Supplementary Information (SI) for Green Chemistry.
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Supplementary Materials

Ethanol-assisted mechanochemical synthesis of MOF-199-derived CuOx/carbon composites with tunable copper species for photo-enhanced Fenton-like dye degradation

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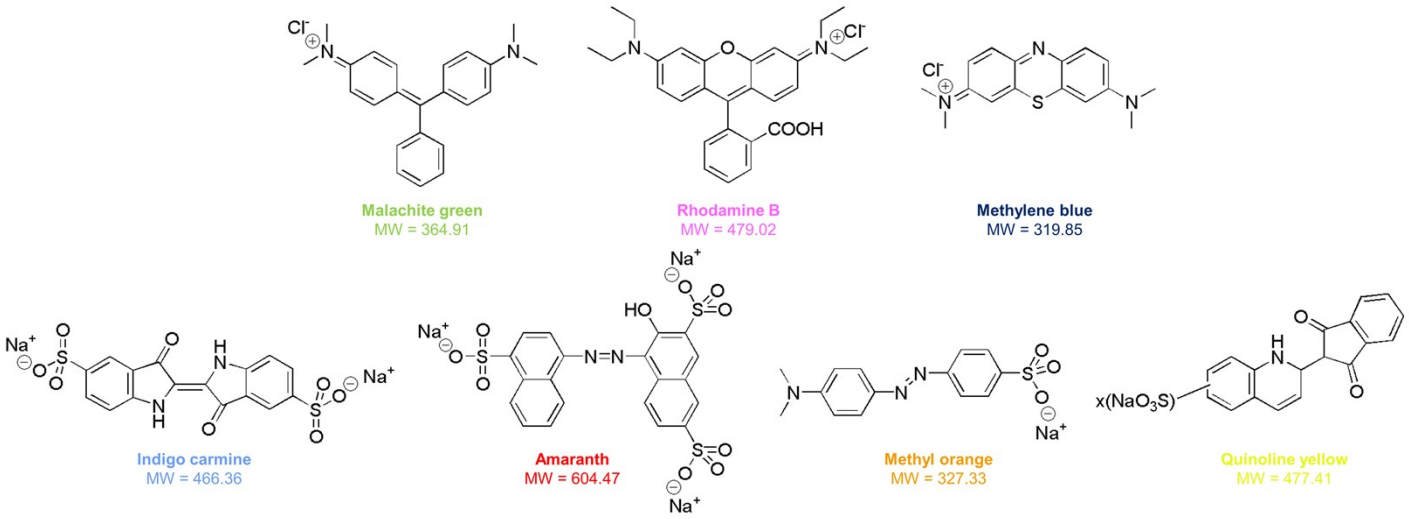

Figure S1. Different organic dyes used in this study and their chemical structures.

