Supplementary Information

Temperature-mediated phase control in high entropy transition metal oxides for hybrid supercapacitor

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Molar Configurational Entropy Calculation

$$S_{Config} = -R\left[\left(\sum_{i=1}^{N} x_i \, ln(x_i)\right)_{cation-site} + \left(\sum_{j=1}^{N} x_j \, ln(x_j)\right)_{anion-site}\right] \quad \text{----------------} \ \mathbf{(1)}$$

Here, x_i and x_j represent the mole fraction of elements present in the cation and anion sites, respectively, and R is the gas constant.

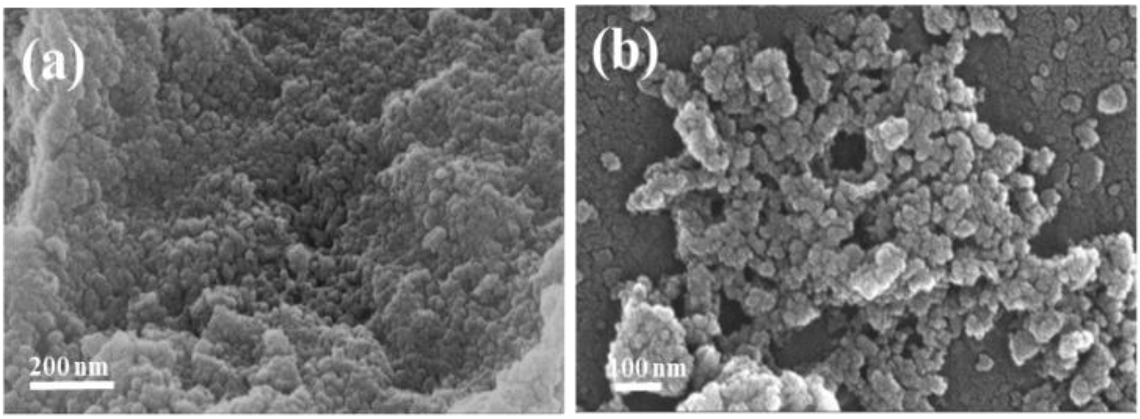

Fig. S1 FESEM images of HEO-110 (a, b).

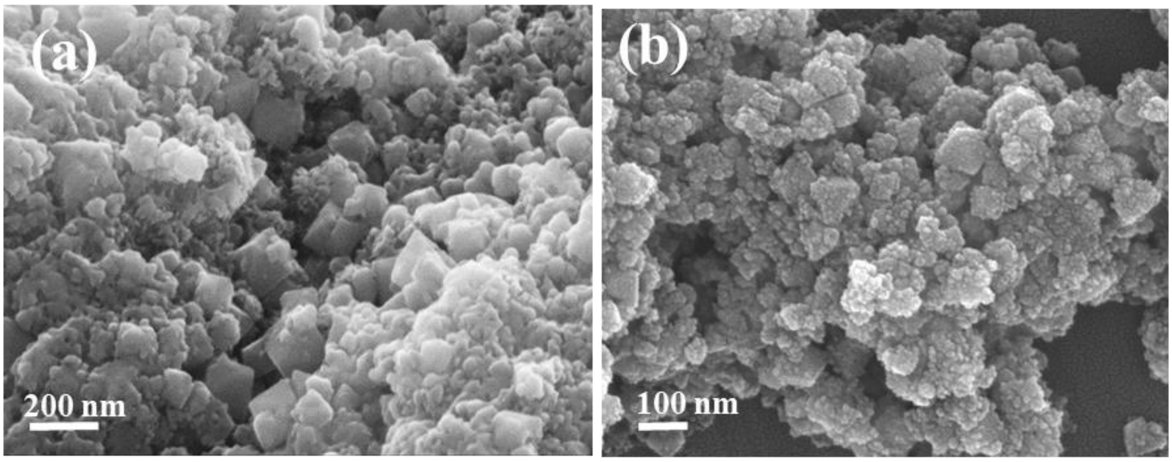

Fig. S2 FESEM images of HEO-170 (a, b).

