Supplementary Materials

Text S1. Effect of composites with different ZIF-67 loads on TC degradation by catalytic H_2O_2.

In order to explore effect of composites with different ZIF-67 loads on TC degradation by catalytic H_2O_2, in addition to the composite ratio of 0.164 g 2-MeIm, other synthetic materials with different loads were prepared, as follows:

$$0.164\ g\ 2\text{-MeIm} + 0.966\ g\ Co(NO_3)_2 \cdot 6H_2O \quad (S1)$$

$$0.328\ g\ 2\text{-MeIm} + 0.096\ g\ Co(NO_3)_2 \cdot 6H_2O \quad (S2)$$

$$0.082\ g\ 2\text{-MeIm} + 0.0483\ g\ Co(NO_3)_2 \cdot 6H_2O \quad (S3)$$

$$0.0328\ g\ 2\text{-MeIm} + 0.01932\ g\ Co(NO_3)_2 \cdot 6H_2O \quad (S4)$$

The results as shown as Fig.S1. The 1.64 g 2-MeIm group achieved 82.2% of TC degradation, while the 0.164 g 2-MeIm group achieved 82.6% of TC degradation. We found 1.64 g 2-MeIm and 0.082 g 2-MeIm had the same degradation rate as the 0.164 g 2-MeIm group, but the degradation rate decreased at 40 min and 50 min. As the ZIF-67 content decreasing, the degradation efficiency was obviously slowed and decreased, which proved that ZIF-67 of the different content played an important role in the catalytic reaction. Therefore, formal experiments used composite of 0.164 g 2-MeIm complexes.

Table S1. The rest of the chemicals used for the experiments.

reagent	purity	Manufacturer and origin
NaOH	AR	Tianjin Wind Ship Chemical Reagent Technology Co., LTD
H_2SO_4	98%	Luoyang chemical reagent factory
$NaHCO_3$	AR	Shanghai Aladdin Co., Ltd
KH_2PO_4	99.5%	Tianjin Kemio Chemical Technology Co., LTD
KNO_3	98.5%	Shanghai MacLean Biochemical Co., Ltd
Na_2SO_4	99%	Shanghai MacLean Biochemical Co., Ltd
Methanol (MeOH), CH_3OH	98%	Tianjin Kemio Chemical Technology Co., LTD
Tert-butyl alcohol (TBA), $C_4H_{10}O$	99%	Tianjin Kemio Chemical Technology Co., LTD
Para-benzoquinone (1.4-PBQ), $C_6H_4O_2$	99%	Shanghai Aladdin Co., Ltd
Furyl alcohol (FFA), $C_5H_6O_2$	98%	Shanghai Aladdin Co., Ltd

Table S2. The content of C, N, O, Fe and Co in CG materials before and after loading ZIF-67 changes.

element		Before loading (%)	After loading (%)
C	C=C	52.7	39.2
	C=N	20.0	27.8
	O=C-O	27.3	33.0
	Total	40.4	39.66
N	Pyridinic N	20.9	15.2
	A-H	34.7	59.4
	graphite	44.4	7.6
	Pyridine-like oxides	--	17.7
	Total	0.89	2.0
O	A-O	100	36.62
	Co-OH	--	63.4
	Total	58.87	53.25
Fe	Fe (II)	65.8	--
	Fe (III)	34.2	--
	Fe chemical compound	--	100
	Total	0.22	0.39
Co	Co (II)	--	54.56
	Co (III)	--	45.44
	Total	--	7.0

Table S3. Changes of C, N, O, Fe and Co elements in ZIF-67/CG before and after catalytic reaction.

element		Before reacting (%)	After reacting (%)
C	C=C	39.2	51.0
	C=N	27.8	24.7
	O=C-O	33.0	24.3
	Total	39.66	42.45
N	Pyridinic N	15.2	13.7
	B-H	59.4	66.6
	graphite	7.6	16.53
	Pyridine-like oxides	17.7	3.14
	Total	2.0	1.2
O	C-O	36.62	42.4
	Co-OH	63.4	57.6
	Total	53.25	47.3
Fe	Fe (II)	--	--
	Fe (III)	--	--
	Fe chemical compound	100	100
	Total	0.39	2.63
Co	Co (II)	54.56	66.6
	Co (III)	45.44	33.4
	Total	7.0	6.41

