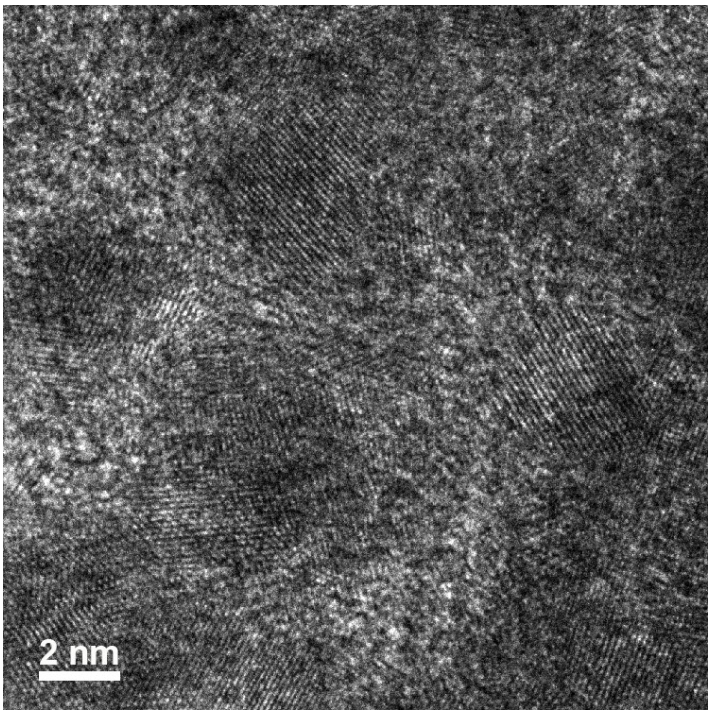


Figure S4. The enlarged HRTEM pattern of **Figure 4(c)**.

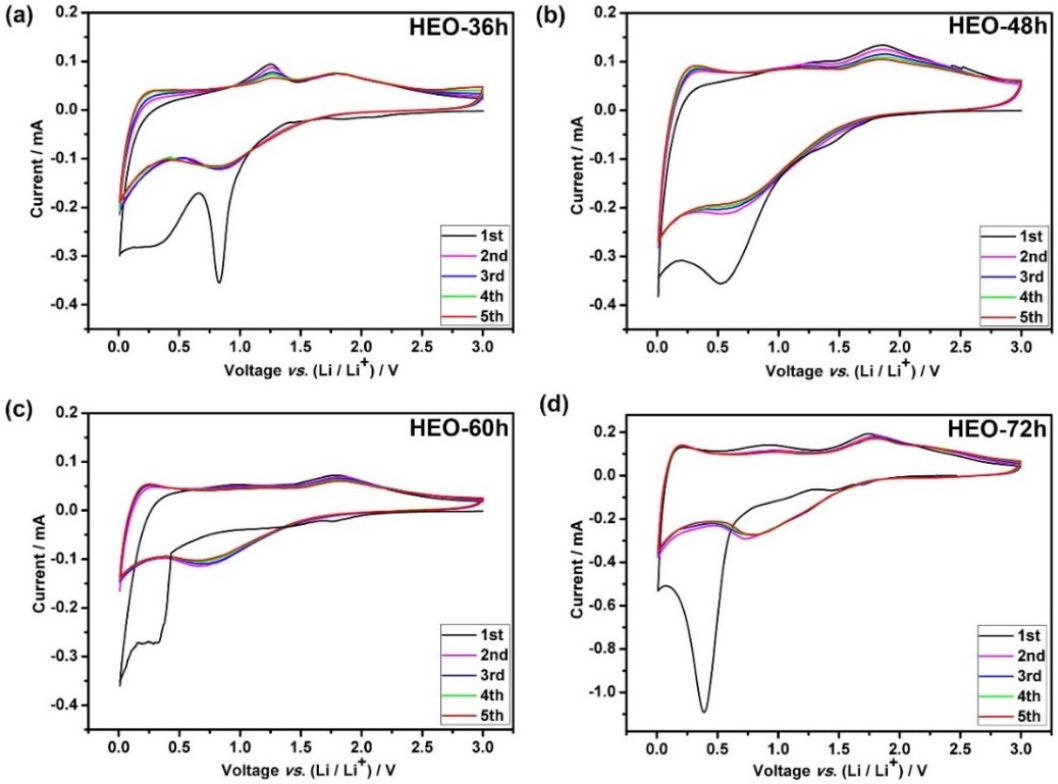


Figure S5. CV curves of (a) HEO-36h, (b) HEO-48h, (c) HEO-60h and (d) HEO-72h electrodes at a scan rate of 0.2 mV s^{-1}.