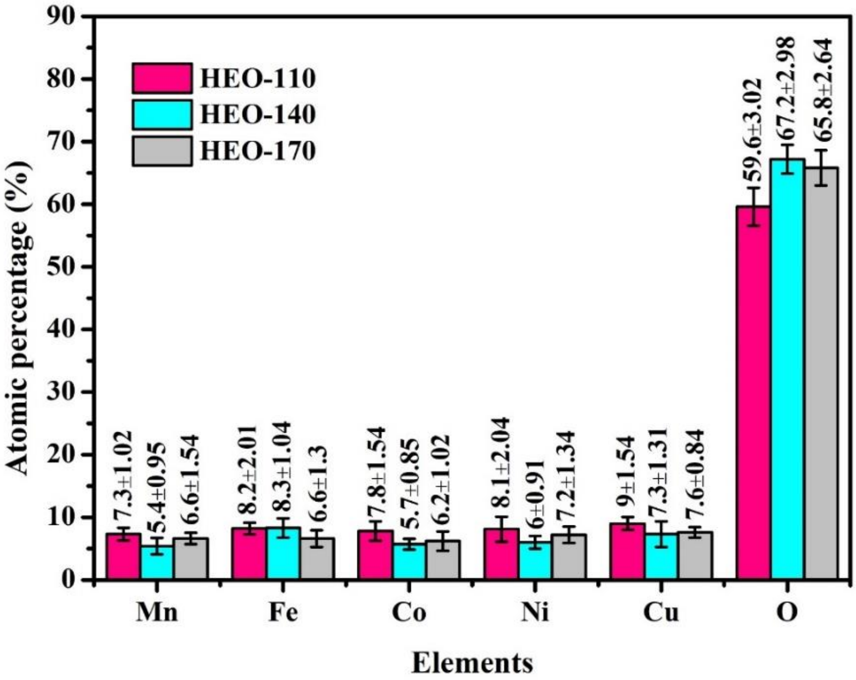

Fig. S3 Atomic percentages of each element in HEOs.

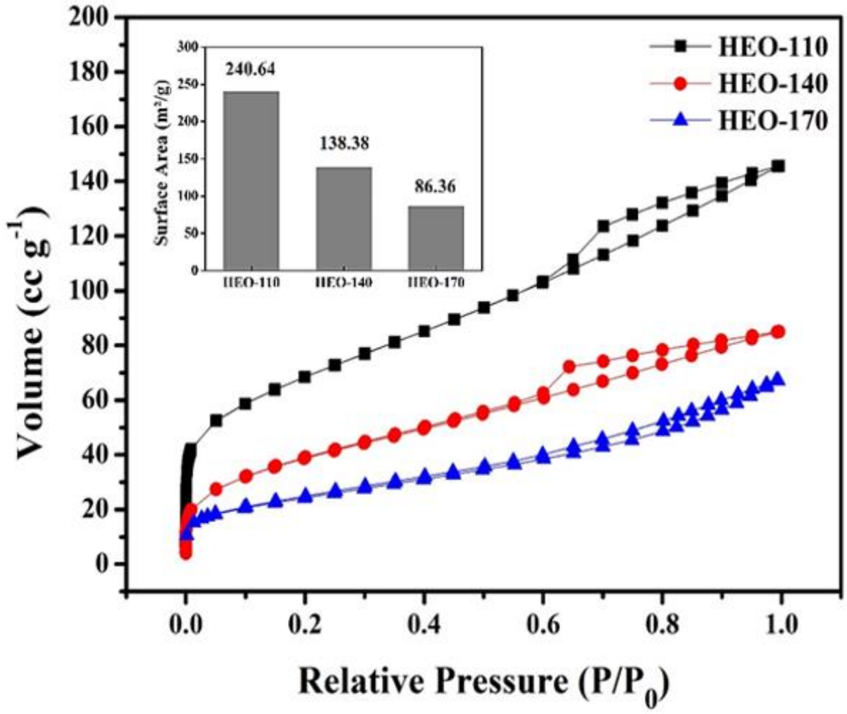

Fig. S4 N_2 adsorption-desorption isotherm of HEO-110, HEO-140 and HEO-170. The inset is the BET surface area of respective HEOs.