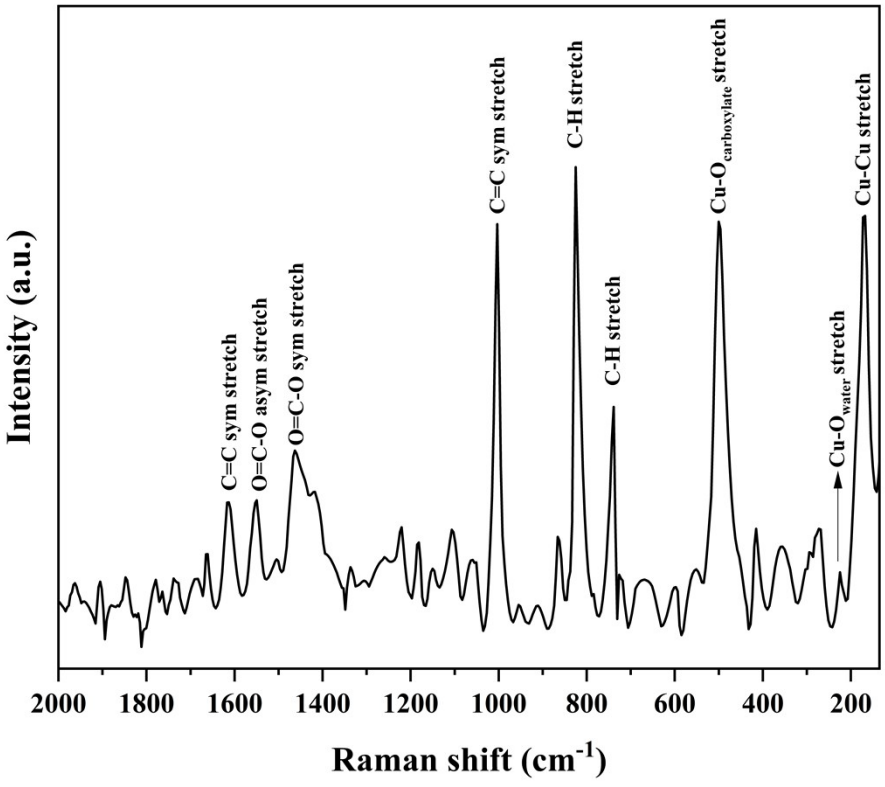

Figure S2. Raman spectrum of MOF-199.

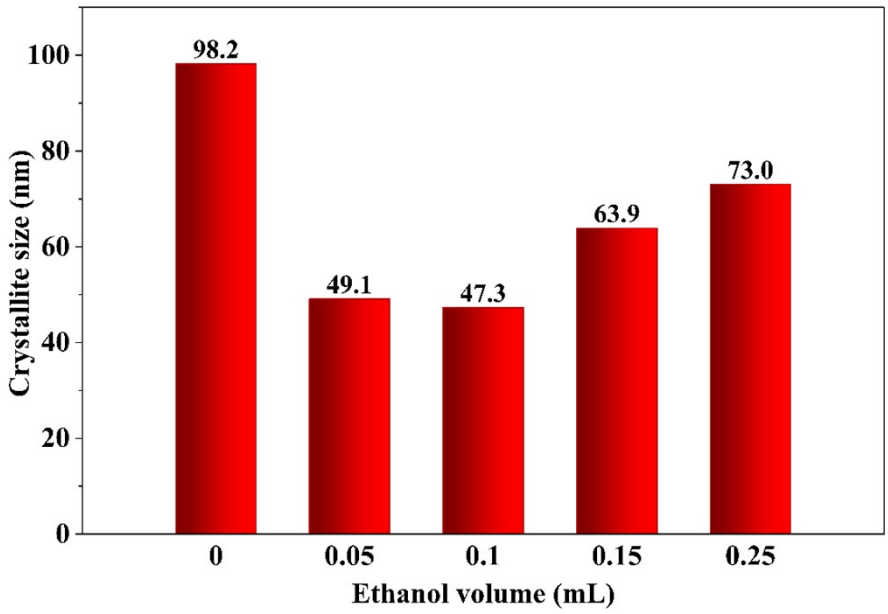

Figure S3. Crystallite size of ethanol volume-varied MOF-199 samples.

The ethanol-volume-varied MOF-199 samples were subsequently pyrolyzed at 700 °C to obtain Cu_xO_y/C-700-V (where V denotes the ethanol volume added during the grinding process) to investigate the influence of ethanol on copper phase evolution after carbonization. The relative contents of Cu, Cu_2O, and CuO were quantified from the XRD patterns (**Fig. S4**) using the Rietveld refinement method.