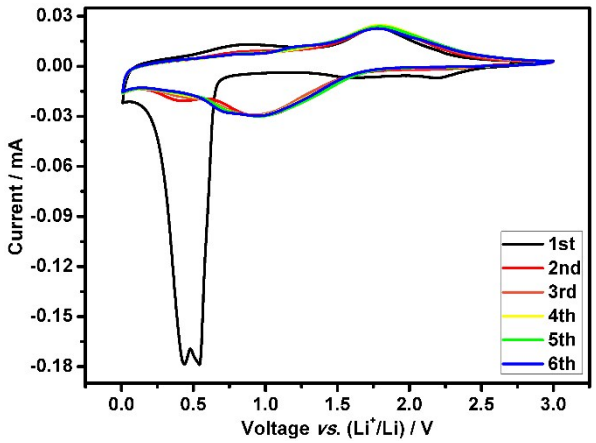


Figure S6. CV curve of HEO-Film electrode at a scan rate of 0.2 mV s^{-1}.

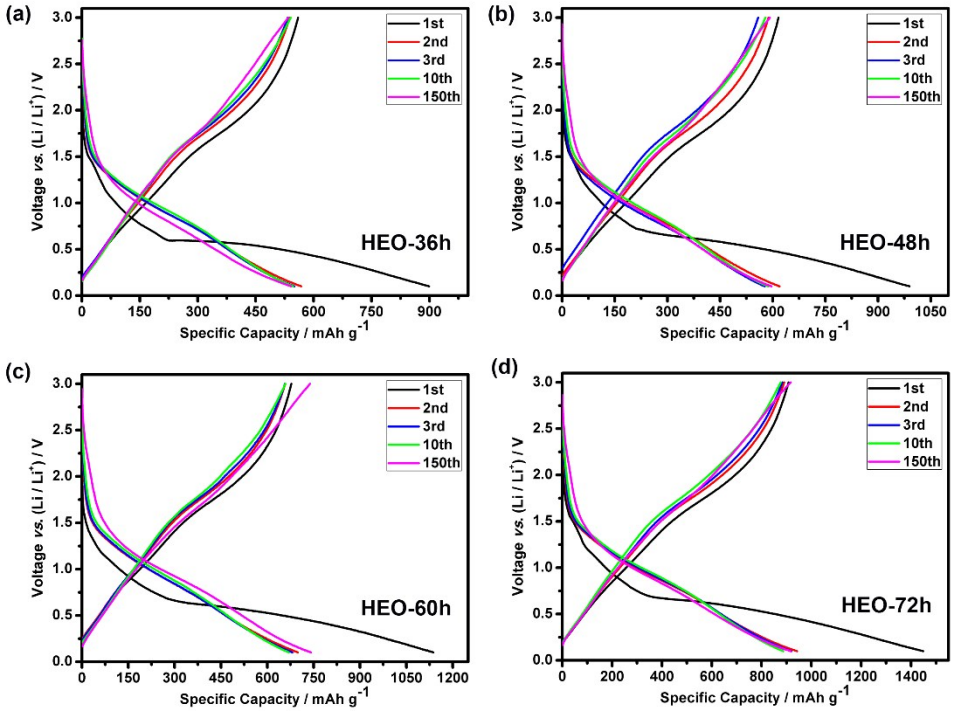
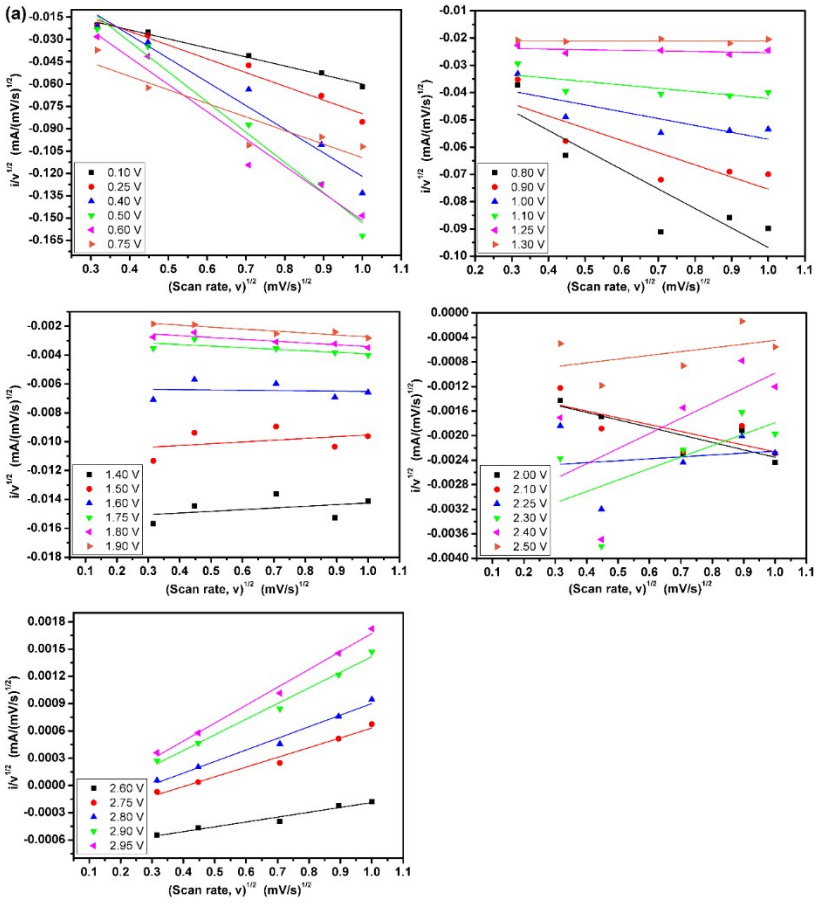