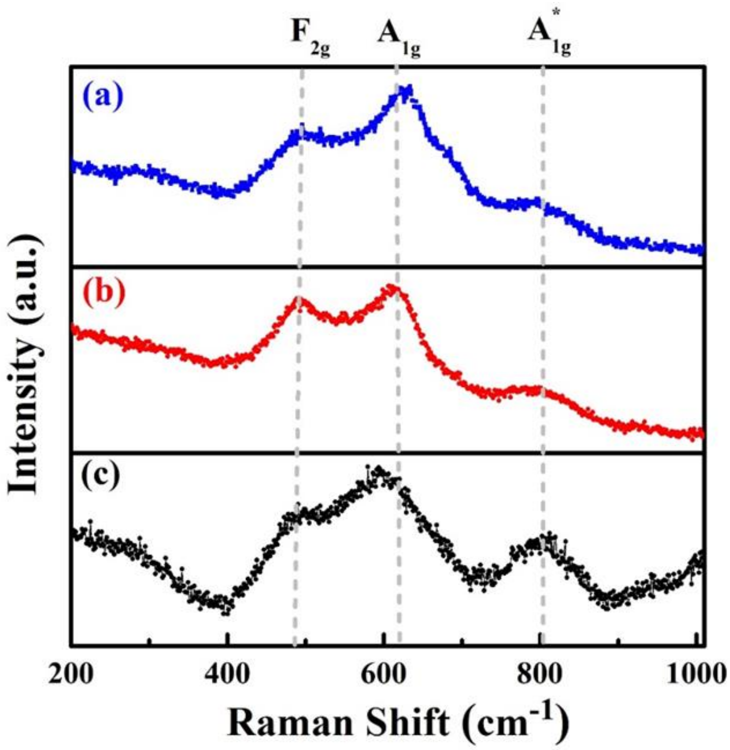

Fig. S5 Raman spectra of synthesized HEOs; (a) HEO-170, (b) HEO-140, (c) HEO-110.

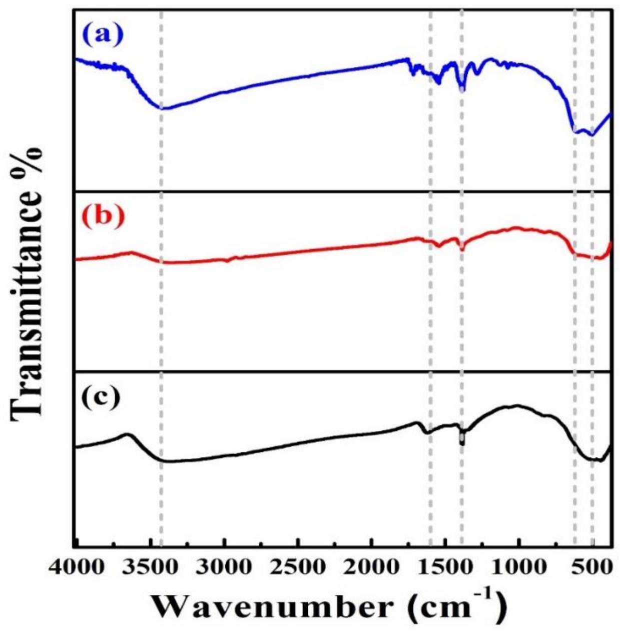

Fig. S6 IR spectra of synthesized HEOs; HEO-170 (a), HEO-140 (b), HEO-110 (c).

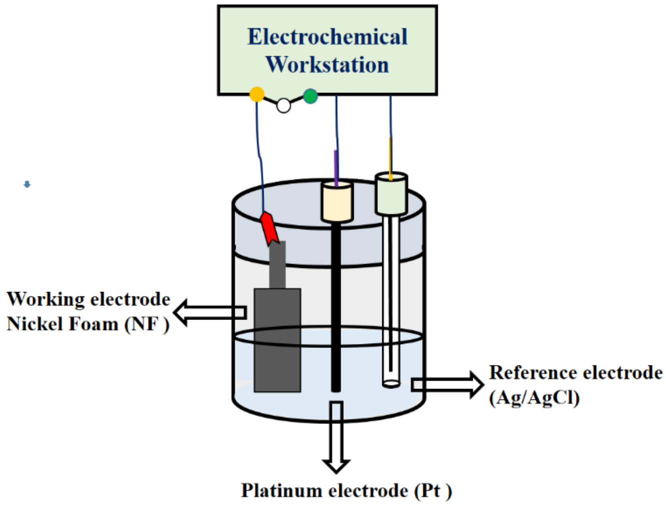

Fig. S7 Schematic diagram for the electrochemical study of electrode materials.

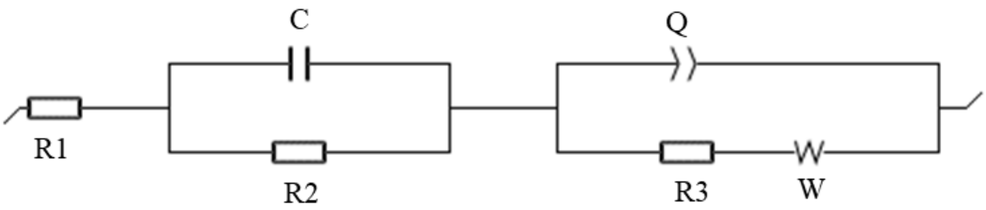

Fig. S8 Equivalent circuit model of EIS fitting.

