Supporting information

High-Entropy Oxide Synthesis in Concentrated Alkaline Solutions for Plasma-Catalytic Formaldehyde Oxidation

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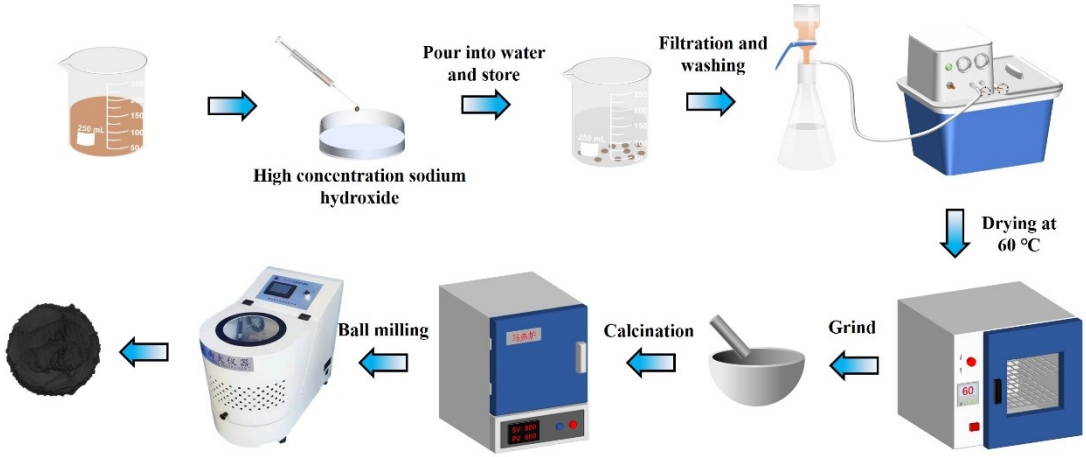

Fig. S1 Process flow diagram for the preparation of HEO catalysts via high-concentration sodium hydroxide coprecipitation method

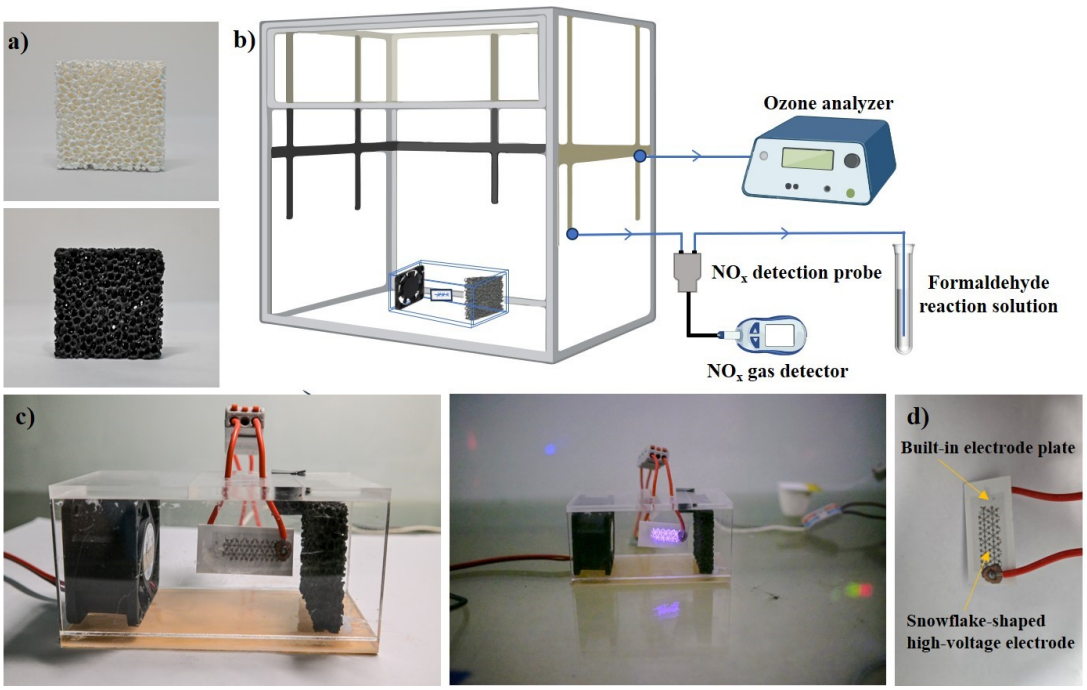

Fig. S2 a) Blank foam ceramics and foam ceramics loaded with HEO catalysts, b) Schematic diagram of the plasma catalytic degradation experimental setup for HCHO, c) Plasma catalytic system and an operational plasma catalytic system, d) Discharge electrode.

