

Supporting Information

# Laser-Induced Nitrogen-Doped Graphene Composite Iron–Cobalt Hydroxide for Methylene Blue Degradation via Electrocatalytic Activation of Peroxymonosulfate

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## Text S1 Chemicals and materials

Methylene blue (MB), Cobalt nitrate ( $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ), Ferric nitrate nonahydrate ( $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ), Sodium chloride (NaCl), Sodium bicarbonate ( $\text{NaHCO}_3$ ), Sodium sulfate anhydrous ( $\text{Na}_2\text{SO}_4$ ), Sodium hydroxide (NaOH), Sodium thiosulfate ( $\text{Na}_2\text{S}_2\text{O}_3$ ), Monosodium phosphate ( $\text{NaH}_2\text{PO}_4$ ), Ethanol, Rhodamine b (RhB), Methyl orange (MO), 2,2,6,6-tetramethyl-4-piperidinol (TEMP), 5,5-dimethyl-1-pyrroline N-oxide (DMPO), methanol (MeOH), tert-butyl alcohol (TBA), and p-benzoquinone benzoquinone (*p*-BQ) were supplied from Macklin Chemical Reagent Co., China. Peroxymonosulfate (PMS,  $\text{KHSO}_5 \cdot 0.5\text{KHSO}_4 \cdot 0.5\text{K}_2\text{SO}_4$ ) was supplied from Sigma Chemical Co., Ltd (St Louis, MO, USA).

## Text S2 Characterization

The external morphology and internal structure of the samples were characterized by scanning electron microscopy (SEM). The phase and crystal structure of the catalyst were analyzed by X-ray diffractometer (XRD, Cu K $\alpha$ ). X-ray photoelectron spectroscopy (XPS) was used to confirm the elemental composition of the sample surface. Linear scanning voltammetric curves (LSV) and open circuit voltage curves (OCPT) were measured on an electrochemical workstation. The active species during the reaction were identified by electron paramagnetic resonance (EPR). The intermediate products of the degradation process were measured and analyzed by ion trap-time of flight mass spectrometry (LC-MS, SHIMADZU). The concentration of metal ions was measured by inductively coupled plasma tandem mass spectrometer (ICP-MS, Agilent 720ES).

**Table S1** Recently reported catalytic performance of comparable electrocatalysts

Electrode	Catalysts	Reaction conditions	Degradation efficiency	Reaction rate constant $k_{\text{obs}}$	Ref.
Graphite plates	[Fe-MOF] = 0.4 g/L	[Cu-EDTA] = 1 mM; [PS] = 4 mM; [Current density] = 2.86 mA/cm <sup>2</sup>	100min 100%	0.0415min <sup>-1</sup>	[1]
Ti plates	[ACS-850] = 0.1 g/L	[phenol] = 50 mg/L; [PS] = 15 mM; [Current density] = 2 mA/cm <sup>2</sup>	60min 100%	0.081min <sup>-1</sup>	[2]
BDD	/	[SMZ] = 50 mg/L; [PS] = 0.4 g/L; [pH] = 4; [Current density] = 21 mA/cm <sup>2</sup>	15min 100%	/	[3]
LIG	/	[MB] = 5mg/L; [Applied voltage] = 2.5V	25h 96%	/	[4]
LIG	/	[Cr (VI)] = 2mg/L; [Applied voltage] = 3V	6h 90%	/	[5]
FeCo-LDH/LI-NDG	/	[MB] = 20mg/L; [PMS] = 0.5 mM; [Applied voltage] = 2V	6min 100%	0.461min <sup>-1</sup>	This work

