Supplementary Information

Promoting Co(II)-EDTA decomplexation by central atom oxidation in contact-electro-catalysis

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Experimental Procedures:

Preparation of the Co(II)-EDTA solution: 373 mg EDTA-2Na and 293 mg $Co(NO_3)_2 \cdot 6H_2O$ were added into 1000 ml deionized water and stirred for 24 h to obtain a pale pink transparent 1 mmol/L Co(II)-EDTA solution.

CEC experiment process: FEP was added into 30 ml of Co(II)-EDTA solution, which was fully mixed by magnetic stirring, and 2 ml of solution was taken for subsequent test after a certain period of reaction. Then 1 mL of the reaction solution was added with 100 μL 0.5 mol/L NaOH solution, which was fully mixed and kept for 5 h for complete precipitation. The precipitation and supernatant were retained for testing.

1H NMR test sample: Take a certain amount of Co(II)-EDTA solution (i), after CEC treatment (ii) and the supernatant after precipitation (iii) into a lyophilizer to freeze drying for 15 h. Then fully mixed with 540 μL D_2O and 60 μL of 0.8 mmol/L DMSO in D_2O, and transferred into 5 mm NMR tube for analysis by Bruker AVANCE NEO 400 MHz NMR spectrometer.

LC-MS test sample: The LC-MS was obtained by Agilent 1290 UPLC system. Each injection volume was 5 μL, and the analysis and separation of the solution were carried out using the waters BEH C18 column (2.1 × 100 mm, 1.7 μm). The mobile phases were composed of solvent A: 0.1% formic acid aqueous solution and solvent B:

acetonitrile solution with flow rate of 0.3 mL/min. The mass spectrometry analysis was performed using the Agilent qtof 6550, with a scan range of 50-1000 m/z. The temperature of the sheath gas was set at 350℃, with a flow rate of 12 L/min.

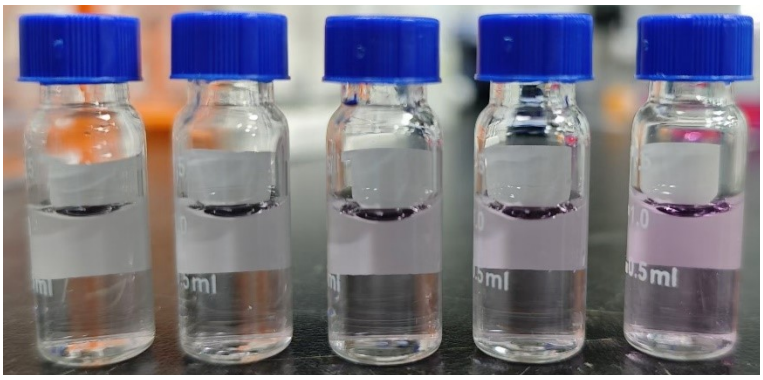

Fig. S1 Color variation of ultrasonication with FEP in 30, 60, 90, 120, 240 minutes.

Fig. S2 Color variation of ultrasonication without FEP in 30, 60, 90, 120, 240 minutes.

