

**Room-temperature fabrication of defective  $\text{CoO}_x\text{H}_y$  nanosheets with abundant oxygen vacancies and high porosity as efficient 5-hydroxymethylfurfural oxidation electrocatalysts**

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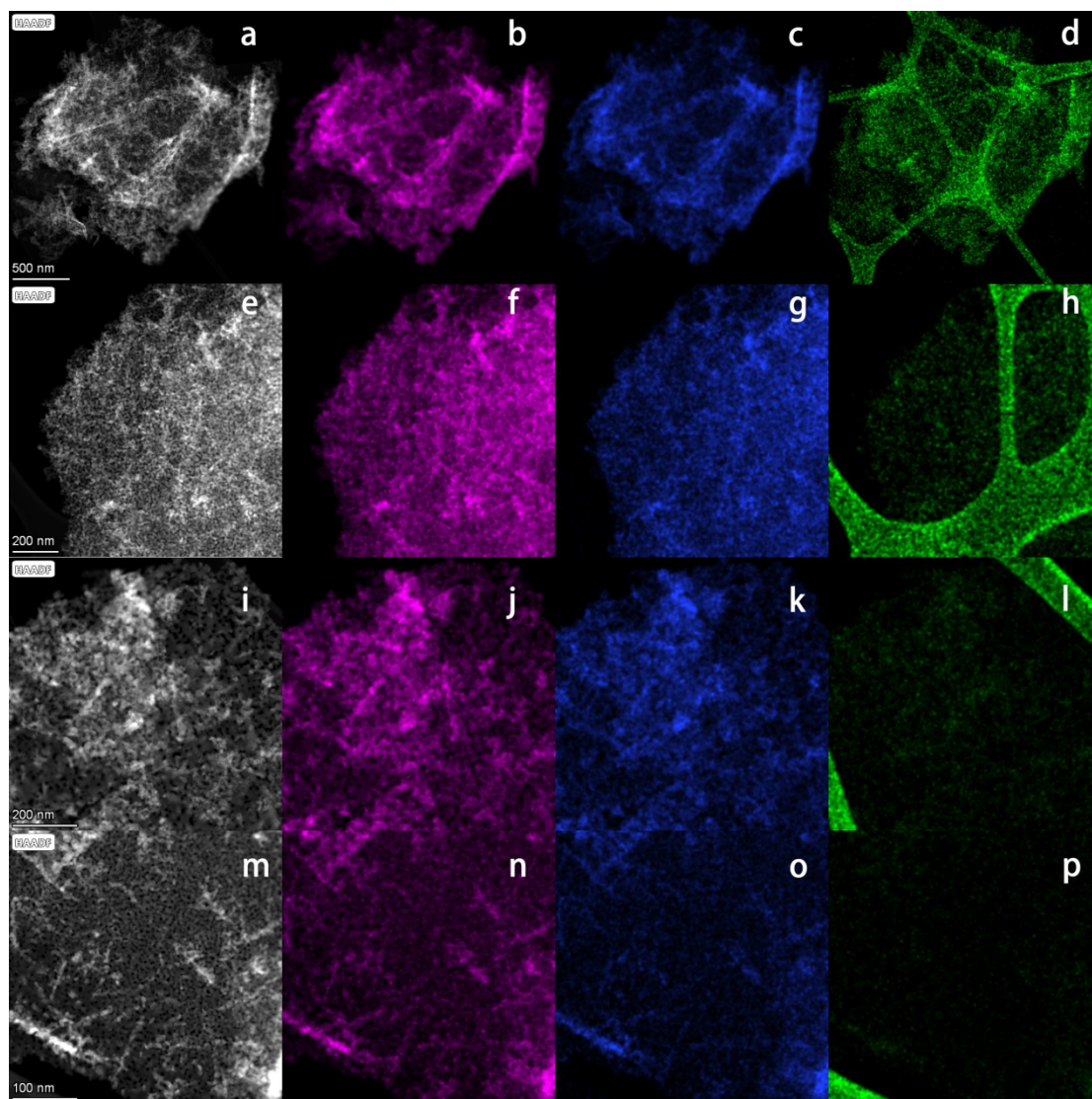
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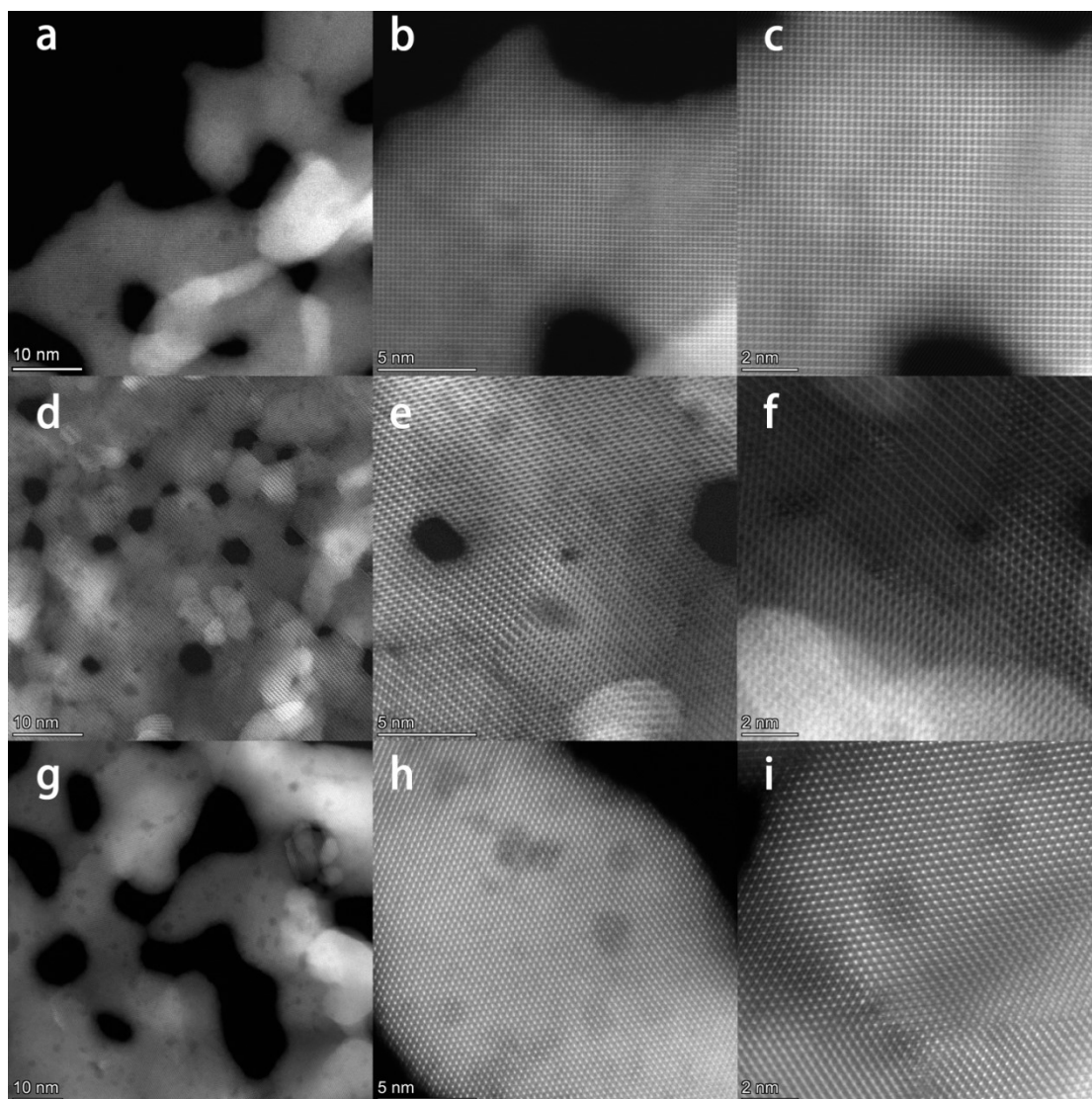
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**Table S1.** The fitted parameters of Nyquist plots of CoO<sub>x</sub>H<sub>y</sub>, CoO<sub>x</sub>H<sub>y</sub>-MA, CoO<sub>x</sub>H<sub>y</sub>-BH, and CoO<sub>x</sub>H<sub>y</sub>-MA/BH.