Figure S7. Discharge-charge voltage profiles of HEO electrodes for the 1st, 2nd, 3rd, 10th, and 150th cycles in the voltage range of 0.01-3.00 V at a current rate of 200 mA g^{-1}.


(a)
$iv^{1/2}$ (mA/(mV/s)$^{1/2}$)
(Scan rate, v)$^{1/2}$ (mV/s)$^{1/2}$
0.10 V
0.25 V
0.40 V
0.50 V
0.60 V
0.75 V
0.80 V
0.90 V
1.00 V
1.10 V
1.25 V
1.30 V
1.40 V
1.50 V
1.60 V
1.75 V
1.80 V
1.90 V
2.00 V
2.10 V
2.25 V
2.30 V
2.40 V
2.50 V
2.60 V
2.75 V
2.80 V
2.90 V
2.95 V

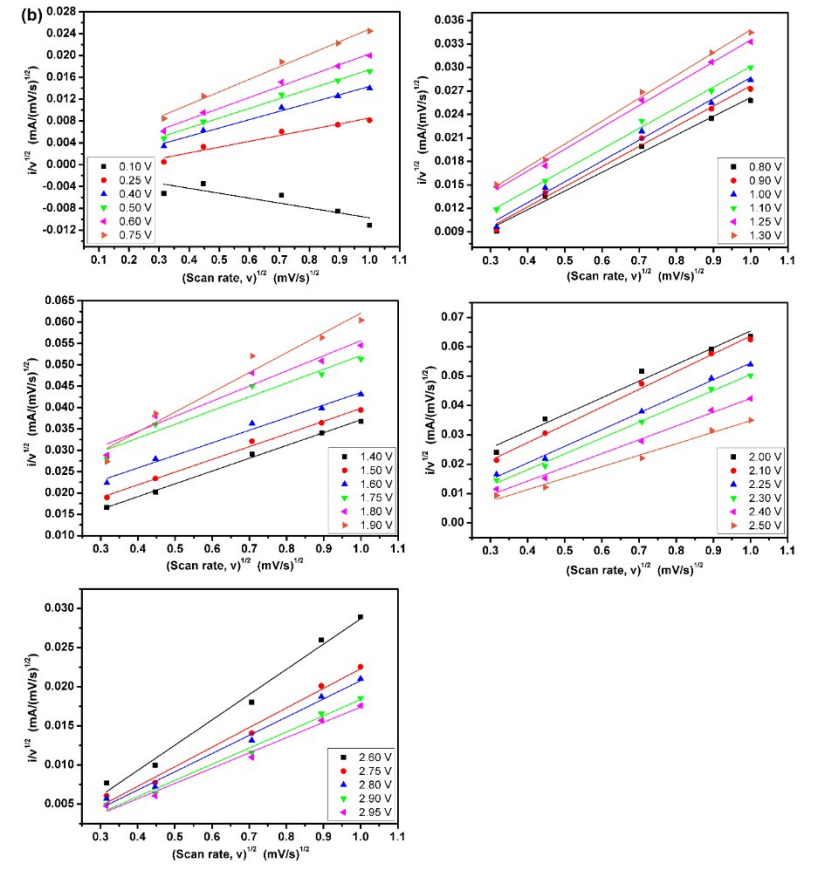

(b)
$iv^{1/2}$ (mA/(mV/s)$^{1/2}$)
(Scan rate, v)$^{1/2}$ (mV/s)$^{1/2}$
0.10 V
0.25 V
0.40 V
0.50 V
0.60 V