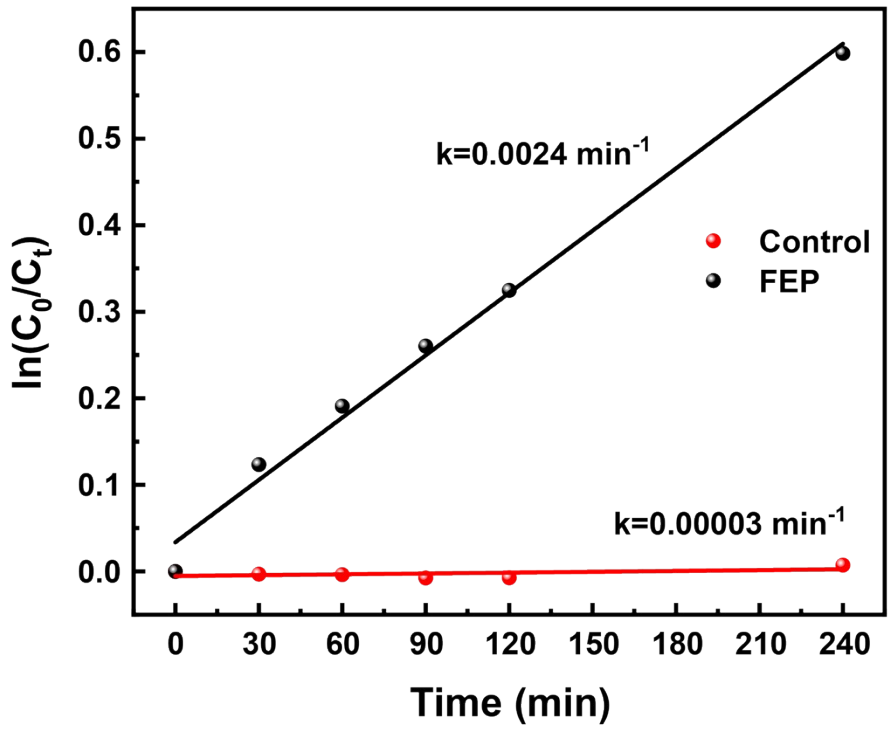

Fig. S3 The kinetics of Co(II)-EDTA decomplexation with or without FEP.

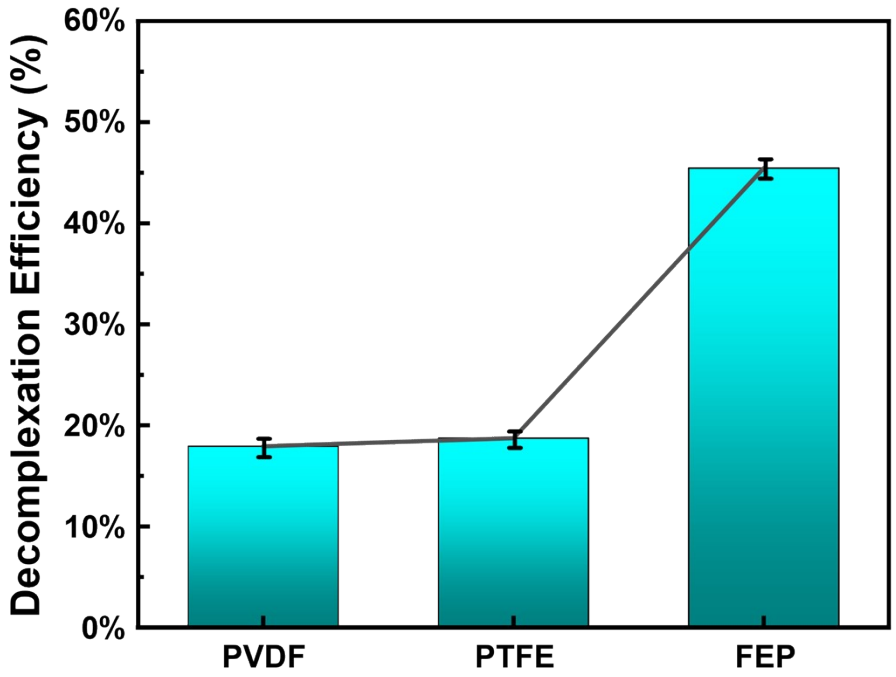

Fig. S4 Comparison of Co(II)-EDTA decomplexation efficiency with different particles for 240 min.