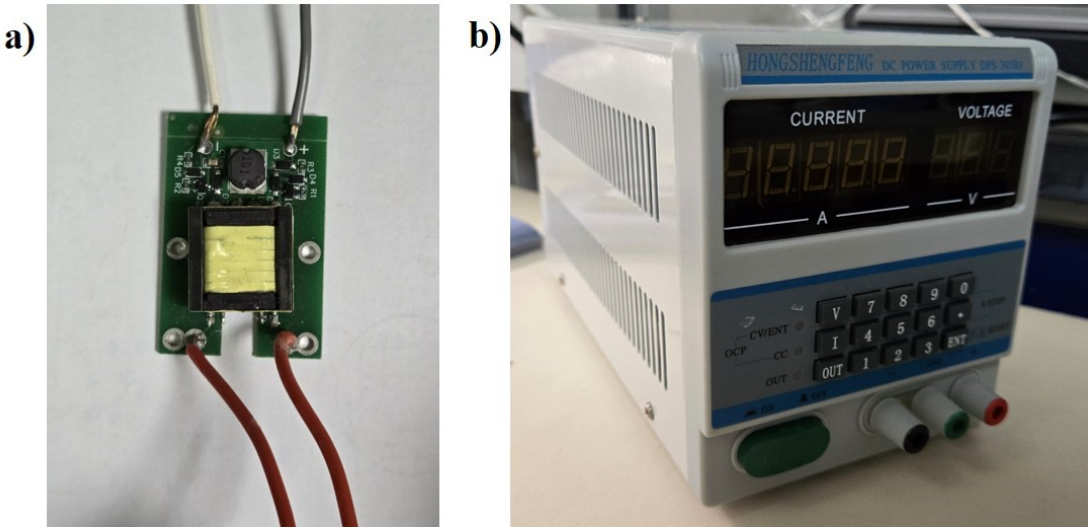

Fig. S3 a) Self-oscillating transformer, b) DC regulated power supply.

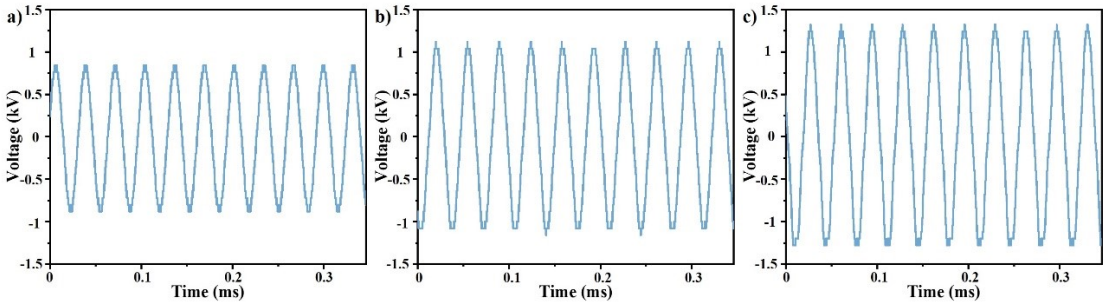

Fig. S4 Waveforms under different output voltages: a) 1.2 kV, b) 1.4kV, c)1.6 kV.

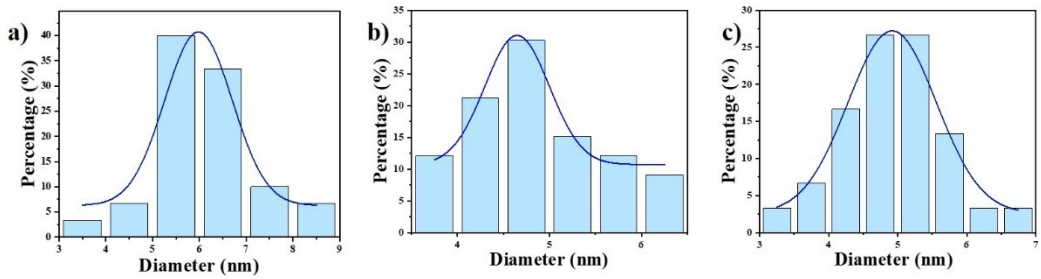

Fig. S5 Particle size distribution histogram of HEO catalysts prepared with different concentrations of sodium hydroxide: a) HEO-2 %-400, c) HEO-10 %-400, i) HEO-20 %-400.