0.75 V
0.80 V
0.90 V
1.00 V
1.10 V
1.25 V
1.30 V
1.40 V
1.50 V
1.60 V
1.75 V
1.80 V
1.90 V
2.00 V
2.10 V
2.25 V
2.30 V
2.40 V
2.50 V
2.60 V
2.75 V
2.80 V
2.90 V
2.95 V

Figure. S8. Correlation between the scan rates and corresponding currents of the HEO-Film electrode at (a) anodic and (b) cathodic scan according to equation: $i(V)/ v^{1/2} = k_1 v^{1/2} + k_2$.

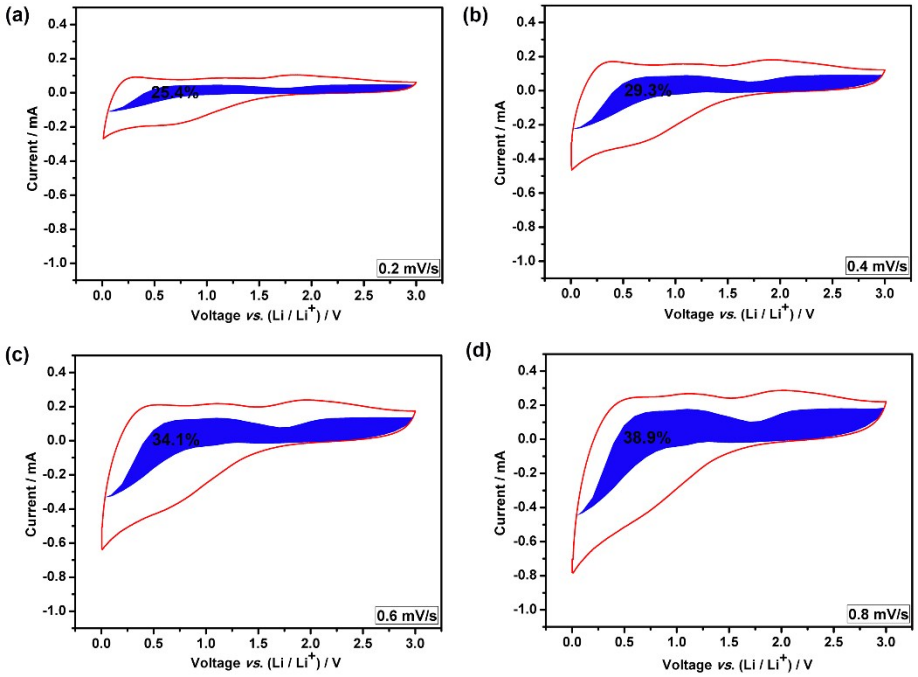


Figure S9. The capacitive contribution (blue shaded region) to the total current contribution (red line) for the HEO-36h electrode at (a) 0.2, (b) 0.4, (c) 0.6, (d) 0.8 mV s^{-1} *vs.* Li^+/Li.