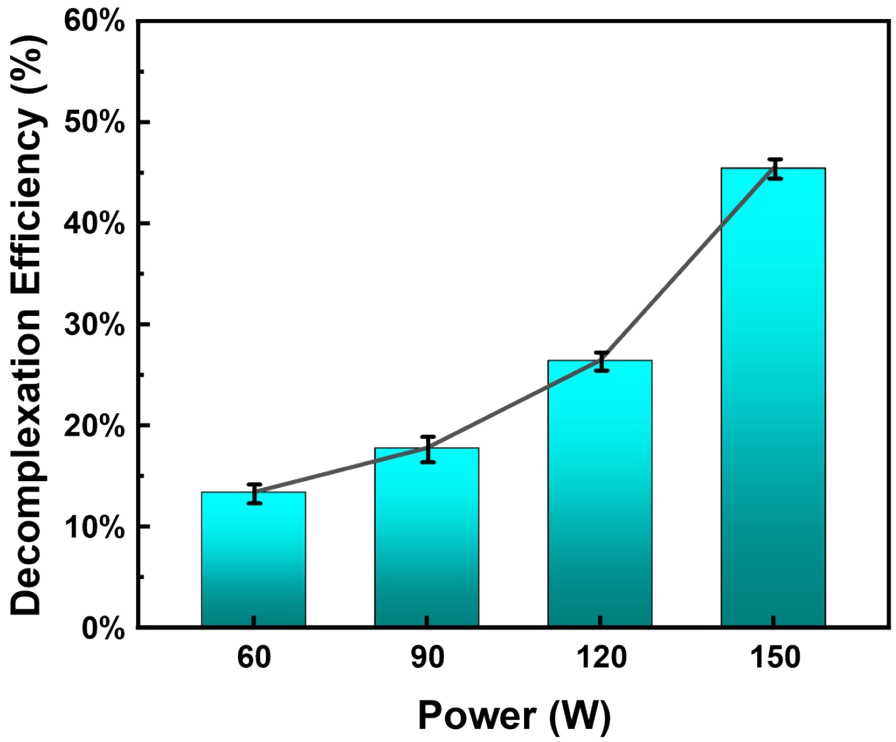

Fig. S5 Comparison of Co(II)-EDTA decomplexation efficiency under different ultrasonic powers for 240 min.

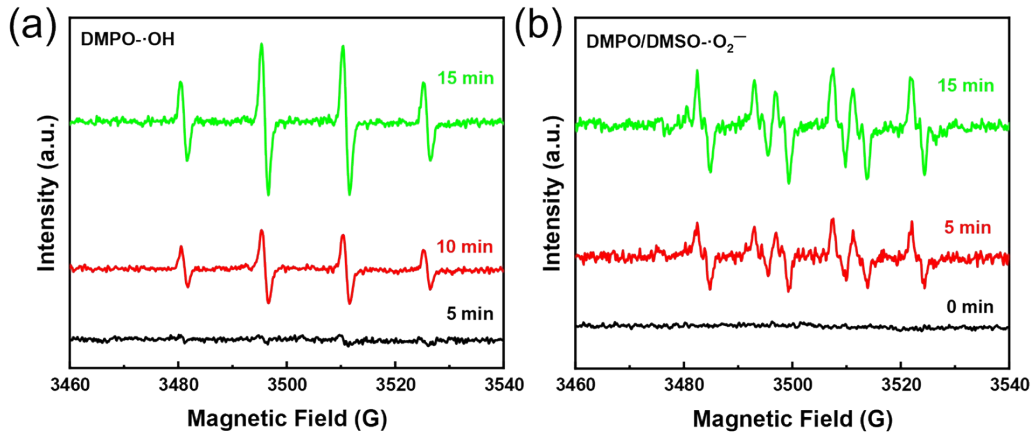

Fig. S6 EPR spectra for (a) DMPO-·OH, (b) DMPO/DMSO-·O_2^- in the presence of FEP particles during different ultrasonication times.