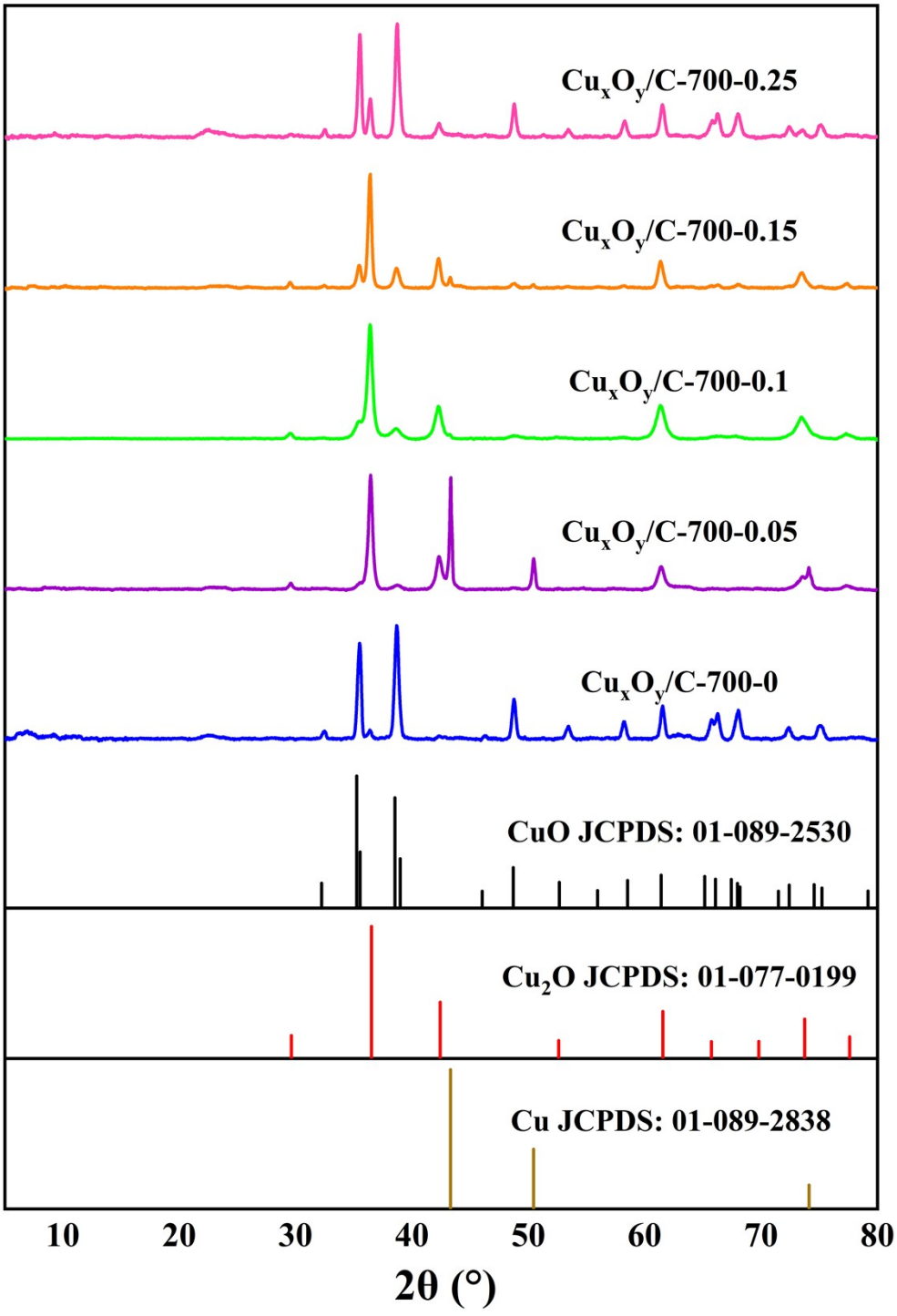

Figure S4. XRD patterns of Cu_xO_y/C-700-V samples.