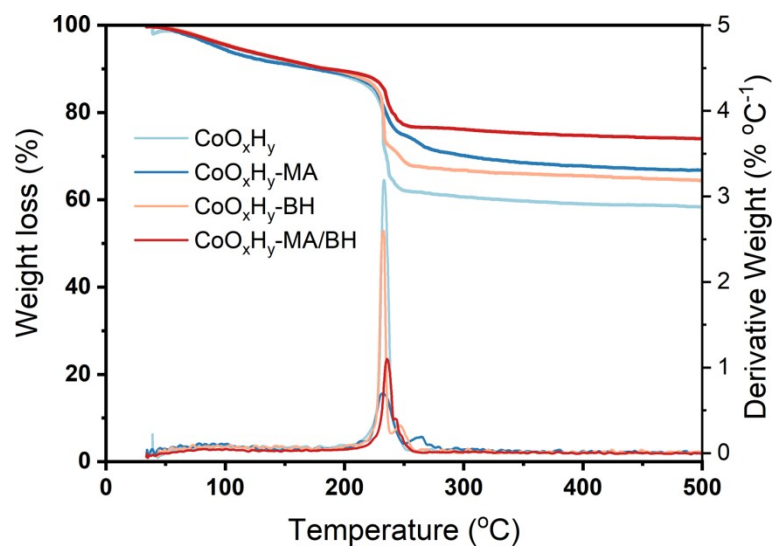
Catalyst	R <sub>s</sub> (Ω)	R <sub>ct</sub> (Ω)	CPE-T (F)	CPE-P
CoO <sub>x</sub> H <sub>y</sub>	11.6	36.7	0.0006	0.80
CoO <sub>x</sub> H <sub>y</sub> -MA	10.9	8.1	0.0035	0.88
CoO <sub>x</sub> H <sub>y</sub> -BH	10.9	15.1	0.0026	0.77
CoO <sub>x</sub> H <sub>y</sub> -MA/BH	12.6	11.1	0.0022	0.81