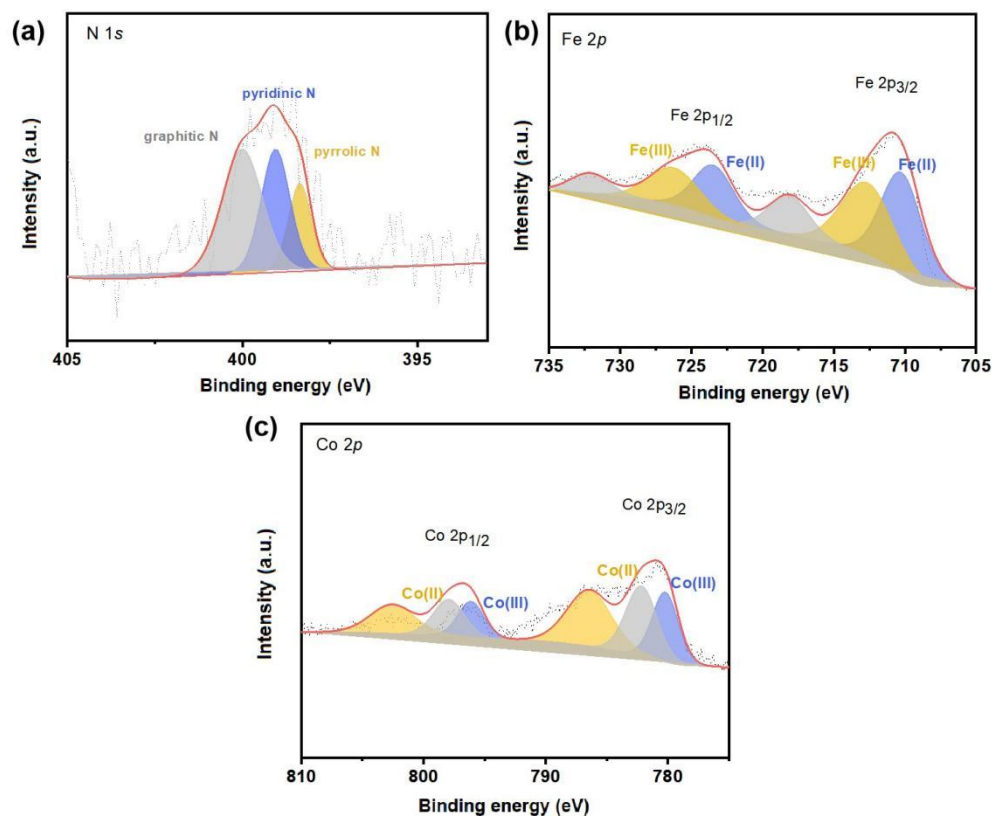


Figure S1. (a) N 1s, (b) Fe 2p, and (c) Co2p XPS spectrum of FeCo-LDH/LI-NDG.

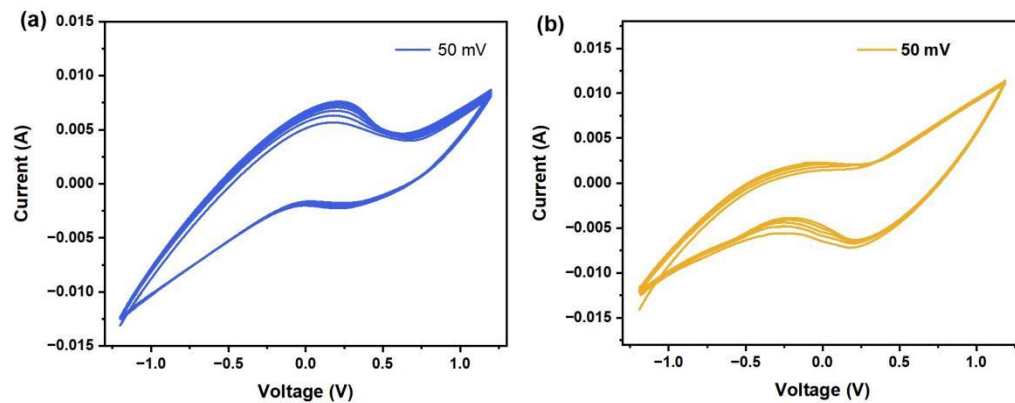


Figure S2. (a) CV curves for 10 consecutive scans of FeCo-LDH/LI-NDG; (a) CV curves for 10 consecutive scans of FeCo-LDH/LI-NDG+PMS.