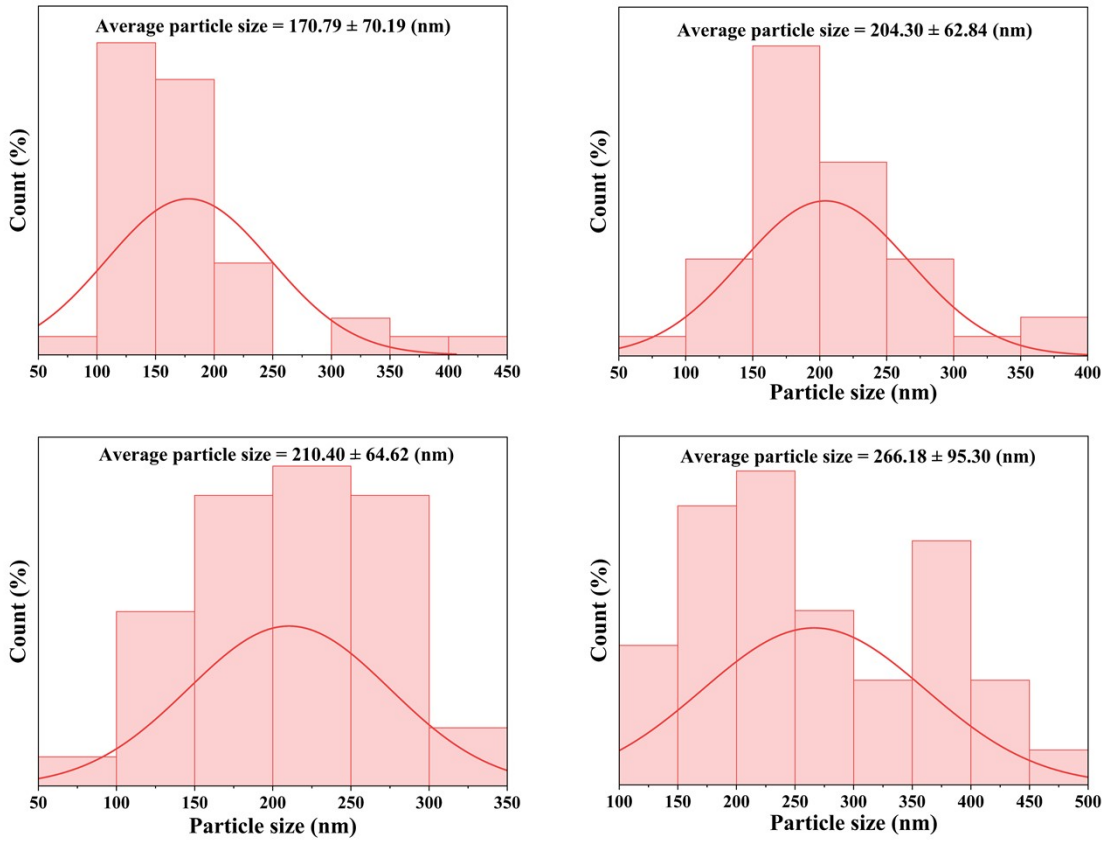

Figure S5. Average particle size of Cu_xO_y/C samples at varied pyrolysis temperatures: a) 500 °C, b) 600 °C, c) 700 °C, d) 800 °C.

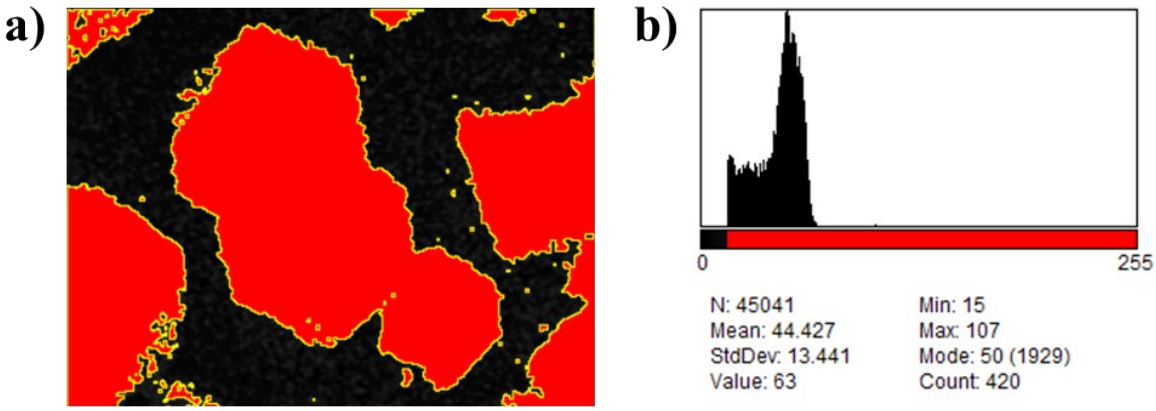

Figure S6. (a) EDS mapping and (b) intensity histogram after processing for statistical analysis.