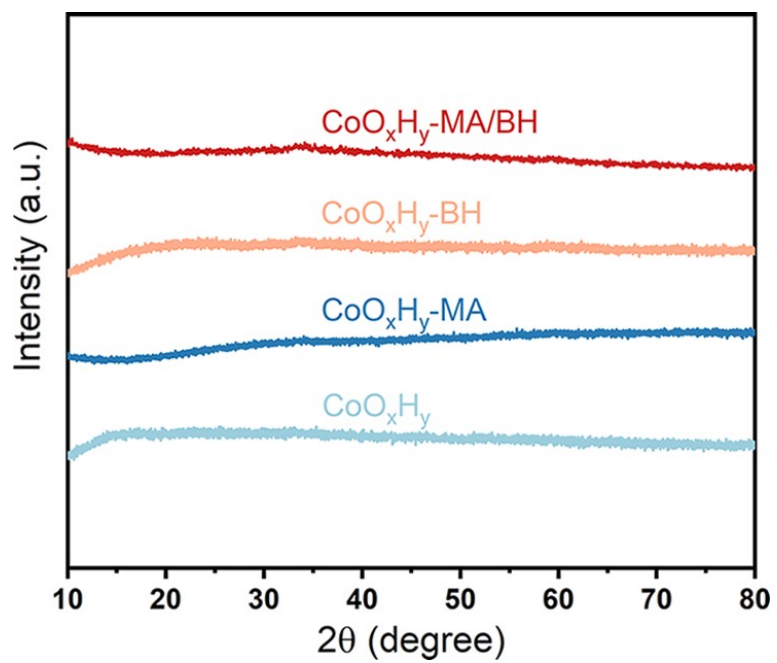
**Figure S1.** (a, e, i, m) The HAADF-STEM images, and the corresponding EDS mappings of (b-d)  $\text{Co}_3\text{O}_4$ -300, (f-h)  $\text{Co}_3\text{O}_4$ -300-MA, (j-l)  $\text{Co}_3\text{O}_4$ -300-BH, and (n-p)  $\text{Co}_3\text{O}_4$ -300-MA/BH. The Co, O, and C elements were represented in the color of magenta, blue, and green, respectively.



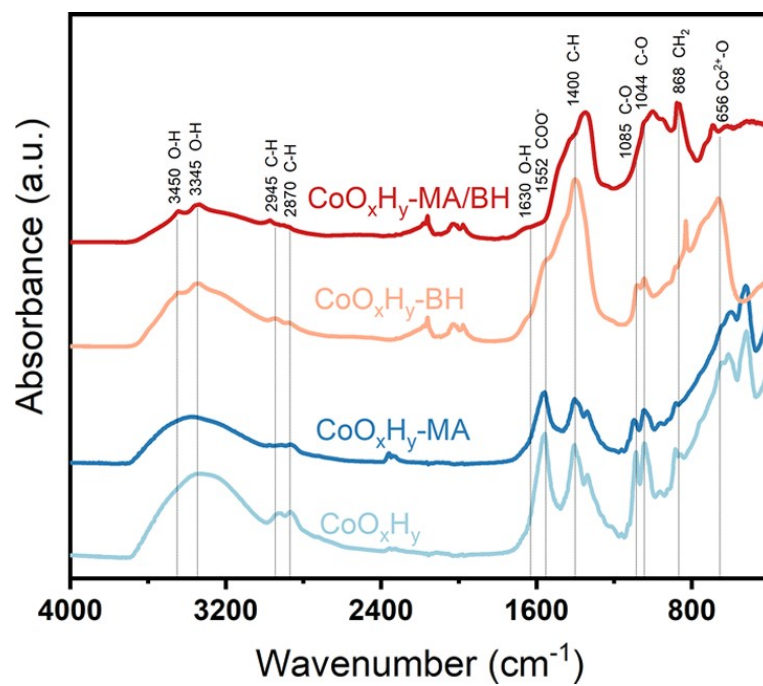
**Figure S2.** HRTEM images with increased magnifications of (a-c)  $\text{Co}_3\text{O}_4\text{-300-MA}$ , (d-f)  $\text{Co}_3\text{O}_4\text{-300-MA}$ , and (g-i)  $\text{Co}_3\text{O}_4\text{-300-MA/BH}$ .