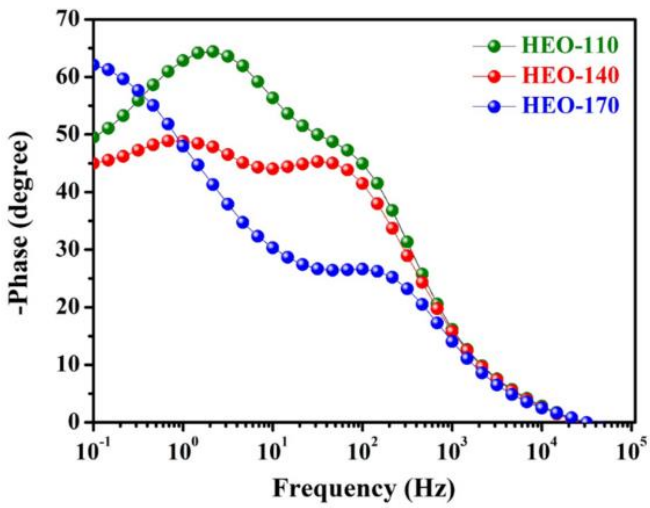

Fig. S9 The Bode plot of the temperature varied high entropy electrode material.

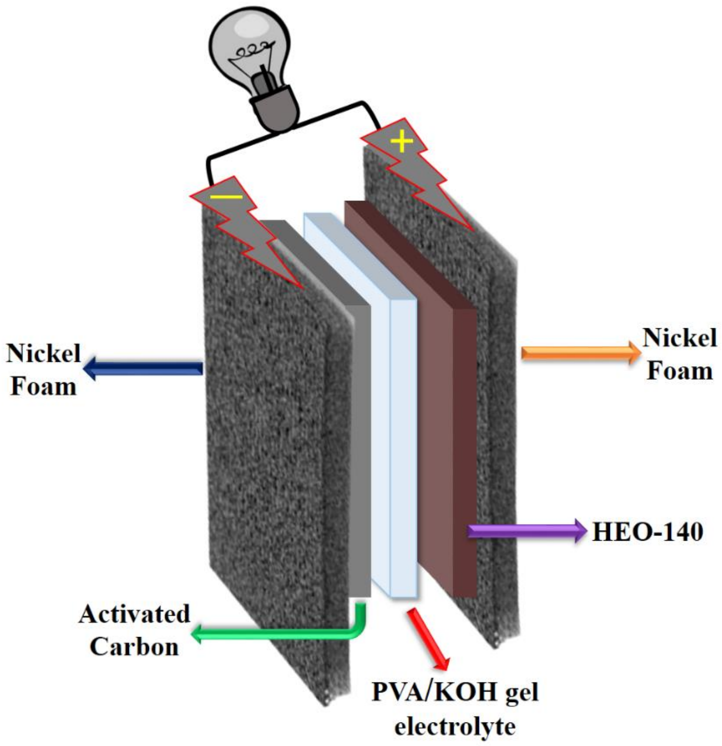

Fig. S10 A diagrammatic representation of the fabrication of the device.