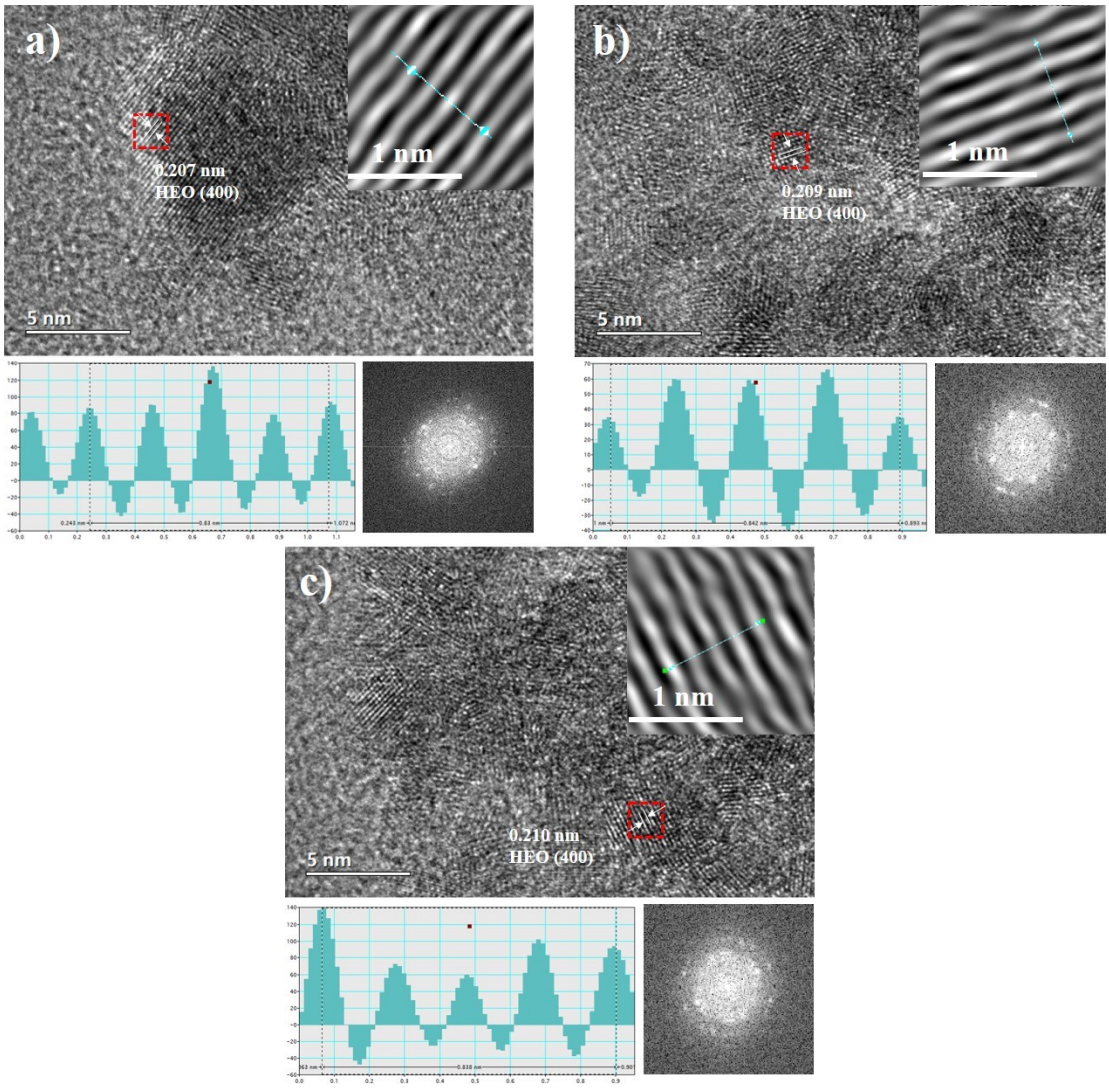

Fig. S6 HRTEM images of HEOs prepared with different sodium hydroxide concentrations (the inset shows the lattice spacing measurement within the red box and the corresponding diffraction spot pattern): a) HEO-2%-400, b) HEO-10%-400, c) HEO-20%-400.

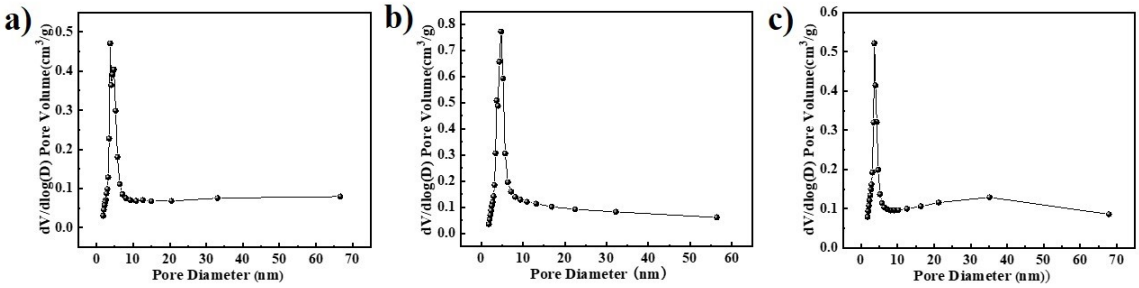

Fig. S7 Pore size distribution curves: a) HEO-2 %-400, b) HEO-10 %-400, c) HEO-20 %-400.