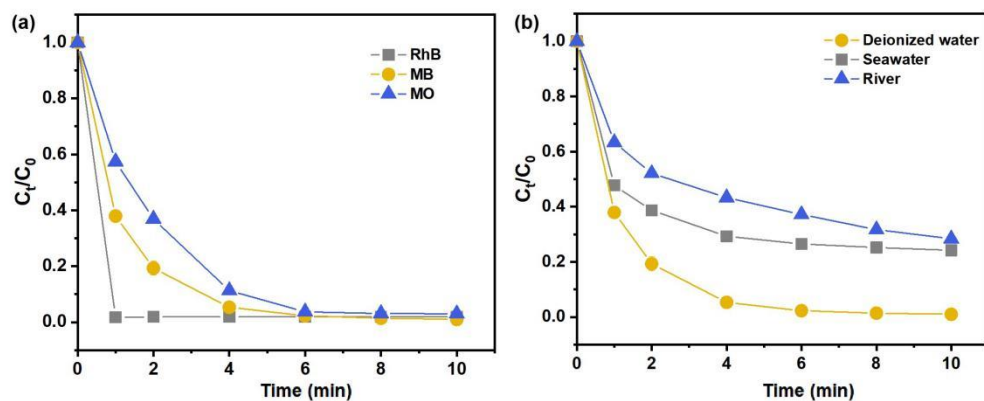


Figure S3. The degradation efficiency of FeCo-LDH/LI-NDG/EC/PMS system with (a) various contaminants and (b) various water bodies. Reaction conditions:  $[MB] = [RhB] = [MO] = 20 \text{ mg/L}$ ;  $[PMS] = 0.5 \text{ mM}$ ;  $[Applied \text{ voltage}] = 2 \text{ V}$ ;  $[Initial \text{ pH}] = 6.8$ .

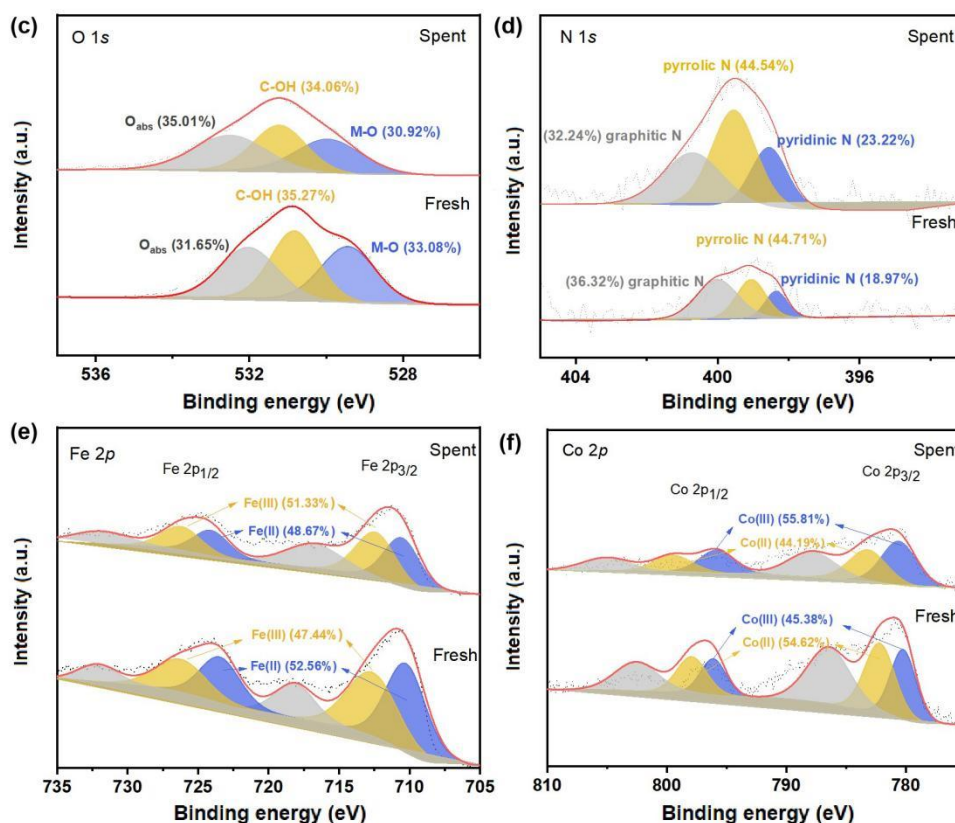


Figure S4. (a) O1s, (b) N1s, (c) Fe 2p, and (d) Co 2p XPS spectrum of spent and fresh FeCo-LDH/LI-NDG.

## References

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