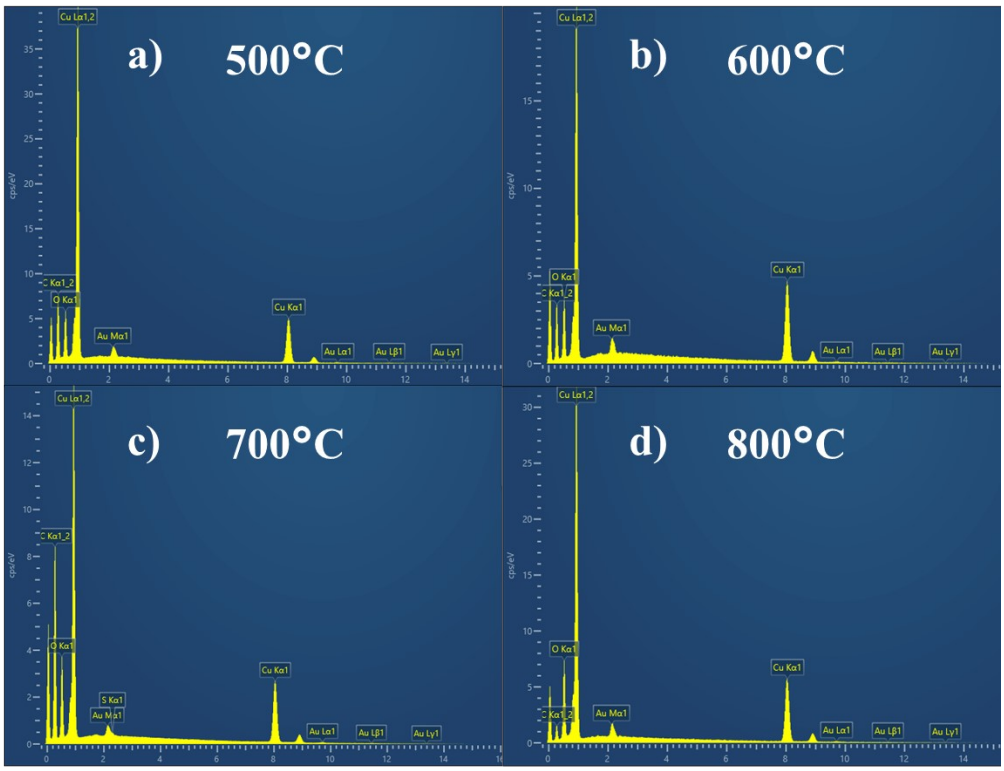

Figure S7. EDX spectrum of Cu_xO_y/C samples at varied pyrolysis temperatures: a) 500 °C, b) 600 °C, c) 700 °C, d) 800 °C.

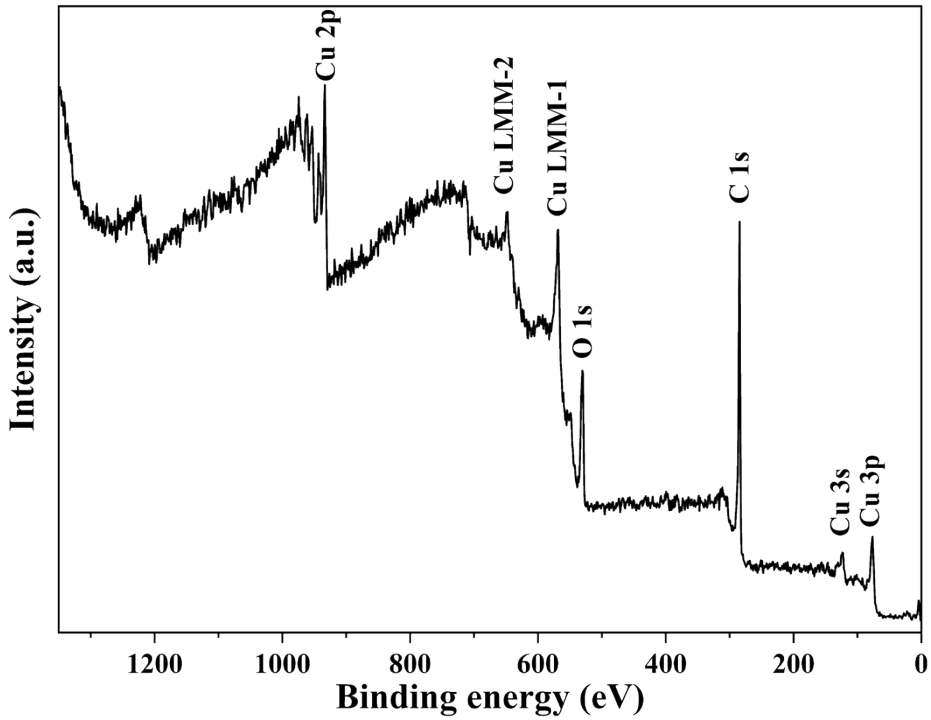

Figure S8. XPS survey spectrum of Cu_xO_y/C-700 sample.