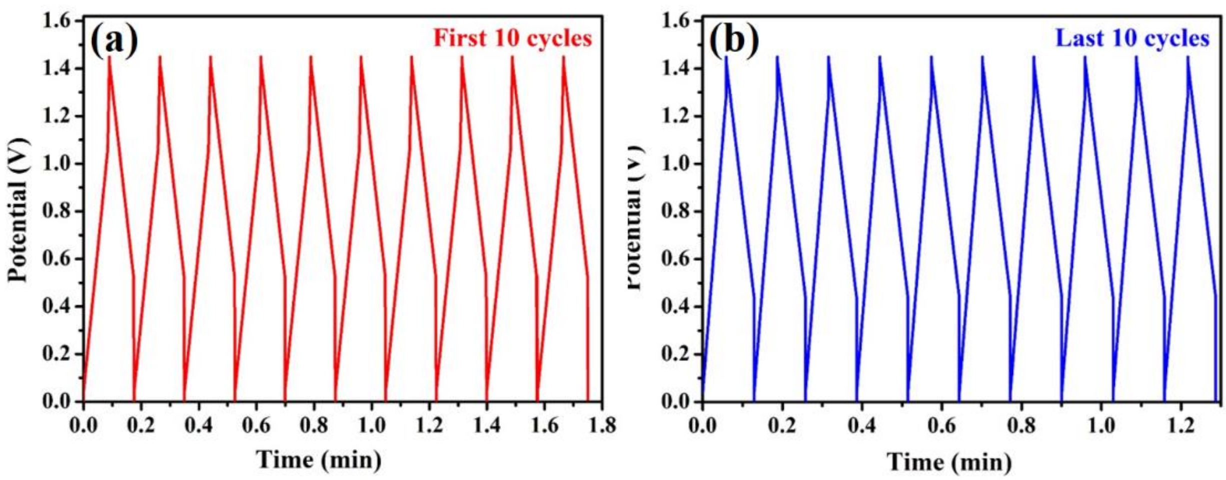

Fig. S11 The stability data in terms of first 10 cycles and last 10 GCD cycles.

Table S1 Average elemental percentage of five metal species in HEO-110, HEO-140 and HEO-170 determined by ICP-OES.

Materials	Mn (Elemental %)	Fe (Elemental %)	Co (Elemental %)	Ni (Elemental %)	Cu (Elemental %)
HEO-110	7.6	7.9	7.6	8.2	8.9
HEO-140	5.8	8.1	5.8	7.0	7.2
HEO-170	6.9	6.8	6.4	7.1	7.8

Table S2 Impedance parameter of the electrode material.

Electrode Material	Parameters		
	R_s (Ω)	R_{CT} (Ω)	C_L (F)
HEO-110	0.57	0.63	0.0038
HEO-140	0.49	0.56	0.0117
HEO-170	1.16	1.73	0.0047

Table S3 Comparison of energy parameters of HEO-140 electrode materials with other reported HEOs.

High Entropy Oxides [Method of synthesis]	Measurements					References
	Three-electrode		Two-electrode			
	Capacitance (Electrolyte)	**Retention (Cycle)**	**Capacitance**	**Energy density & Power density**	**Retention (Cycle)**	
$(FeCoCrMnNi)_3O_4$ [*PEG assisted HEO precursor followed by calcination (450 ºC)*]	204 F/g at 0.5 A/g (1M KOH)	60.4% (10,000)	149 F/g at 0.5 A/g	53 $Whkg^{-1}$ & 400.8 Wkg^{-1}	33% (10,000)	Ceramics International, 50 (7) (2024), 10292-10304
$(Al_{0.2}Co_{0.2}Cr_{0.2}Mn_{0.2}Ni_{0.2})_3O_4$ [*Reverse co-precipitation and calcination (550 ºC)*]	318 mAh/g at 1 A/g (3M KOH)	-----	-----	-----	-----	Chemistry Select, 7 (5) (2022),e202104015
$(CuNiFeMnCo)_3O_4$ **[*a)*** *Sol-gel and Combustion (350 ºC).* ***b)*** *Reverse co-precipitation and calcination (350 ºC)*]	a) 38.46F/g at 5 mV/s (2 M KOH) b) 34.24 F/g at 5 mV/s (2 M KOH)	a) 90% (1,000) b) 99% (1,000)	-----	-----	-----	Energy Storage, 6 (1) (2024), e538
$(Co_{0.2}Cr_{0.2}Fe_{0.2}Mn_{0.2}Ni_{0.2})_3O_4$ [*Polyacrylamide gel method followed by calcination (800 ºC)*]	384 F/g at 1 A/g (6 M KOH)	60% (2000)	75 F/ g at 1 A/g	24 $Whkg^{-1}$ & 746 Wkg^{-1}	45.6% (5000)	Journal of Energy Storage, 73 (2023), 109182
$(FeCoCrMnMg)_3O_4$ [*Solid-state reaction (900 ºC)*]	193.7 F/g at 1 A/g (1 M KOH)	51% (1000)	-----	-----	-----	Journal of Inorg. Materials, 36 (4) (2021), 425-430
$(CoCrFeMnNi)_3O_4$ [*Reverse co-precipitation and calcination (750 ºC)*]	239 F/g at 0.5 A/g (2 M KOH)	51% (1000)	-----	-----	-----	Journal of Energy Storage, 42 (2021), 103004
$(MnFeCoNiCu)_3O_4$ **[*Hydrothermal synthesis (140 ºC)*]**	**216.2 F/g at 1 A/g (3 M KOH)**	-----	**62 F/g at 1 A/g**	**25.17 $Whkg^{-1}$ & 1006.8 Wkg^{-1}**	**86.7% (5000)**	**This Work**