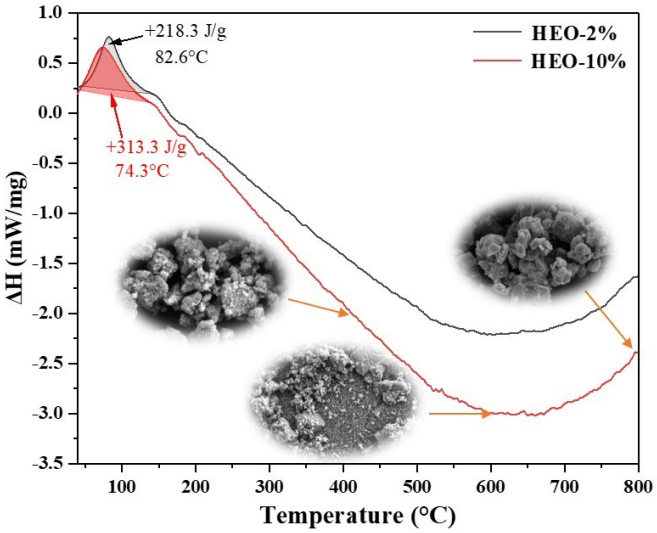

Fig. S8 DSC analysis of phase transition in HEO catalyst at different temperatures.

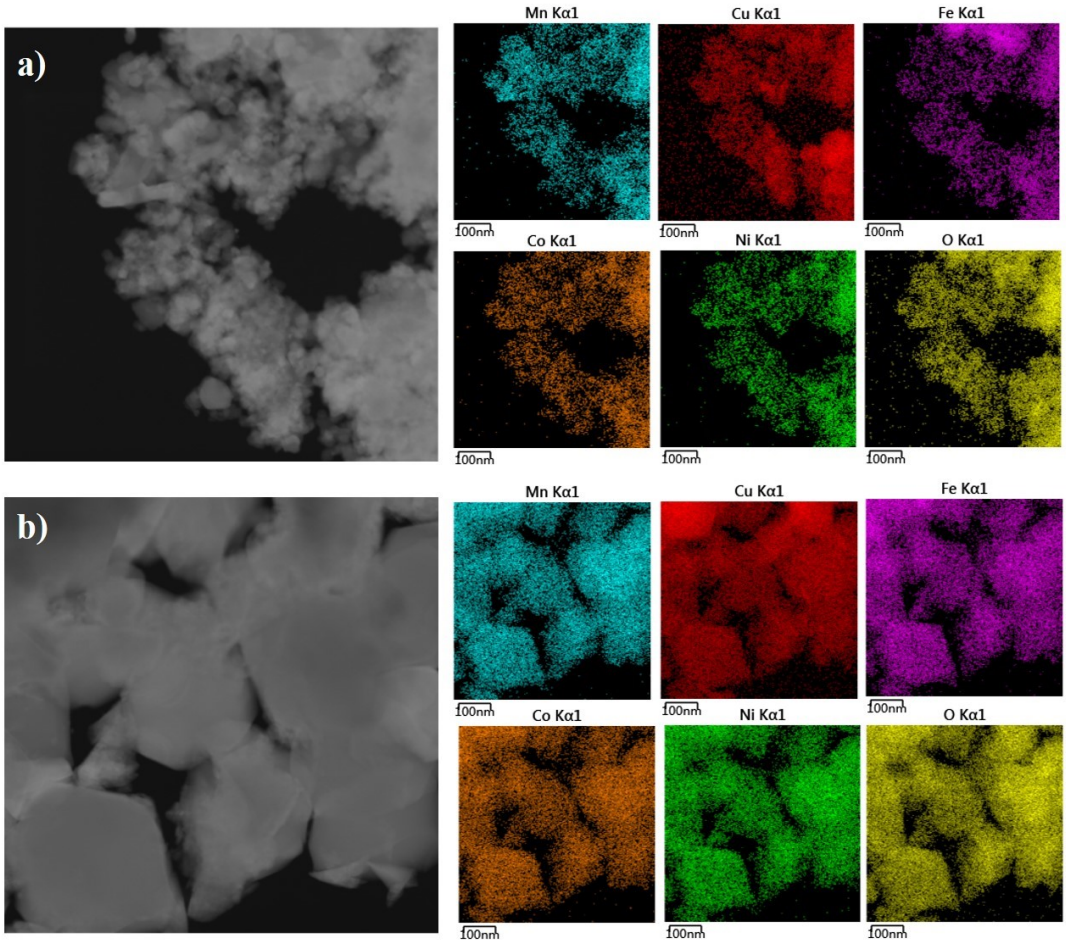

Fig. S9 EDS elemental mapping images of the a) HEO-10 %-600, b) HEO-10 %-800.

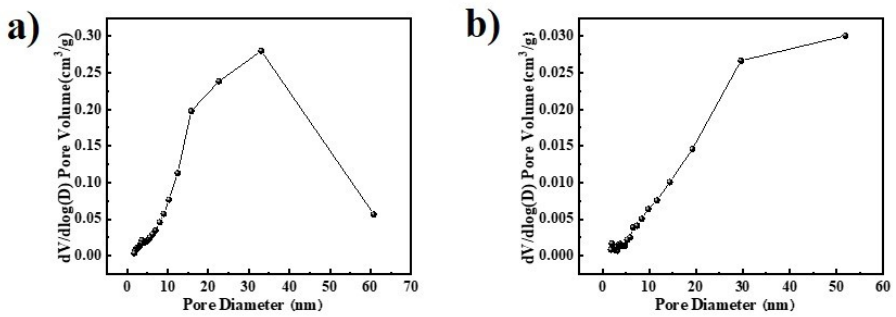

Fig. S10 Pore size distribution curves: a) HEO-10%-600, b) HEO-10%-800.