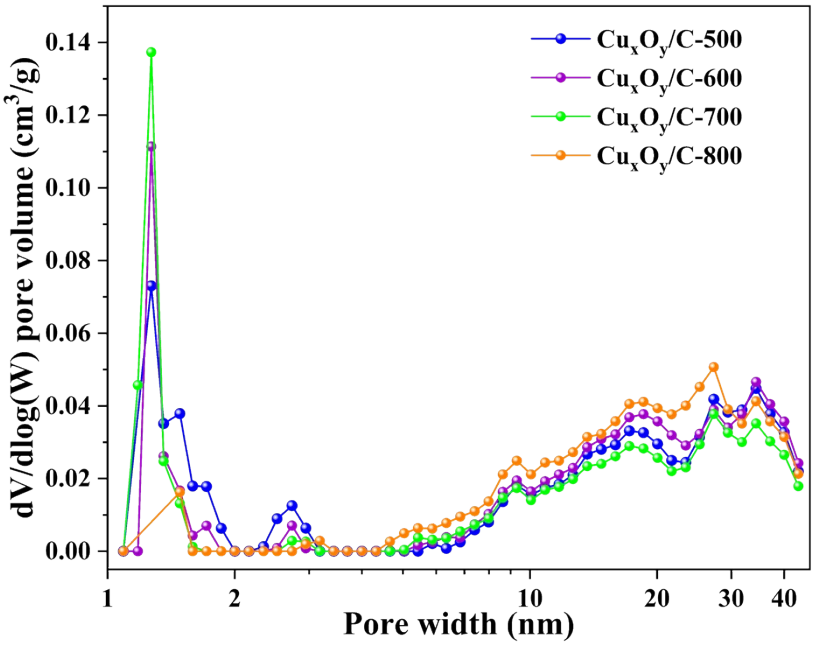

Figure S9. Pore size distribution of Cu_xO_y/C samples at varied pyrolysis temperatures.

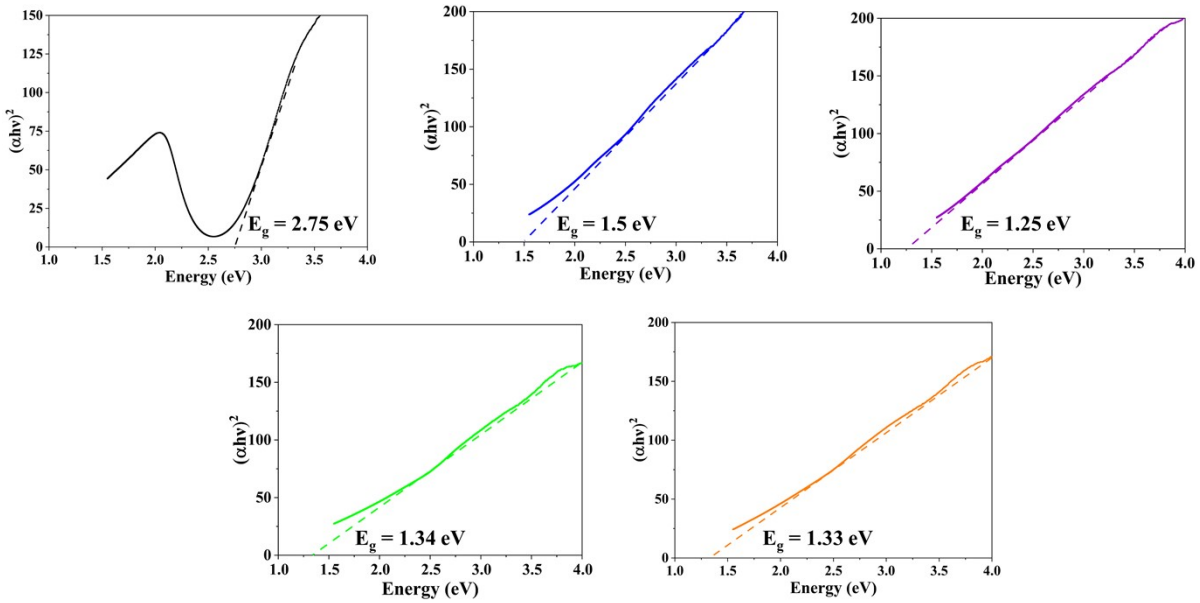

Figure S10. Tauc plots and extrapolated direct E_g data of a) MOF-199 and Cu_xO_y/C samples pyrolysis at varied temperatures: b) 500 °C, c) 600 °C, d) 700 °C, e) 800 °C.

