

Supplementary Information

Double pyramid stacked CoO nano-crystals induced by graphene at low temperature as highly efficient Fenton-like catalysts

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Catalytic Activity Evaluation.

The degradation effects of CoO@graphene for dye pollutants were investigated by using 10 ppm RhB as a model dye. At room temperature, 10 mg CoO@graphene was added to 20 mL 10 mg L⁻¹ RhB aqueous solution placed in the 50 mL reactor and then stirred to start the adsorption process. Then 1 mL solution was taken at 0, 15, 30, 45, 60, 90 and 120 minutes, respectively, and centrifuged in a centrifuge at 6000 rpm for 5 min to wipe off the CoO@graphene. Then, the adsorption reaction ended. After reaching adsorption equilibrium, 100 µL PMS (18 g/L) was added to launch the Fenton-like reaction. The 1 mL solution was removed at the same time interval as the adsorption process, and the CoO@graphene was also removed. The absorbance of the extracted solution was measured by UV-visible absorption spectrometry (UV-1600, Mapada, China). The effects of different pH values (pH=3, 5, 7, 9 and 11) on the adsorption and Fenton catalysis of CoO@graphene were investigated by adjusting the pH value of the solution with 0.1M H₂SO₄ or 0.1M NaOH solution. Similarly, all other conditions being equal, we explored the effect of different amounts of PMS (30 mg/L, 60 mg/L, 90 mg/L, 120 mg/L) on the adsorption and catalytic properties. The catalytic efficiency of the catalyst treated with adsorption for 2 h was compared with that of the catalyst not treated with adsorption.

PMSO and PMSO₂ were detected By High-performance liquid chromatography (HPLC, Thermo U3000). A Phenomenex Kinetex F5 column (150 mm × 4.6 mm, 2.6 µm particle size) was used for the separation of products. The mobile phase consisted of 30 v% ultrapure water (A) and 70 v% acetonitrile (B), and the flow rate was 1.0 mL·min⁻¹. The column temperature was kept constant at 25 °C. Sample injection volume was 20 µL.

Stability Experiment

200 mg of CoO@graphene were added to 500 mL of an aqueous RhB solution at 10mg L⁻¹, placed in 1000 mL beakers and mixed to start the adsorption process at room temperature. After reaching adsorption equilibrium, the first three hours reflect the adsorption effect, while the last two hours initiate the catalytic degradation process after

the addition of 5 mL of PMS (18 g/L). Similarly, the 1 mL solution was centrifuged and the remaining RhB was measured using UV-vis absorption spectroscopy (UV-1600, Mapada, China). Subsequently, the CoO@graphene after the adsorption catalytic reaction was collected, washed, and dried. This process was repeated with the dried sample until the catalyst was reused for the 5 time.

PS2: Calculations

The removal efficiency of RhB was determined using equation (1) by normalising the measured absorbance using the peak at 554 nm as the numerator and the absorbance at 0 min as the denominator:

$$\eta = C/C_0 \quad (1)$$

Where, the concentration of pollutants at a specific reaction time (t) is represented by C, while C_0 signifies the initial pollutant concentration. Calculation of k value.

The RhB degradation kinetics was fitted by the pseudo first order model and the apparent rate constant (k) was calculated according to eqs (2):

$$\ln(C/C_0) = -kt \quad (2)$$

where C is the pollutant concentration at a certain reaction time (t) and C_0 is the initial pollutant concentration.

PS3: Supplementary figures

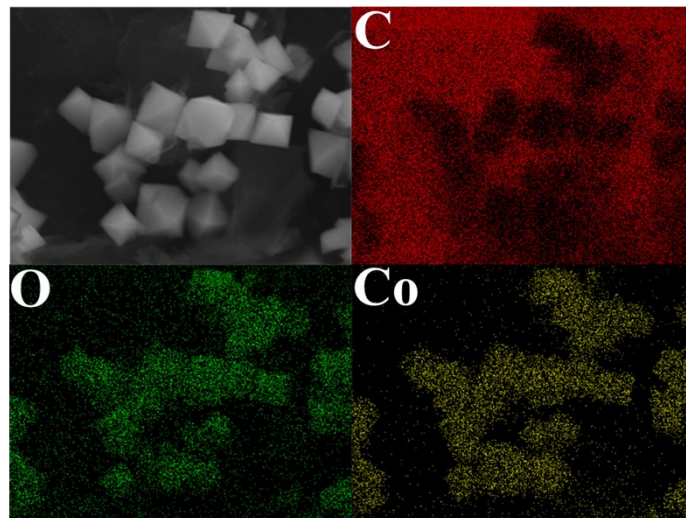
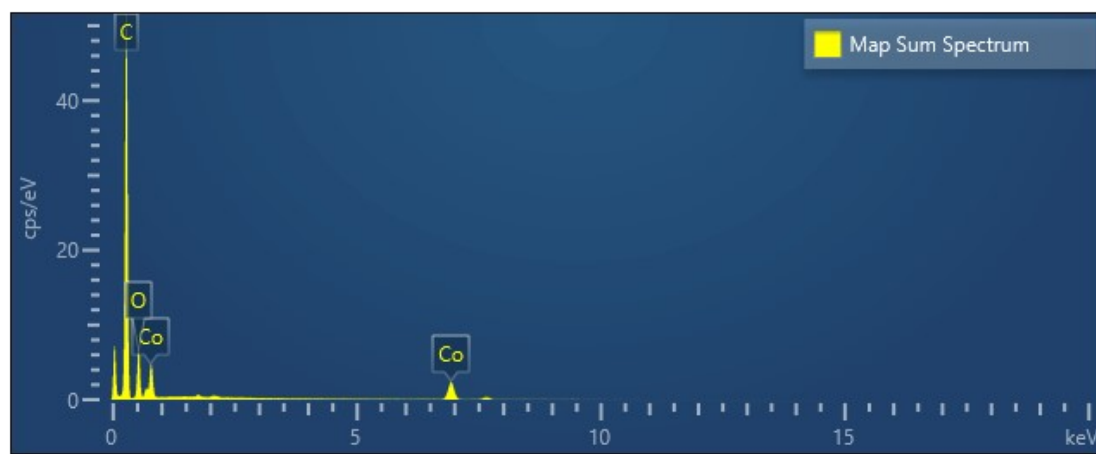