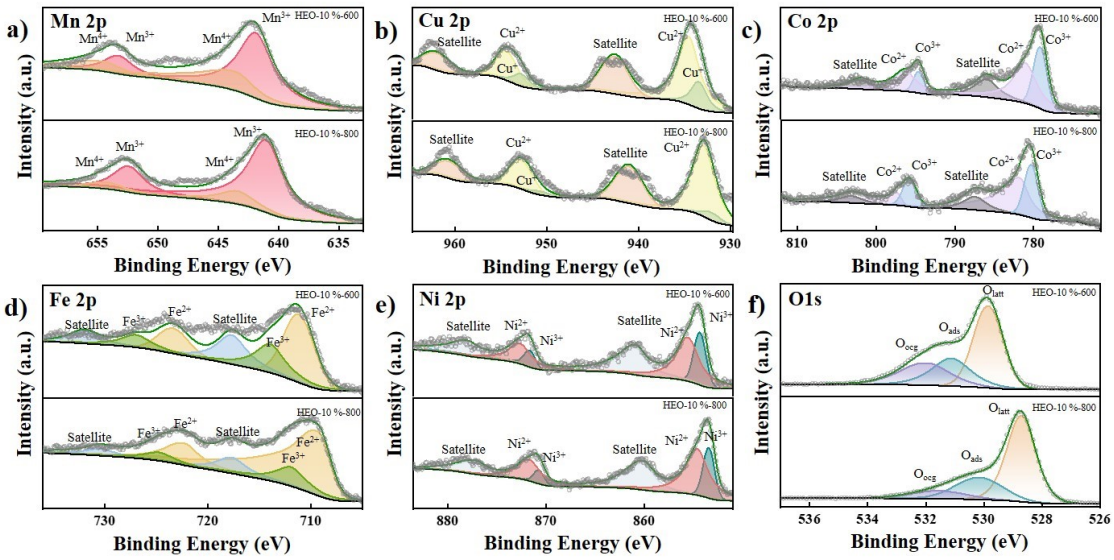

Fig. S11 XPS spectra of HEO catalysts with different calcination temperatures: a) Mn 2p; b) Cu 2p; c) Co 2p; d) Fe 2p; e) Ni 2p; f) O 1s.