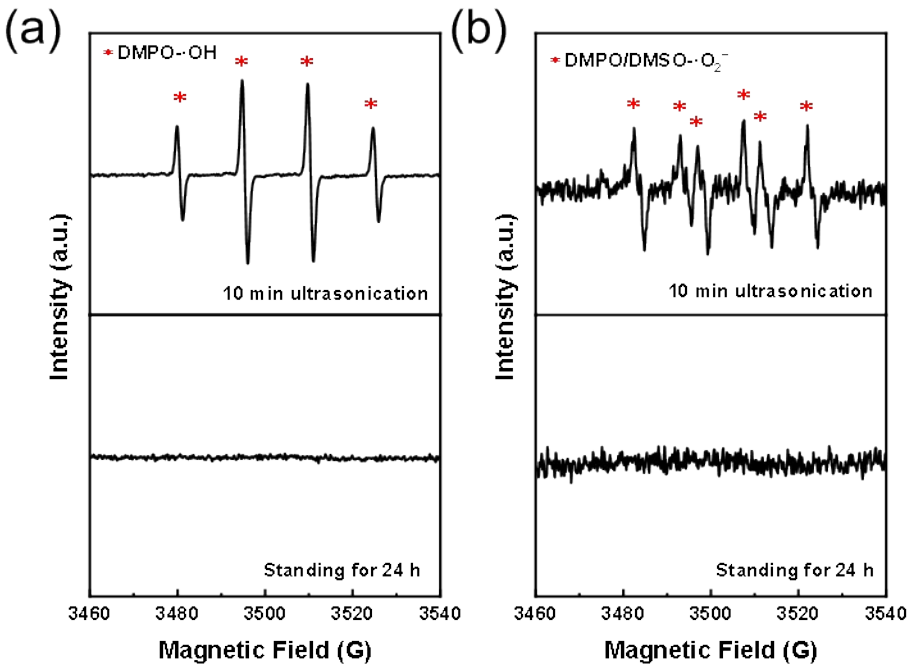

Fig. S7 EPR spectra for (a) DMPO-·OH, (b) DMPO/DMSO-·O_2^- in the presence of FEP particles after 10 min ultrasonication (top) and standing for 24 hours after ultrasonic treatment (bottom).

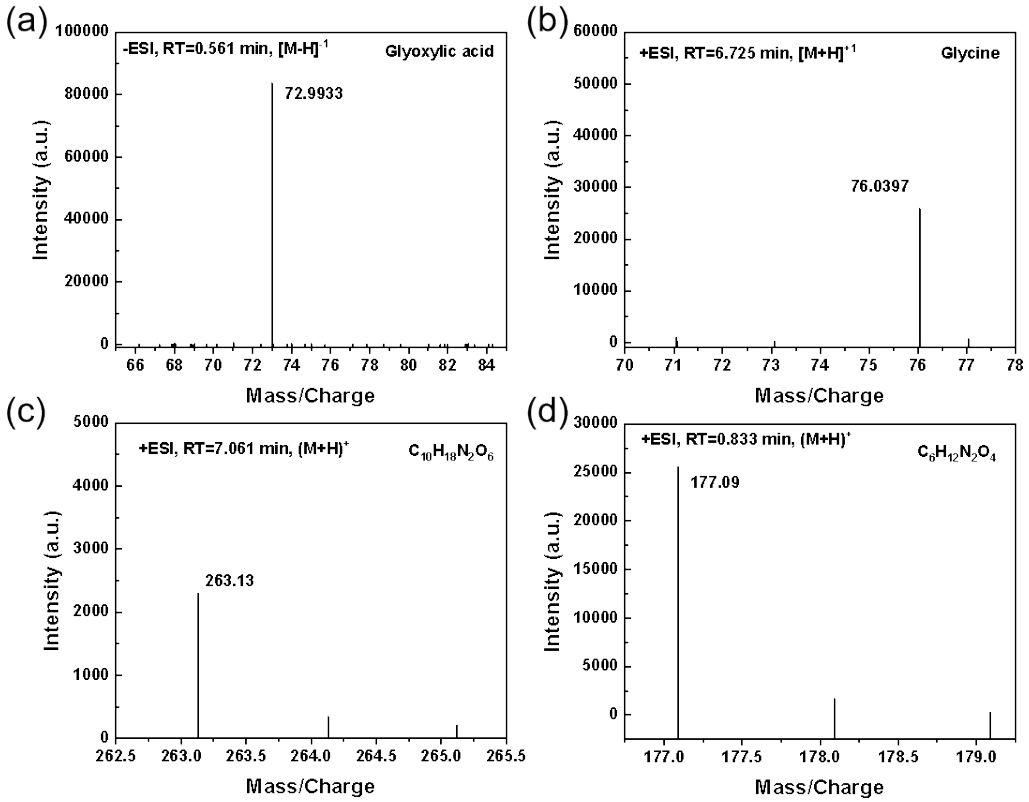

Fig. S8 Mass spectra of degradation products. (a) Glyoxylic acid, $C_2H_2O_3$; (b) Glycine, $C_2H_5NO_2$; (c) ED3A, $C_{10}H_{18}N_2O_6$; (d) ED2A, $C_8H_{12}N_2O_4$.