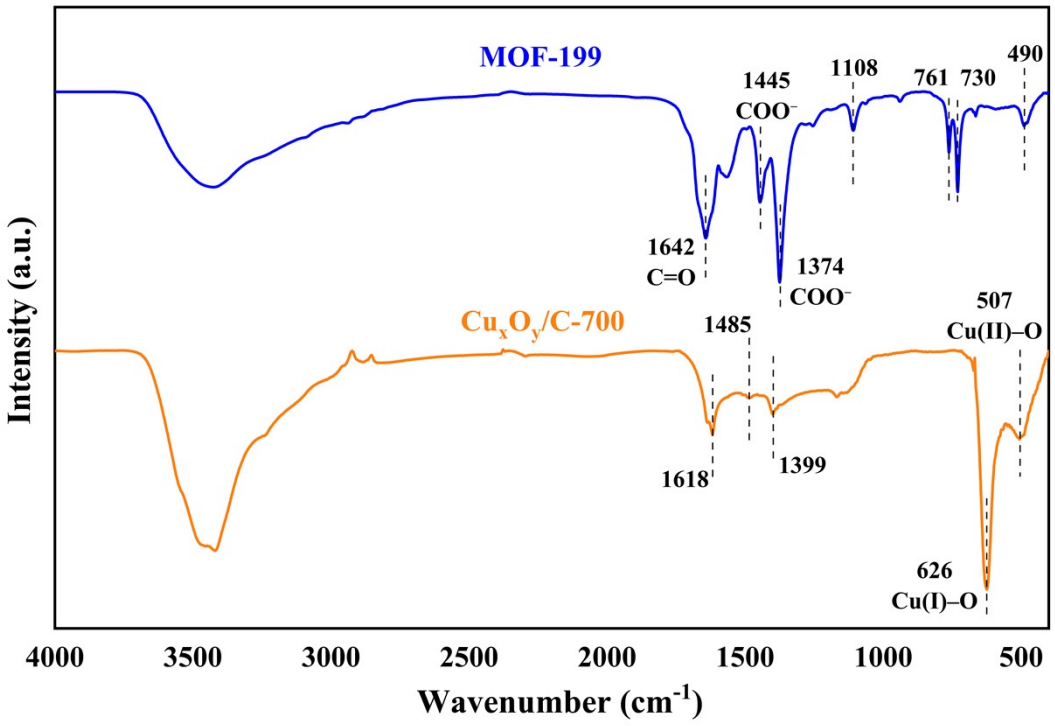

Figure S11. FTIR spectra of MOF-199 and Cu_xO_y/C-700.

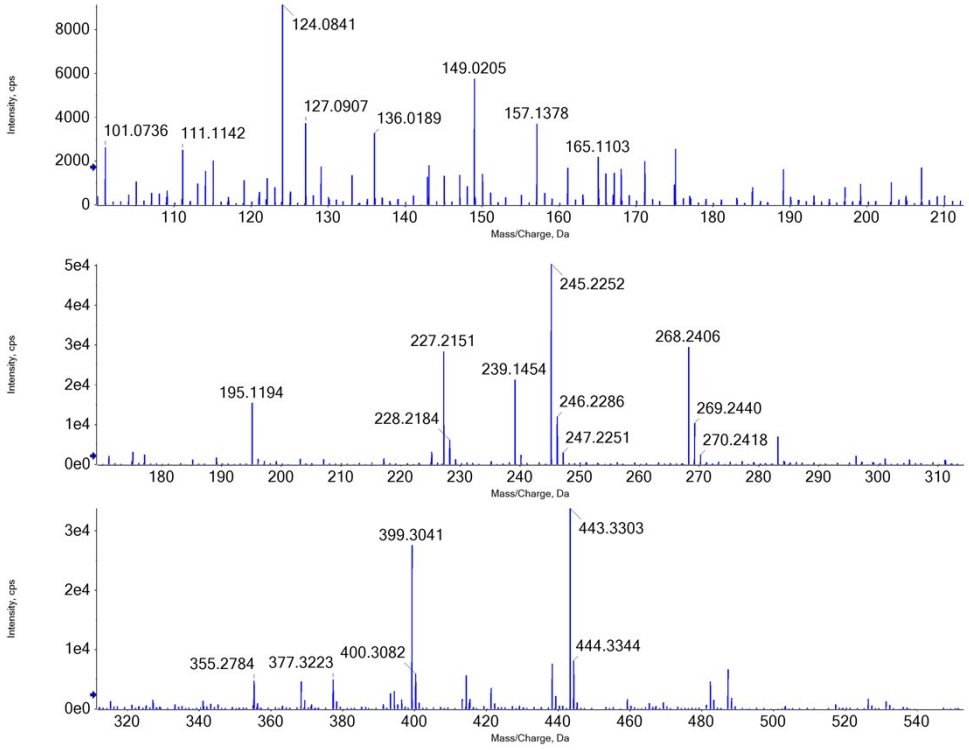

Figure S12. Mass spectra of intermediates in the solution after 60 min of degradation by Cu_xO_y-C/700. Reaction conditions: $[dye]_0$ = 10 mg/L, $[H_2O_2$ 30%] = 6 mL/L, [catalyst] = 0.04 g/L, and room temperature.