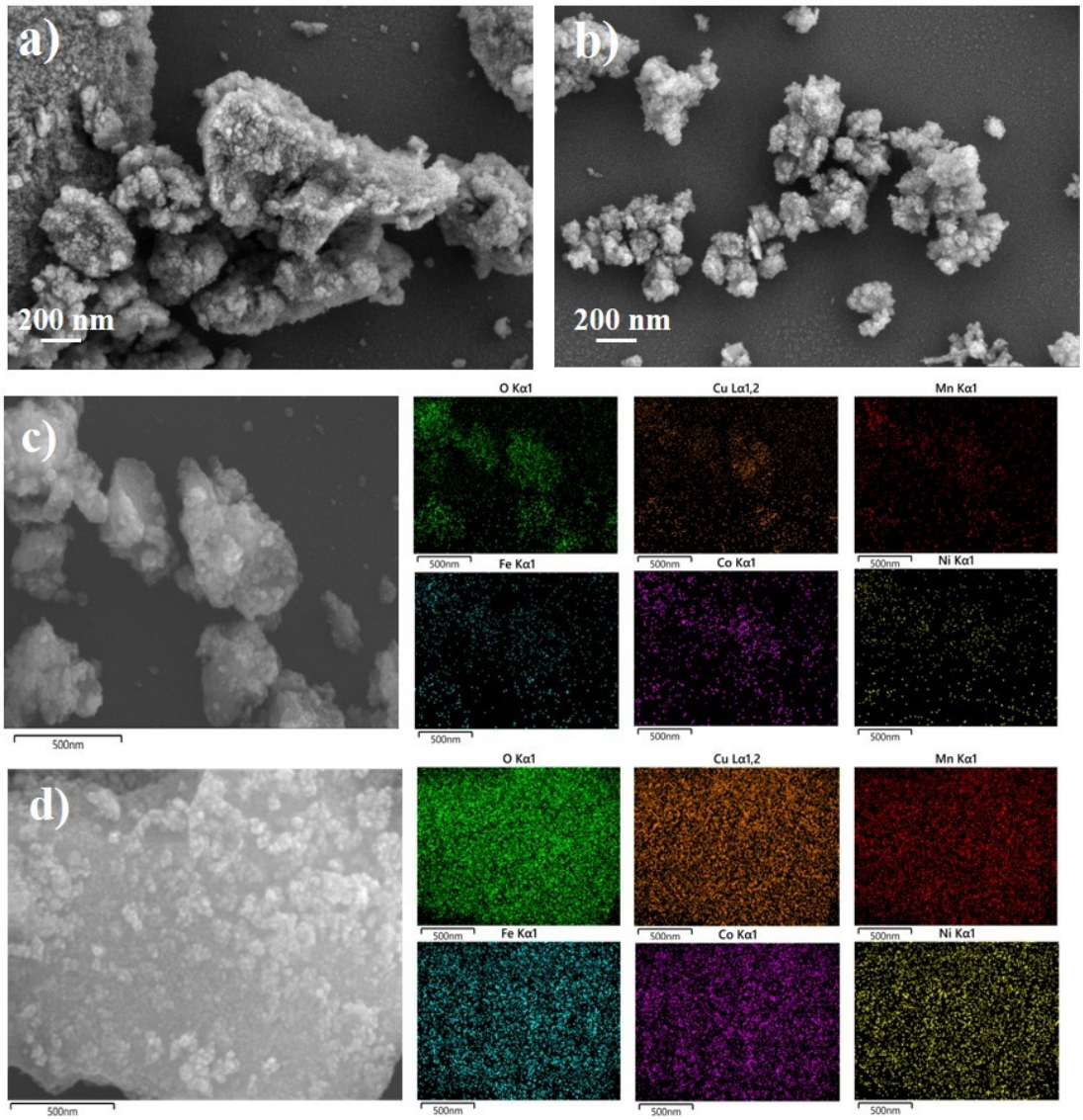

Fig. S12 SEM images of the a) HEO-2% and b) HEO-10 %; EDS elemental mapping images of the c) HEO-2 %, d) HEO-10 %.

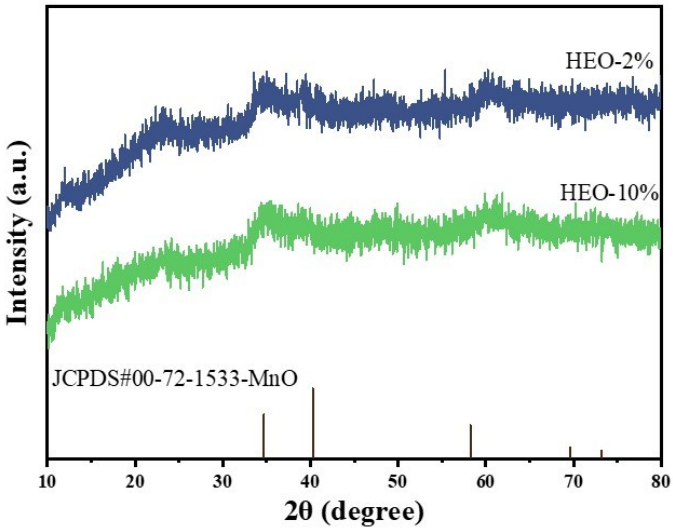

Fig. S13 XRD pattern of the naturally air-dried material.

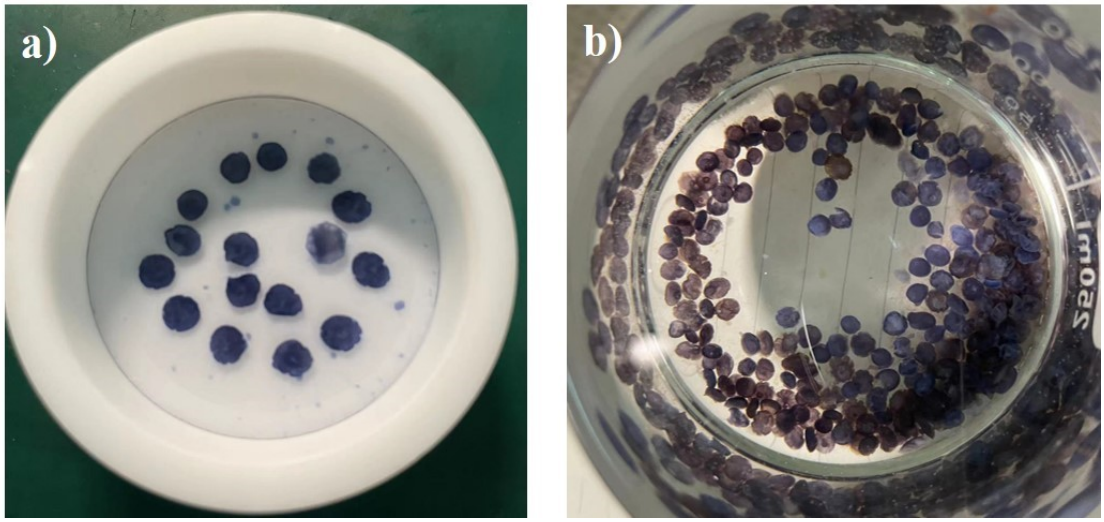

Fig. S14 a) Precipitation reaction in a polytetrafluoroethylene container, b) Collected precipitate.