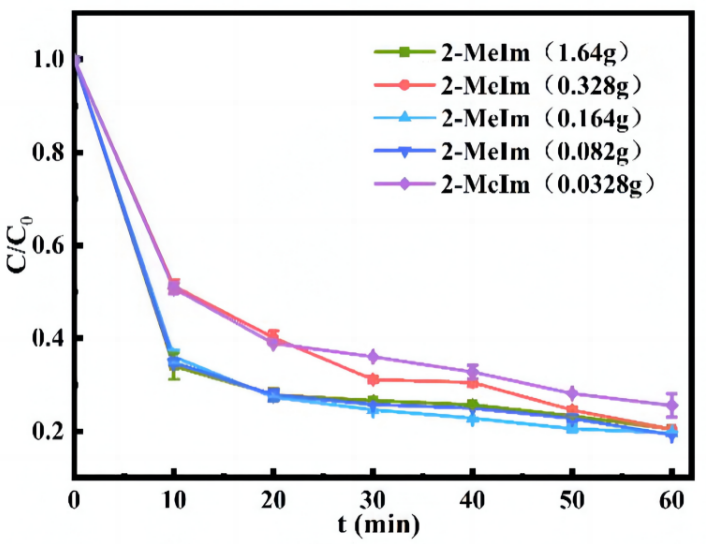

Fig. S1. Effect of CG loads with different ZIF-67 masses on the efficiency of TC degradation. Experimental condition: t = 60 min, C_{TC} = 20 mg/L, $V_{(TC)}$=100 mL, dosa(ZIF-67/CG) = 0.6 g/L, dosa(H_2O_2) = 2 mM, pH = 6.2, T = 25℃.

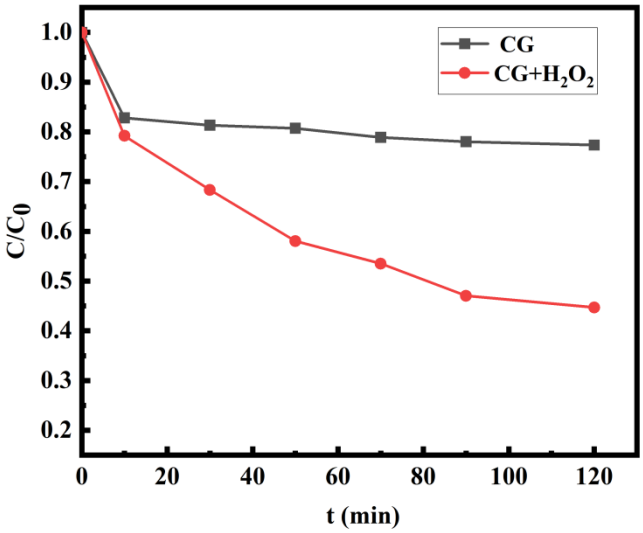

Fig. S2. The TC removal efficiency by sole CG and CG activated H_2O_2 system. Experimental condition: t = 120 min, C_{TC} = 40 mg/L, $V_{(TC)}$ = 50 mL, pH = 6.2, dosa(H_2O_2) = 2 g/L, T = 25℃.

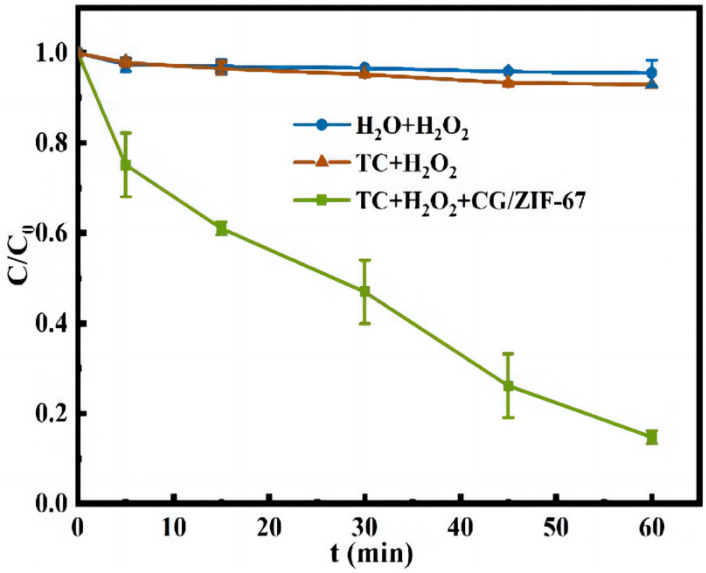

Fig. S3. Consumption of H_2O_2 in the different systems. Experimental condition: t = 60 min, dasa dosa(H_2O_2) = 2 mM, C_{TC} = 20 mg/L, $V_{(TC)}$ = 100 mL, $dosa_{(ZIF-67/CG)}$ = 0.6 g/L, pH = 6.2, T = 25°C.