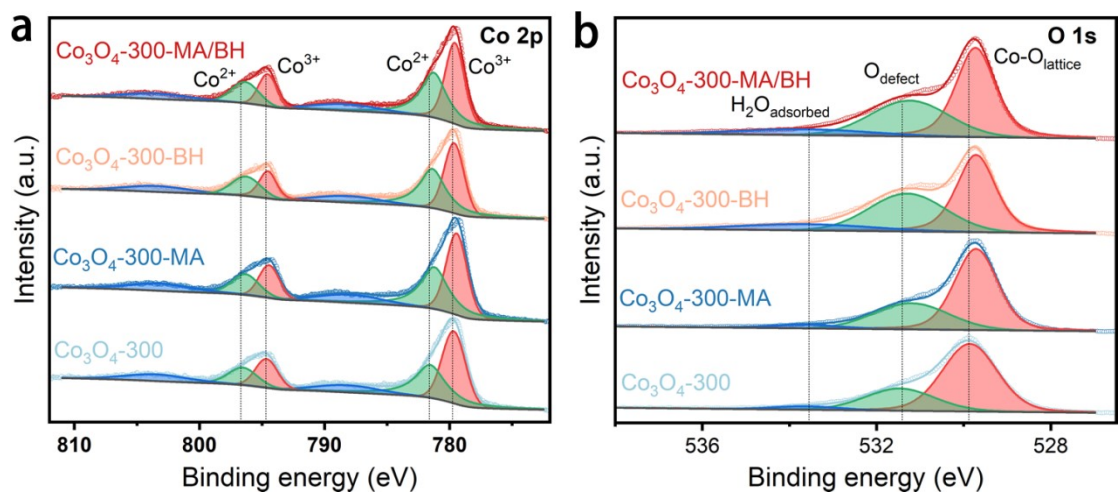
**Figure S3.** TGA/DTG profiles of  $\text{CoO}_x\text{H}_y$ ,  $\text{CoO}_x\text{H}_y\text{-MA}$ ,  $\text{CoO}_x\text{H}_y\text{-BH}$ , and  $\text{CoO}_x\text{H}_y\text{-MA/BH}$ .



**Figure S4.** XRD patterns of  $\text{CoO}_x\text{H}_y$ ,  $\text{CoO}_x\text{H}_y\text{-MA}$ ,  $\text{CoO}_x\text{H}_y\text{-BH}$ , and  $\text{CoO}_x\text{H}_y\text{-MA/BH}$ .

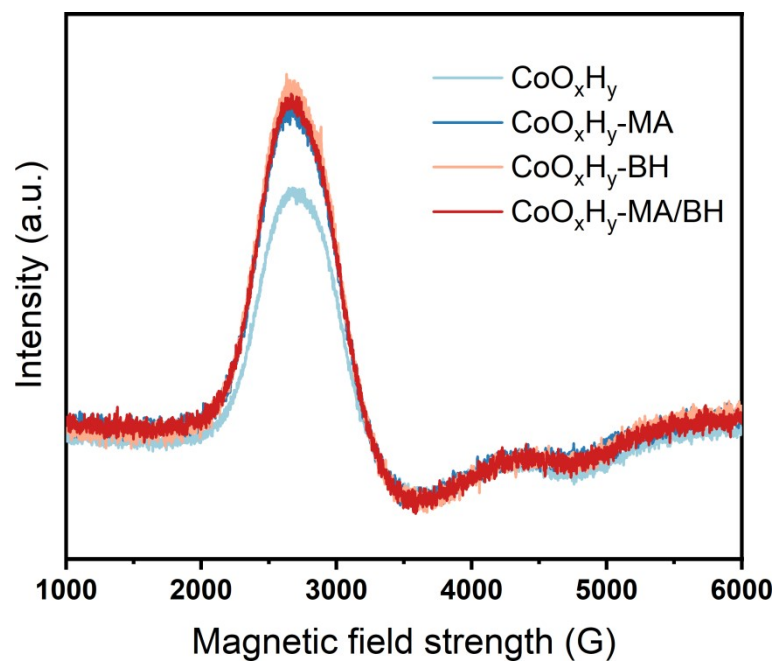


**Figure S5.** FTIR spectra of CoO<sub>x</sub>H<sub>y</sub>, CoO<sub>x</sub>H<sub>y</sub>-MA, CoO<sub>x</sub>H<sub>y</sub>-BH, and CoO<sub>x</sub>H<sub>y</sub>-MA/BH.