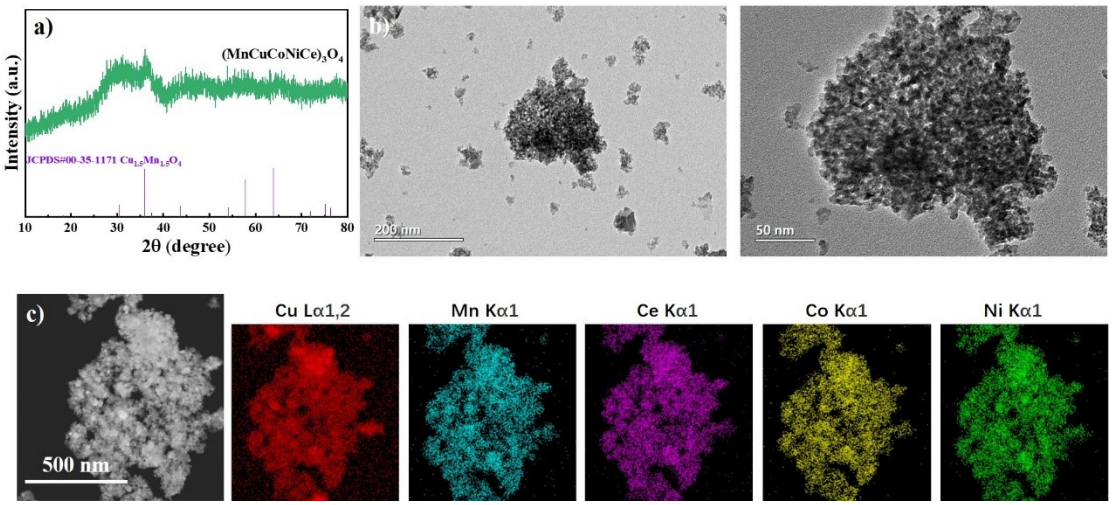

Fig. S15 $(MnCuCoNiCe)_3O_4$: a) XRD pattern, b) TEM image, c) EDS elemental mapping images.

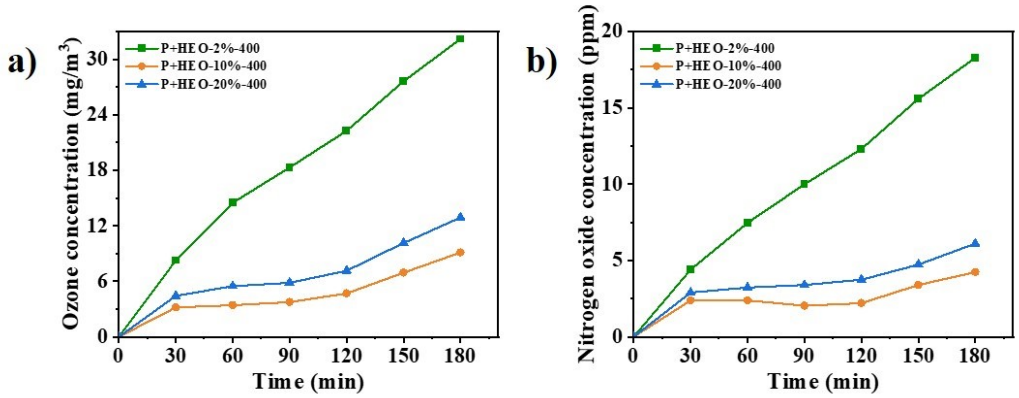

Fig. S16 The by-product yield of HEO catalysts prepared under different conditions at 1.6 kV, 60% RH, and 18 m^3/h gas flow rate: a) O_3 yield; b) NO_x yield.

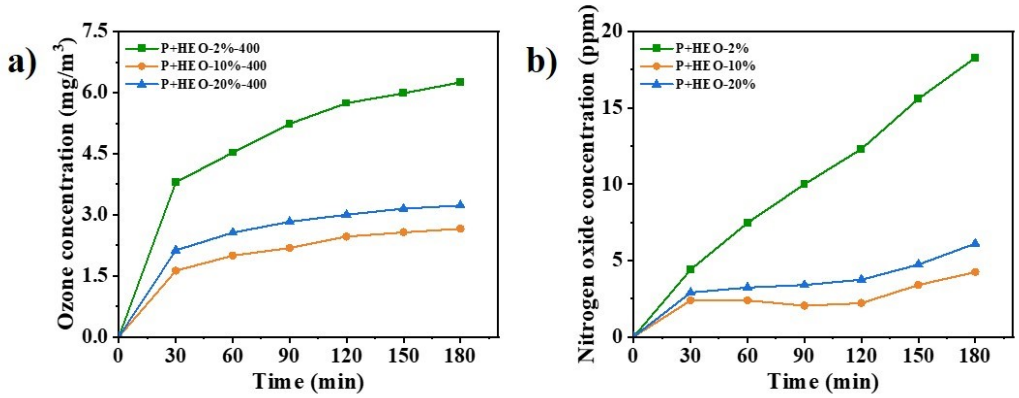

Fig. S17 The by-product yield of HEO catalysts prepared under different conditions at 1.4 kV, 80% RH, and 18 m^3/h gas flow rate: a) O_3 yield, b) NO_x yield.