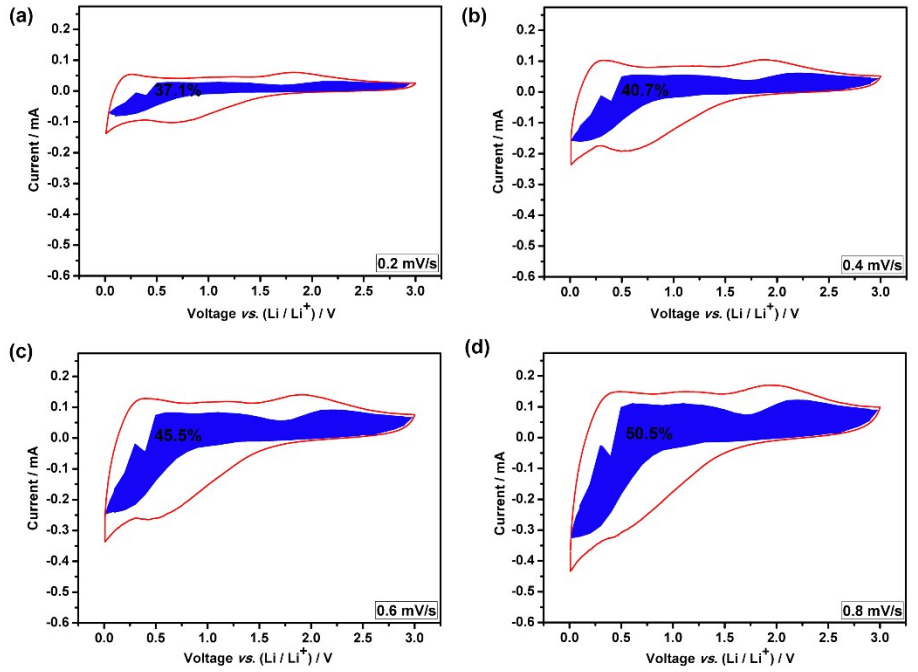


Figure S10. The capacitive contribution (blue shaded region) to the total current contribution (red line) for the HEO-48h electrode at (a) 0.2, (b) 0.4, (c) 0.6, (d) 0.8 mV s^{-1} *vs.* Li^+/Li.

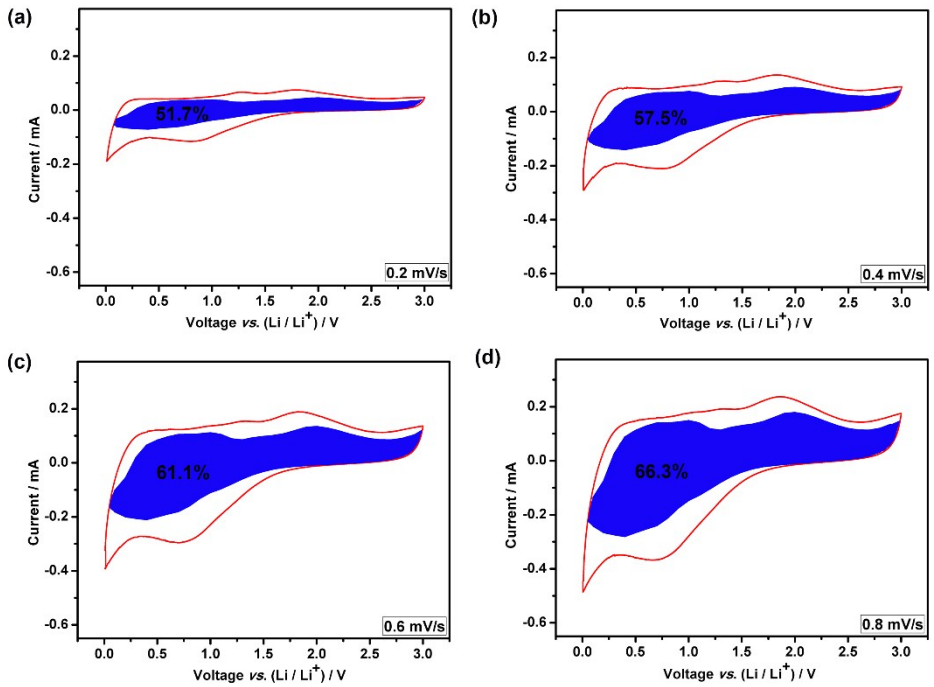


Figure S11. The capacitive contribution (blue shaded region) to the total current contribution (red line) for the HEO-60h electrode at (a) 0.2, (b) 0.4, (c) 0.6, (d) 0.8 mV s^{-1} *vs.* Li^+/Li.