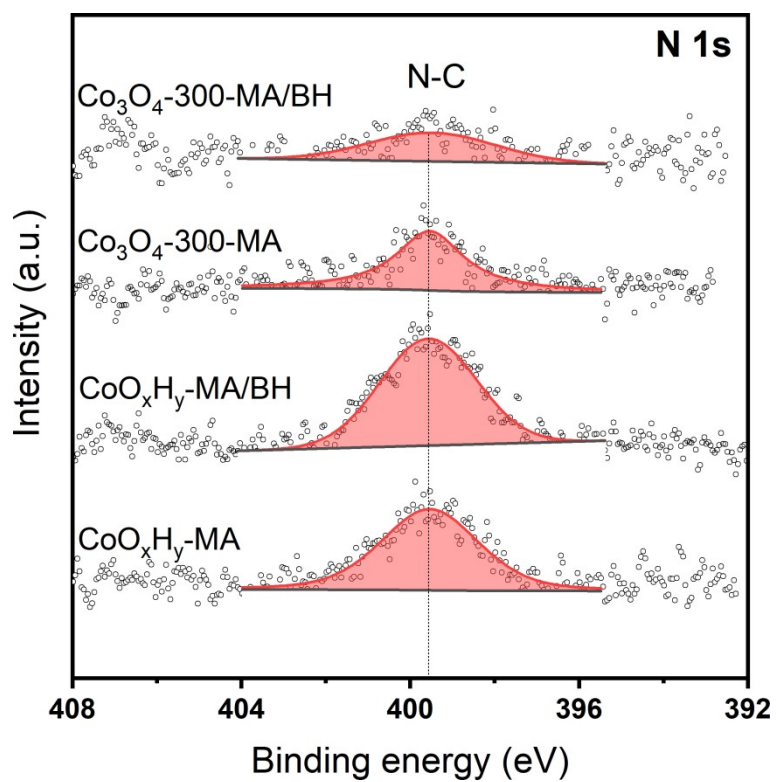


**Figure S6.** XPS (a) Co 2p and (b) O 1s spectra of  $\text{Co}_3\text{O}_4\text{-300}$ ,  $\text{Co}_3\text{O}_4\text{-300-MA}$ ,  $\text{Co}_3\text{O}_4\text{-300-BH}$ , and  $\text{Co}_3\text{O}_4\text{-300-MA/BH}$ .