Fig. S1. SEM image and EDS analysis of the CoO@graphene.



Element	Wt%	At%
C	68.50	83.05
O	13.83	12.59
Co	17.67	4.37
Total:	100.00	100.00

Table. S1 The mass and atom ratio of CoO@graphene in the catalytic converter.

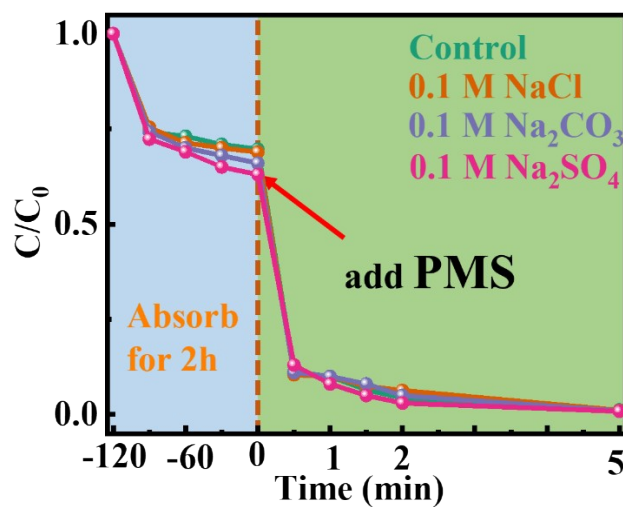


Fig. S2 Degradation efficiency of CoO@G/PMS catalytic system in the presence of 0.1 M NaCl, Na₂SO₄ and Na₂CO₃. Reaction conditions: [catalyst] = 0.5 g/L, [PMS] = 90 mg/L, [RhB] = 10 mg/L and pH 7.0.

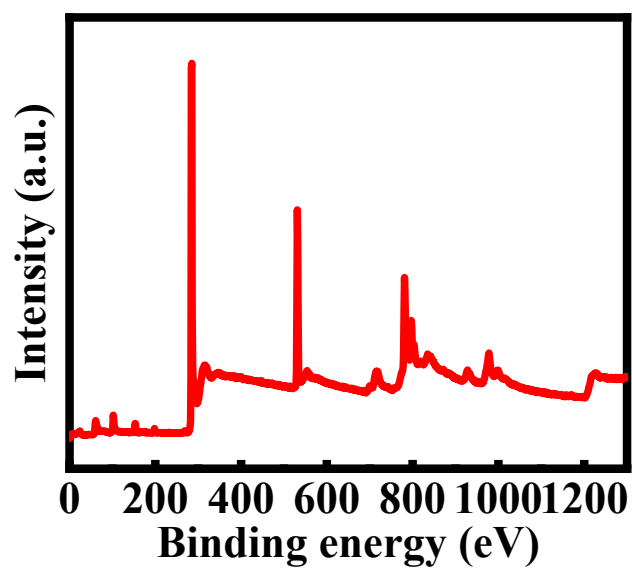


Fig. S3. The full-scale XPS survey of CoO@graphene.

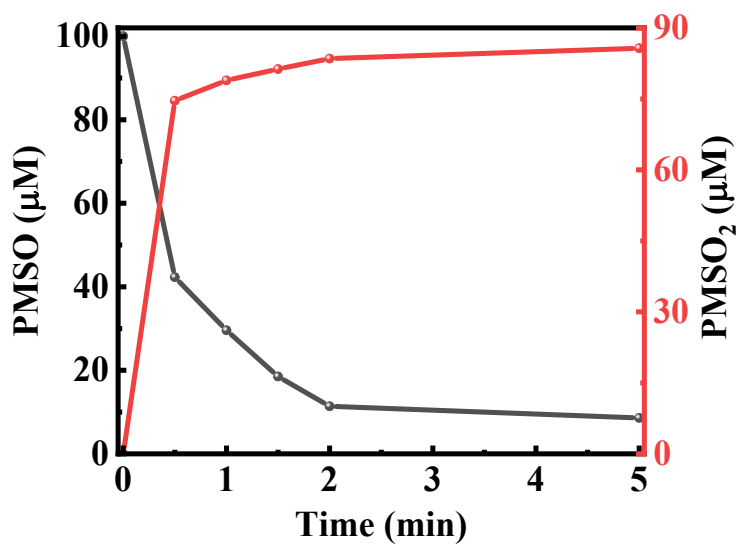


Fig. S4 Production of PMSO₂ and consumption of PMSO in the CoO@graphene/PMS system.