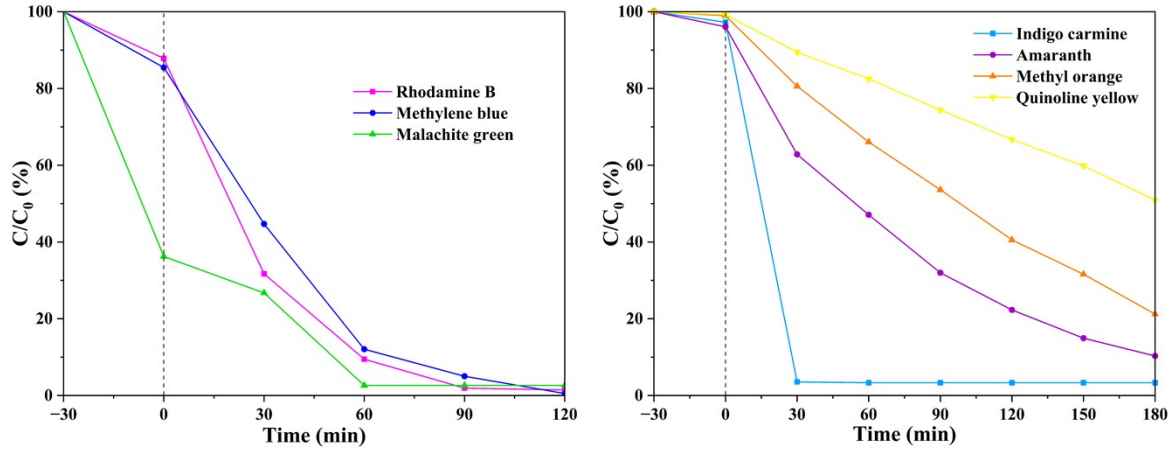

Figure S13. Photo-Fenton catalytic activity of Cu_xO_y/C-700 on the degradation of a) cationic dyes and b) anionic dyes. Reaction conditions: $[dye]_0$ = 10 mg/L, $[H_2O_2$ 30%] = 6 mL/L, [catalyst] = 0.04 g/L, and room temperature.

Table S1. Copper species composition, total copper content, and textural properties of Cu_xO_y/C samples at varied pyrolysis temperatures.

	Copper species composition[a] (%)			BET surface area (m^2/g)	total pore volume[b] (cm^3/g)	V_{micro}[b] (cm^3/g)	V_{meso}[b] (cm^3/g)	copper content[c] (%)
	Cu	Cu_2O	CuO					
Cu_xO_y/C-500	31.5	51.8	16.6	123.02	0.059	0.030	0.029	55.1
Cu_xO_y/C-600	47.6	40.7	11.6	123.25	0.061	0.030	0.031	58.2
Cu_xO_y/C-700	0.5	82.2	17.3	116.19	0.053	0.026	0.027	68.8
Cu_xO_y/C-800	0.4	25.0	74.6	56.43	0.040	0.013	0.027	70.6

[a]from Rietveld refinement of the XRD patterns.
[b]from DFT analysis of N_2 adsorption-desorption isotherms.
[c]from TGA curves.

Table S2. Comparison of the solvothermal method and ethanol-assisted grinding method.

	Solvothermal method	**Ethanol-assisted grinding method**
$Cu(NO_3)_2.3H_2O$	0.438 g	1.533 g
H_3BTC	0.236 g	0.826 g
Synthesis time	24 h	0.5 h
Synthesis temperature	85 °C	Room temperature
Solvent	3 mL DMF : 4 mL ethanol : 2 mL H_2O	0.1 mL ethanol
Washing solvent	3×8 mL DMF and 3×8 mL ethanol	3×3 mL ethanol
Yield (based on H_3BTC)	84%	82%
Solvent per gram of MOF-199	277.4 mL g^{-1}	13.3 mL g^{-1}