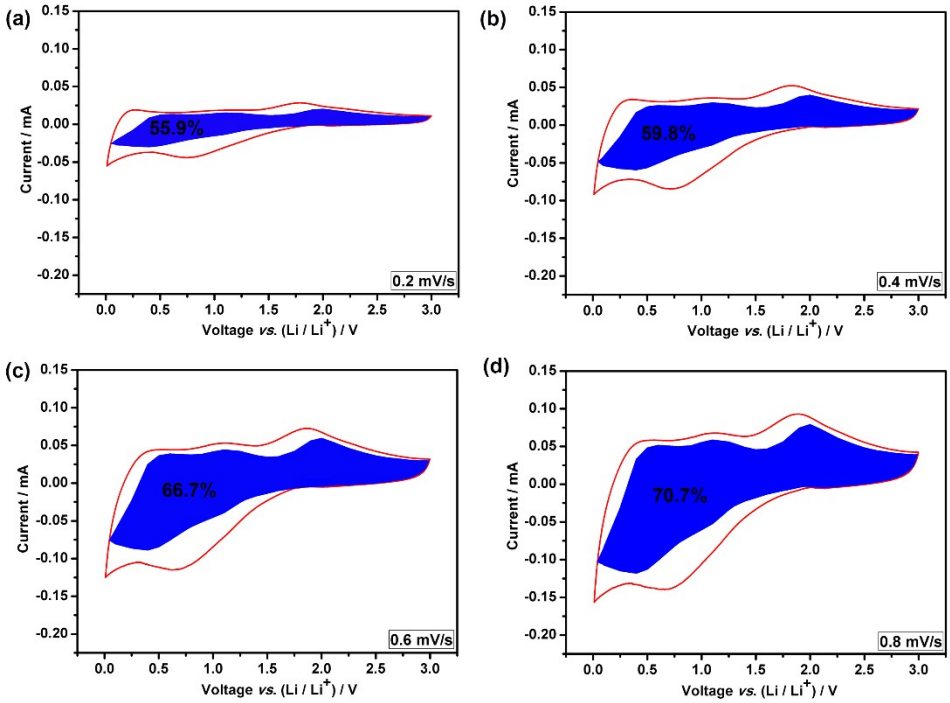


Figure S12. The capacitive contribution (blue shaded region) to the total current contribution (red line) for the HEO-72h electrode at (a) 0.2, (b) 0.4, (c) 0.6, (d) 0.8 mV s^{-1} *vs.* Li^+/Li.

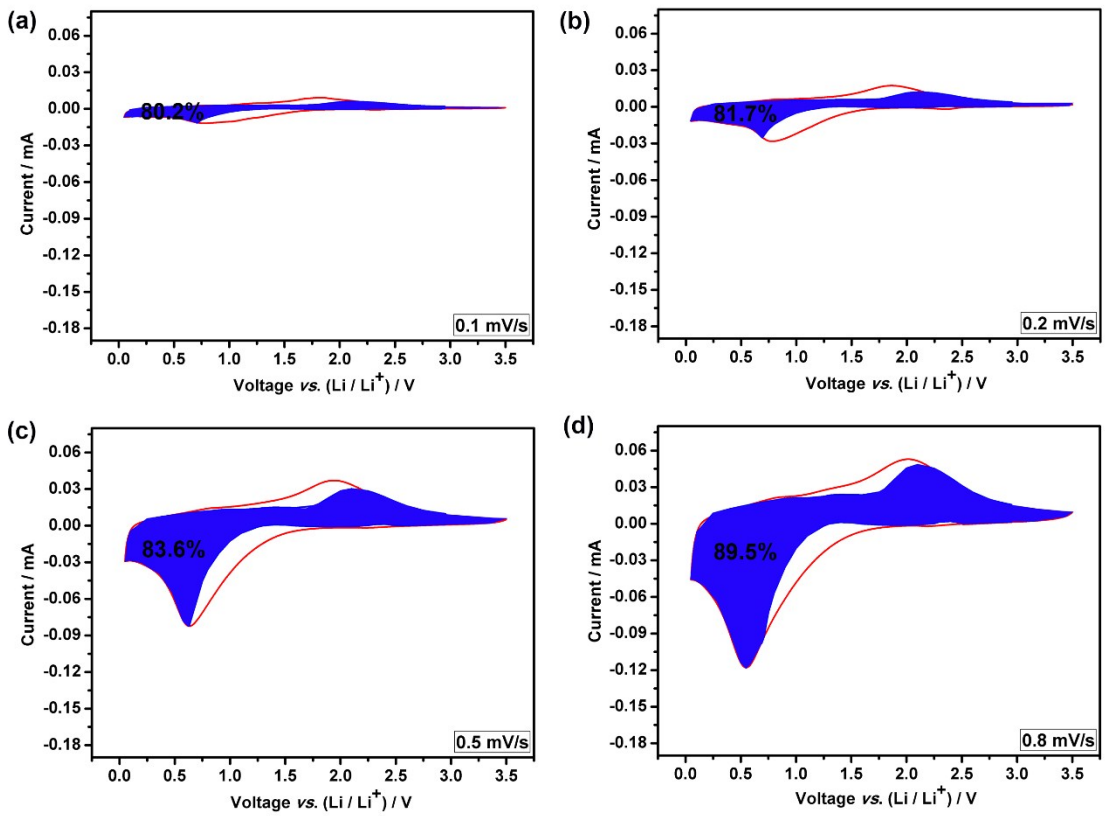


Figure S13. The capacitive contribution (blue shaded region) to the total current contribution (red line) for the HEO-Film electrode at (a) 0.1, (b) 0.2, (c) 0.5, (d) 0.8 mV s^{-1} *vs.* Li^+/Li.