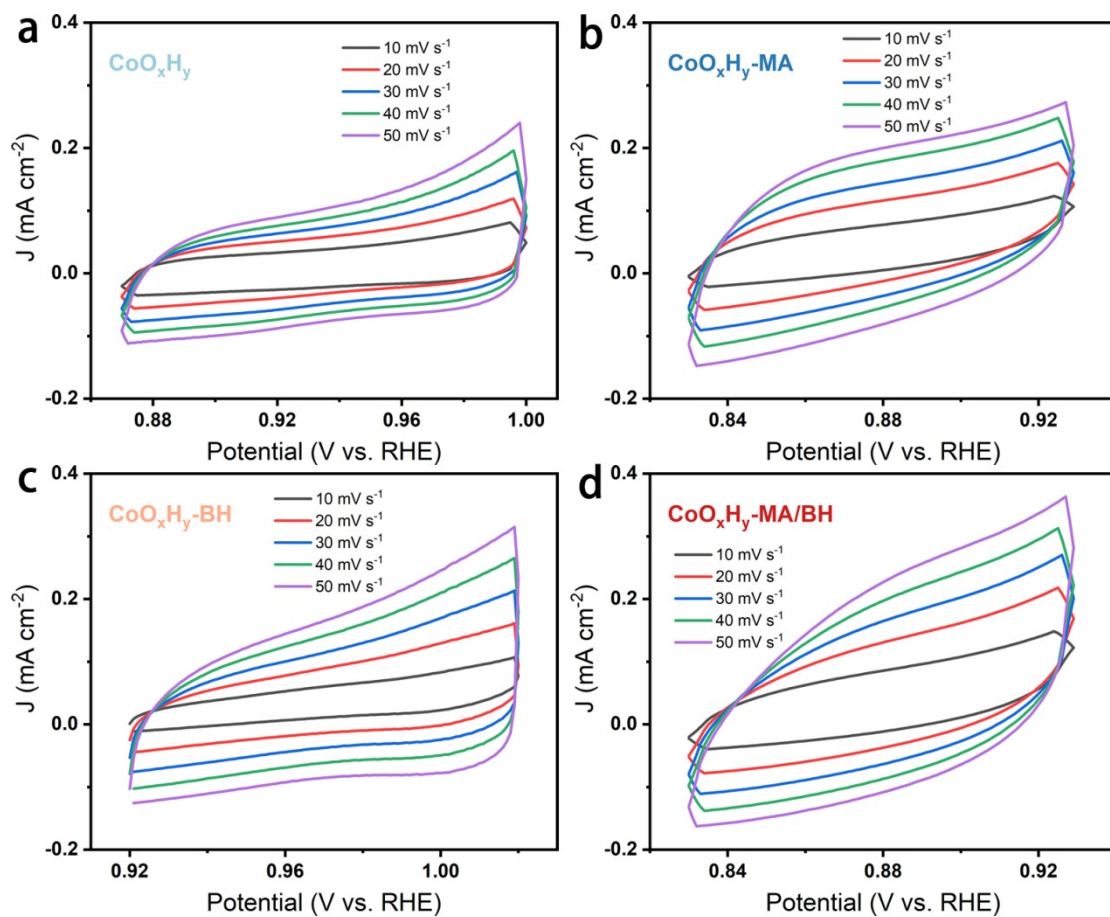




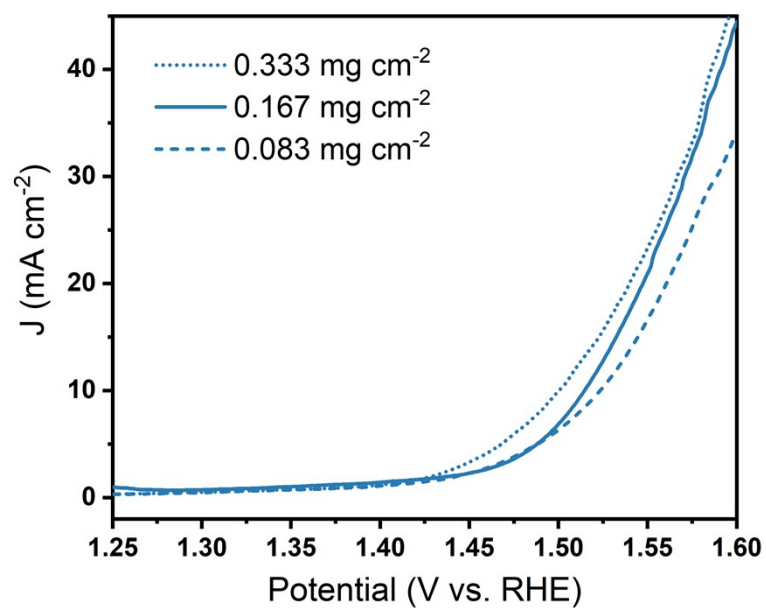
**Figure S7.** EPR signals against magnetic field strength of  $\text{CoO}_x\text{H}_y$ ,  $\text{CoO}_x\text{H}_y\text{-MA}$ ,  $\text{CoO}_x\text{H}_y\text{-BH}$ , and  $\text{CoO}_x\text{H}_y\text{-MA/BH}$ .



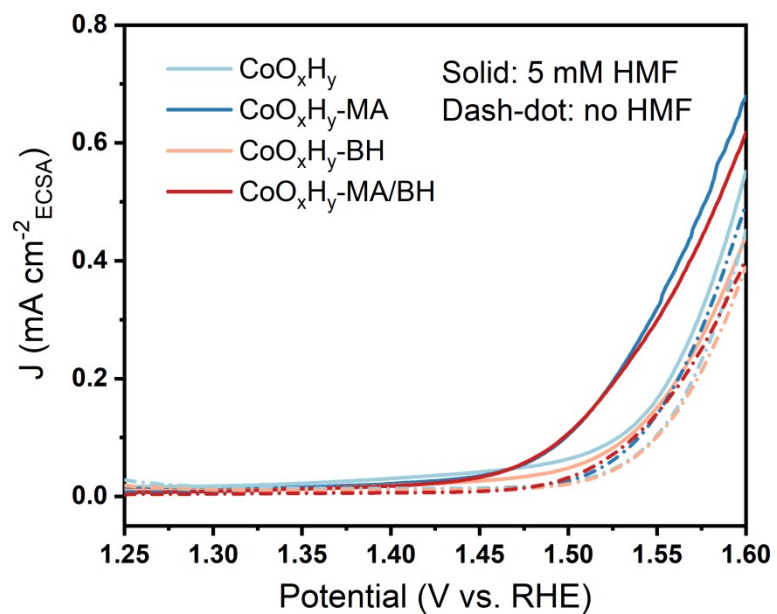
**Figure S8.** XPS N 1s spectra of  $\text{CoO}_x\text{H}_y\text{-MA}$ ,  $\text{CoO}_x\text{H}_y\text{-MA/BH}$ ,  $\text{Co}_3\text{O}_4\text{-300-MA}$ , and  $\text{Co}_3\text{O}_4\text{-300-MA/BH}$ .