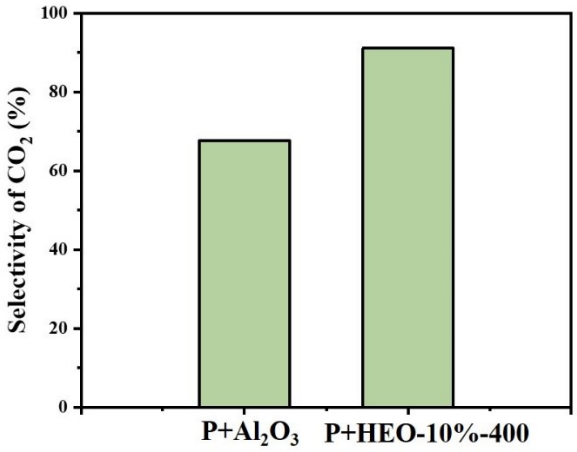

Fig. S18 The CO_2 selectivity of different combinations.

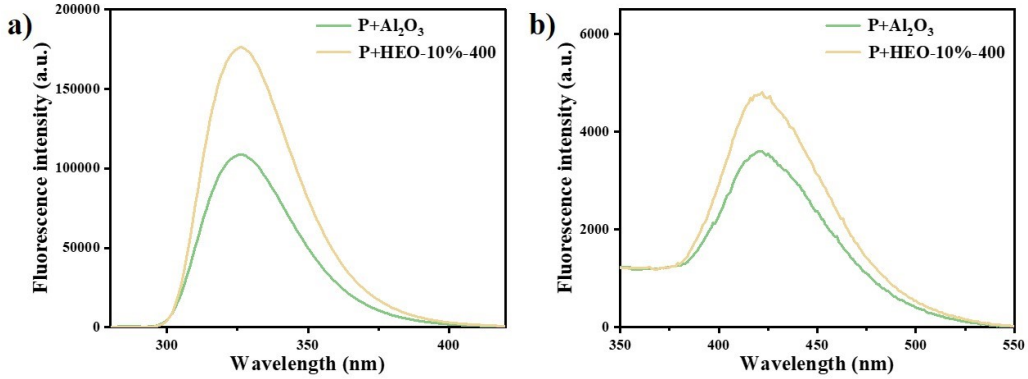

Fig. S19 a) Fluorescence intensity plot of $O_2^{-\cdot}$ captured by different combinations λ_e=260 nm, b) Fluorescence intensity plot of HO· captured by different combinations λ_e=315 nm. Output voltage = 1.4 kV, Temperature = 25 °C, Humidity = 60 %RH.

Table. S1 Plasma system electrical parameters

DC Power Supply		Self-Excited Oscillation Transformer		Discharge Ceramic Plate	
Input Voltage (V)	Input Current (A)	Output Voltage (kV)	Output Current (mA)	Positive Ion Count (*10000)	Negative Ion Count (*10000)
6	0.18	1.2	0.16	41	39
6.5	0.22	1.4	0.18	78	79
7	0.26	1.6	0.21	114	108

Table. S2 The results of the simulated crystalline size calculation.

Sample	HEO-2%-400	HEO-10%-400	HEO-20%-400	HEO-10%-600	HEO-10%-800
Size (nm)	2.4	1.8	1.9	23.7	167.6

K=0.9, λ=0.154 nm

Table. S3 EDS elemental analysis results

Elemental content (wt.%) / Sample	Mn	Cu	Co	Fe	Ni
HEO-2 %-400	15.6	16.0	15.7	15.3	15.8
HEO-10 %-400	15.7	16.2	15.8	15.4	15.7
HEO-20 %-400	15.5	16.8	15.6	14.5	15.7

Table. S4 Structural properties of synthesized HEO catalysts

Sample	Specific surface area (m^2/g)	pore volume (cm^3/g)	average pore diameter (nm)
HEO-10 %-600	46	0.23	19.7
HEO-10 %-800	8	0.04	28.9

Table. S5 Summary table of XPS analysis results of HEO catalysts at different calcination temperatures

Sample	Mn^{3+}/Mn^{4+}	Cu^{+}/Cu^{2+}	Co^{2+}/Co^{3+}	Fe^{2+}/Fe^{3+}	Ni^{2+}/Ni^{3+}	$O_{ads}/(O_{latt}+O_{ads})$
HEO-10 %-600	2.27	0.21	1.56	1.88	2.22	0.33
HEO-10 %-800	5.88	0.10	1.96	3.65	2.56	0.29

Table. S6 ICP test results for the content of each element

Sample	Element	theoretical content (wt.%)	actual content (wt.%)
$(MnCuCoNiCe)_3O_4$	Mn	15.8	15.6
	Cu	15.8	15.5
	Co	15.8	15.7
	Ni	15.8	15.9
	Ce	15.8	16.2