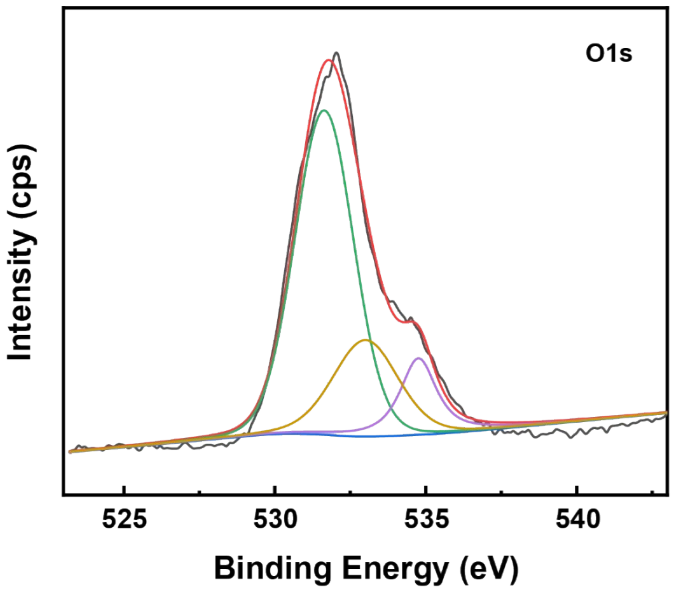

Fig. S9 O 1s spectra of the precipitates.

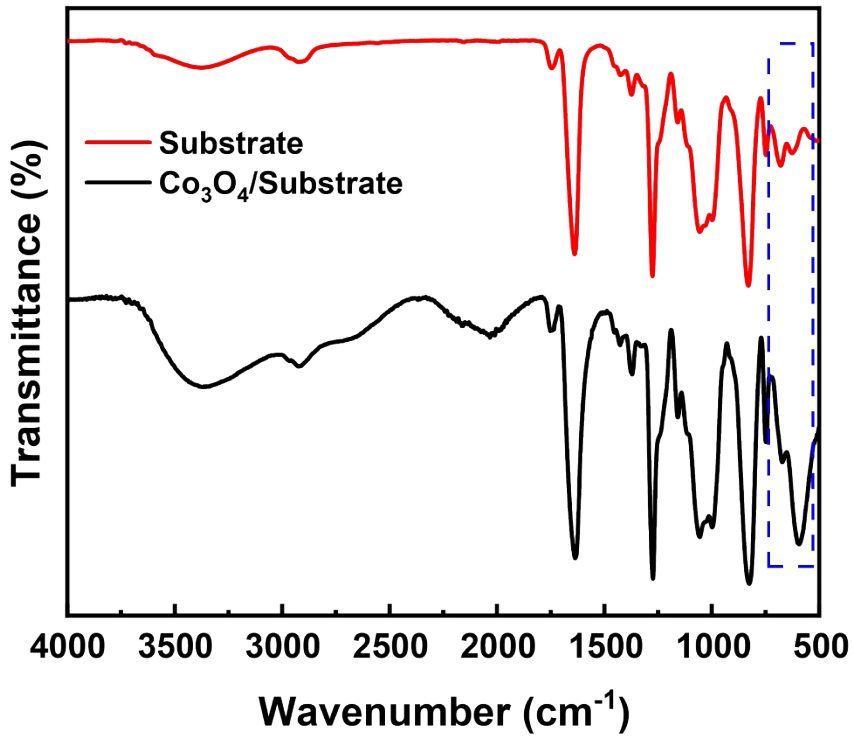

Fig. S10 FTIR spectra of the substrate (red) and the precipitates attached to the substrate (black).