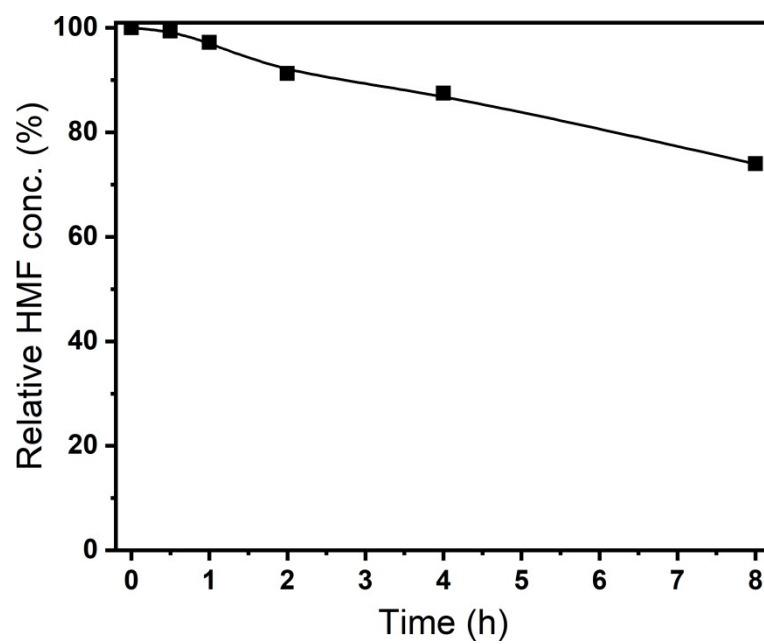
**Figure S9.** CVs at different scanning rates from 10 to 50 mV s<sup>-1</sup> of (a) CoO<sub>x</sub>H<sub>y</sub>, (b) CoO<sub>x</sub>H<sub>y</sub>-MA, (c) CoO<sub>x</sub>H<sub>y</sub>-BH, and (d) CoO<sub>x</sub>H<sub>y</sub>-MA/BH.



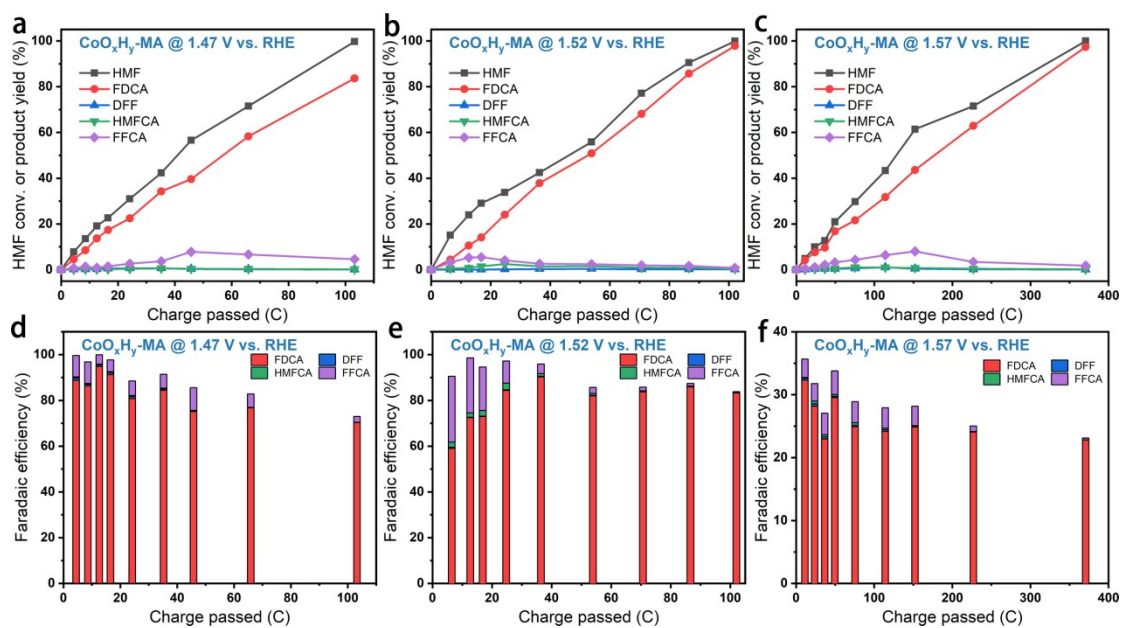
**Figure S10.** The HMFOR LSVs of  $\text{CoO}_x\text{H}_y\text{-MA}$  with different loadings on the glassy carbon electrode in 1 M KOH solution containing 5 mM HMF at a scanning rate of  $5 \text{ mV s}^{-1}$ .