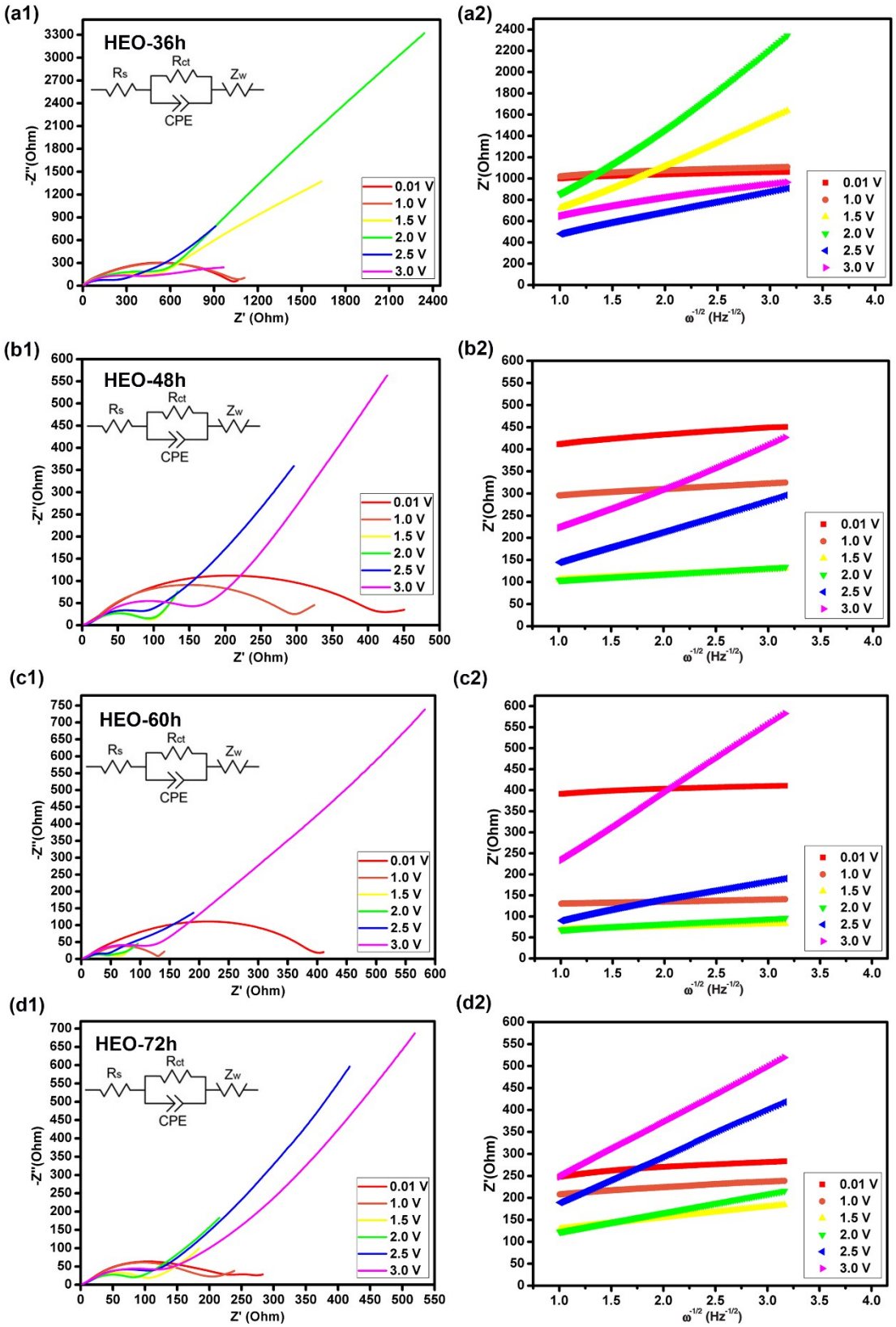


Figure S14. Determination of lithium-ion diffusion coefficients (D) by electrochemical impedance spectroscopy (EIS) measurements according to $D = R^2T^2/2A^2n^4F^4C^2\sigma^2$, where R, T, A, n, F, and C are the gas constant, absolute temperature, electrode area, electron transfer

number, Faraday constant, and lithium-ion molar concentration, respectively. The σ value (Warburg factor) can be calculated by $Z' = R_s + R_{ct} + \sigma\omega^{-1/2}$ from the low-frequency plots of EIS, and ω is the corresponding angular frequency.