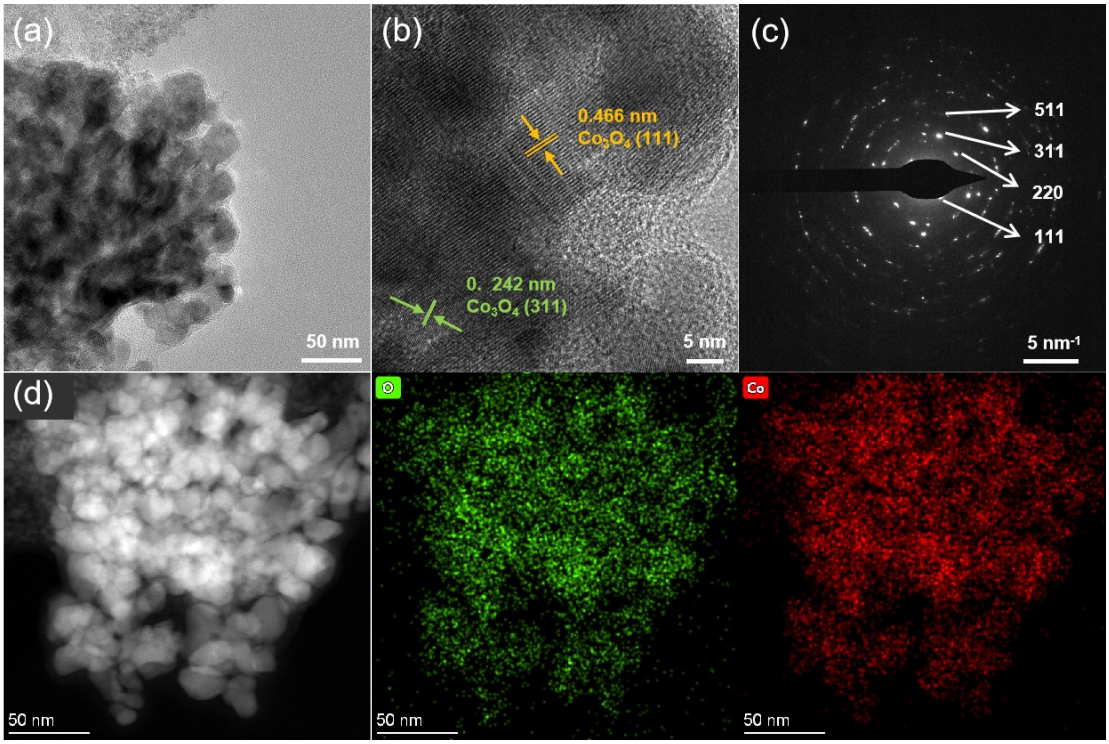

Fig. S11 Characterization of Co_3O_4 nanoparticles. (a) TEM image, (b) HRTEM image, (c) SAED pattern, and (d) Dark-field elemental mappings of the precipitates (Co_3O_4).

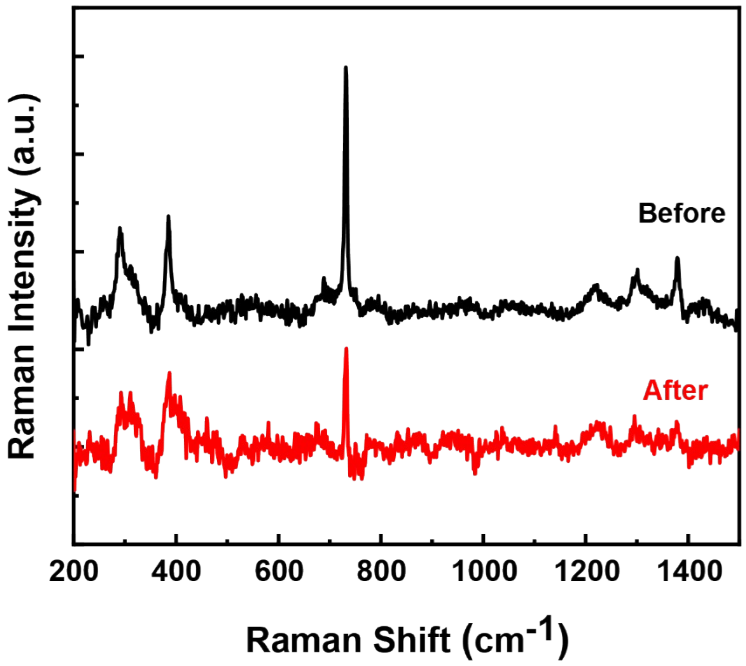

Fig. S12 Raman spectra of FEP before (black) and after (red) the reaction.