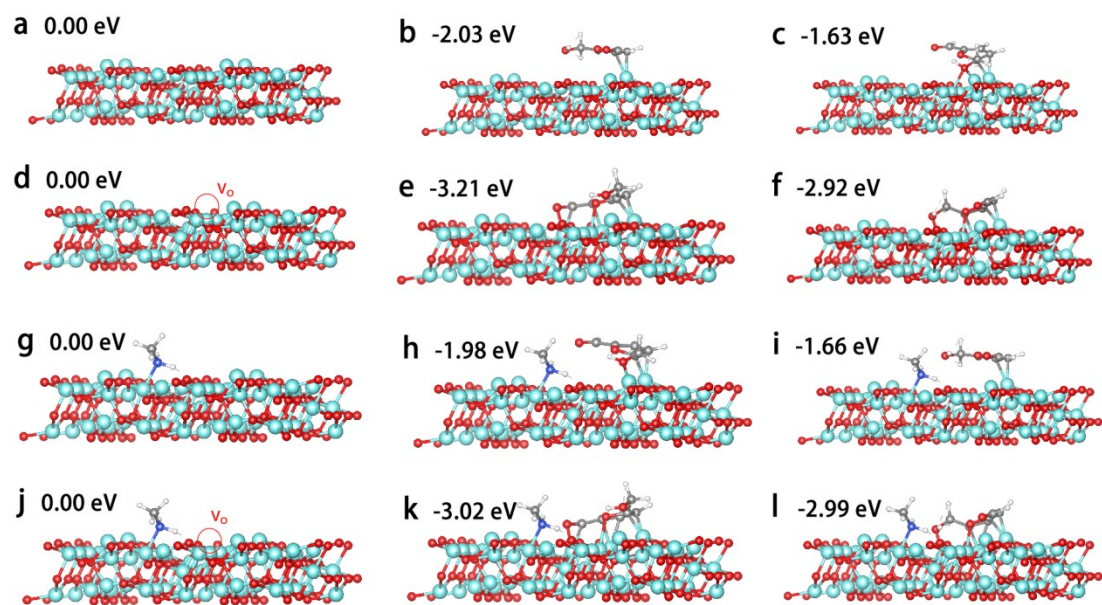
**Figure S11.** The HMF and OER performance of CoO<sub>x</sub>H<sub>y</sub>, CoO<sub>x</sub>H<sub>y</sub>-MA, CoO<sub>x</sub>H<sub>y</sub>-BH, and CoO<sub>x</sub>H<sub>y</sub>-MA/BH after normalization to their electrochemical surface area.



**Figure S12.** HMF thermodegradation of 5 mM HMF in 1 M KOH without electrocatalysts.



**Figure S13.** (a, b, c) The conversion of HMF and the yields of oxidation products, and (d, e, f) the corresponding Faradaic efficiency of CoO<sub>x</sub>H<sub>y</sub>-MA in 1.0 M KOH containing 5 mM HMF at different potentials of (a, d) 1.47, (b, e) 1.52, and (c, f) 1.57 V, respectively.



**Figure S14.** DFT-calculated adsorption configurations for  $\text{Co}_3\text{O}_4$  catalysts: (a, b, c) (3 1 1) surface, (d, e, f) in the presence of surface oxygen vacancy ( $V_{\text{O}}$ ) site, (g, h, i) in the presence of chemisorbed methylamine (MA) molecule, and (j, k, l) in the presence of  $V_{\text{O}}$  and MA. (b, d, g, j) represent the adsorption behavior of HMF starting from its -CHO group, and (c, f, i, l) represent the adsorption behavior of HMF starting from its -C-OH group.