Samples	R_s [Ω]	R_{ct} [Ω]	σ [Ω $Hz^{1/2}$]	D_{Li}[cm^2 s^{-1}]
HEO-36h (0.01 V)	1.89	979.21	27.3	9.38×10^{-13}
HEO-36h (1.0 V)	1.83	991.15	39.1	4.60×10^{-13}
HEO-36h (1.5 V)	1.75	274.32	425.0	3.93×10^{-15}
HEO-36h (2.0 V)	1.41	117.23	682.4	1.52×10^{-15}
HEO-36h (2.5 V)	1.45	291.33	195.7	1.81×10^{-14}
HEO-36h (3.0 V)	1.87	511.79	148.6	3.21×10^{-14}
HEO-48h (0.01 V)	3.87	391.54	18.3	2.13×10^{-12}
HEO-48h (1.0 V)	3.02	280.48	13.1	4.11×10^{-12}
HEO-48h (1.5 V)	2.89	90.03	11.9	5.09×10^{-12}
HEO-48h (2.0 V)	3.05	87.89	13.4	3.88×10^{-12}
HEO-48h (2.5 V)	2.71	71.75	69.7	1.47×10^{-14}
HEO-48h (3.0 V)	2.39	122.08	93.1	8.14×10^{-14}
HEO-60h (0.01 V)	4.41	380.46	8.7	3.59×10^{-14}
HEO-60h (1.0 V)	3.49	122.21	4.9	1.21×10^{-13}
HEO-60h (1.5 V)	3.11	58.85	6.7	6.19×10^{-14}
HEO-60h (2.0 V)	3.10	53.61	12.4	1.75×10^{-14}
HEO-60h (2.5 V)	2.50	44.71	45.9	1.33×10^{-15}
HEO-60h (3.0 V)	2.30	64.47	163.8	1.05×10^{-16}
HEO-72h (0.01 V)	2.84	234.00	15.9	7.05×10^{-12}
HEO-72h (1.0 V)	2.40	192.65	14.2	8.39×10^{-12}
HEO-72h (1.5 V)	2.20	101.20	25.9	2.60×10^{-12}
HEO-72h (2.0 V)	2.29	77.21	43.3	9.35×10^{-13}
HEO-72h (2.5 V)	2.25	78.90	106.9	1.51×10^{-13}
HEO-72h (3.0 V)	2.30	119.95	124.9	1.15×10^{-13}

Table S1. The kinetic parameters of different HEO samples at cut-off potential of 0.01, 1.0 1.5, 2.0, 2.5 and 3.0 V.

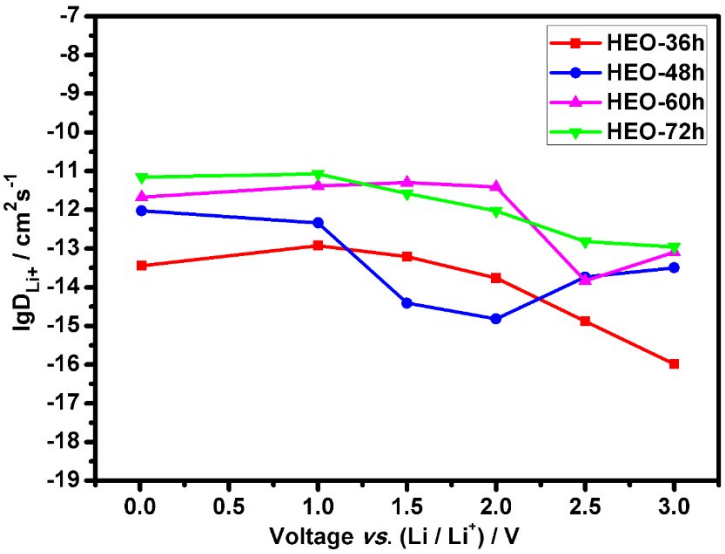


Figure S15. The lithium-ion diffusion coefficients (*D*) of different HEO samples at cut-off potential of 0.01, 1.0 1.5, 2.0, 2.5 and 3.0 V.