Supplementary Materials for:

Efficient degradation of organic dyes using peroxymonosulfate activated by magnetic graphene oxide

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Text S1 Materials and characterizations

Coomassie brilliant blue G250 (CBB), methylene blue (MB), Rhodamine B (RhB), basic blue 17 (BB17), methyl orange (MO) and disodium hydrogen phosphate (Na_2HPO_4, 99%) were purchased from J&K Co., Ltd. Potassium peroxomonosulfate (PMS, $2KHSO_5 \cdot KHSO_4 \cdot K_2SO_4$, $KHSO_5 \geq 42.8\%$), tert-butanol (TBA, 99%), furfuryl alcohol (FFA, 98%), benzoquinone (BQ, 99%) and methyl phenyl sulfoxide (PMSO, 98%) were obtained from Aladdin Co., Ltd. Methanol (99.5%) and sodium sulfate (Na_2SO_4, 99%) were purchased from Tianjin Fuchen Chemical Reagent Co., Ltd. Sodium chloride (NaCl, 99.5%) and sodium bicarbonate ($NaHCO_3$, 99.5%) were obtained from Tianjin Guangfu Chemical Reagents and Tianjin Tianhe Chemical Reagents, respectively. Ethanol (99.7%), sulfuric acid (H_2SO_4, AR), sodium hydroxide (NaOH, 96%) and deuterium oxide (D_2O, 99.9%) were obtained from Tianjin Keman Reagents, Tianjin Kemiou Reagents, Tianjin Jinfeng Reagents and Shanghai Yien Reagents, respectively. Nafion® solution (5 wt.%) was purchased from Dupont. Tap water was obtained in Dalian Maritime University and used directly. Lake water and sea water were derived from the Xinhai Lake on campus of Dalian Maritime University and the Heishijiao Bay of Dalian respectively and treated with 0.22 μm membrane filters before use. Deionized water was used except for the experiments with real waters.

The specific surface areas (S_{BET}) were calculated using the Brunauer–Emmett–Teller (BET) method by selecting the suitable relative pressure ranges to obtain correlation coefficients (r) larger than 0.999 and positive C constants. The total pore

volumes (V_t) were derived from the saturated points on the isotherms. The average pore sizes (D_p) were calculated by $D_p=4V_t/S_{BET}$. For the open circuit potential (OCP) test, 50 μL Nafion was added into 650 μL ethanol and then mixed with 10 mg MGO catalyst. After ultrasound treatment for 60min, 50 μL of the mixture was withdrawn, dripped on a glassy carbon electrode (GCE) and dried naturally. The MGO coated GCE (MGO-GCE) was obtained by repeating the dripping step three times and then dipped in 0.1 M Na_2SO_4 overnight to maintaining a stable potential. The open circuit potential (OCP) of MGO-GCE was monitored using Ag/AgCl as the reference electrode and Pt sheet as the counter electrode in 0.1 M Na_2SO_4. PMS (20 mM) and CBB (50 mg/L) was added into the solution at ~100 s and ~1000 s respectively.

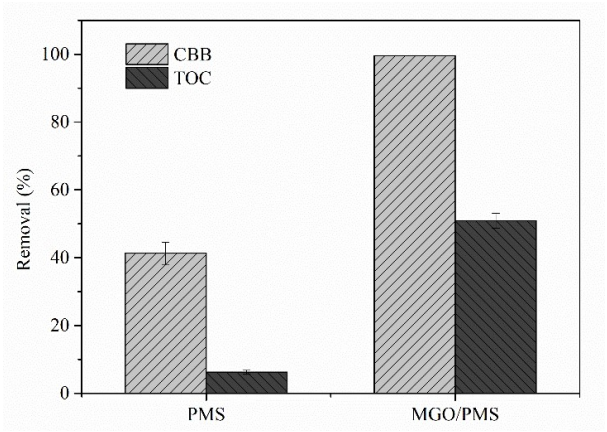

Figure S1 CBB and TOC removals obtained with PMS and the MGO/PMS system

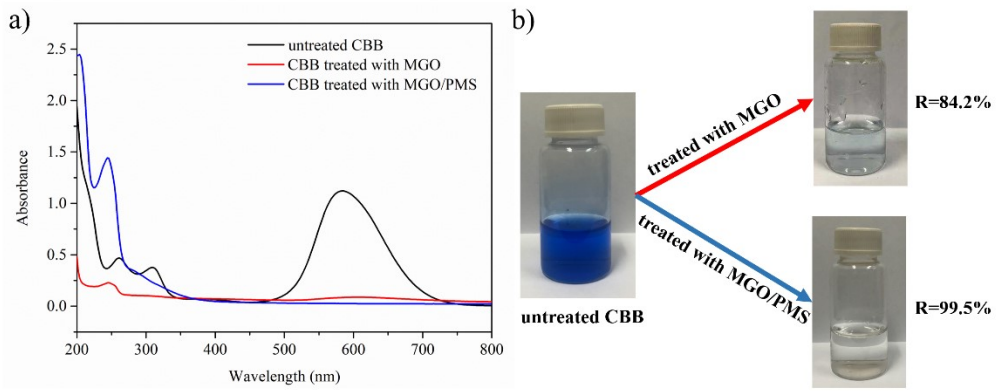

Figure S2 The UV-visible spectra of parent CBB, CBB after adsorption and CBB after catalytic oxidation (a) and the optical photos before and after treatment (b)

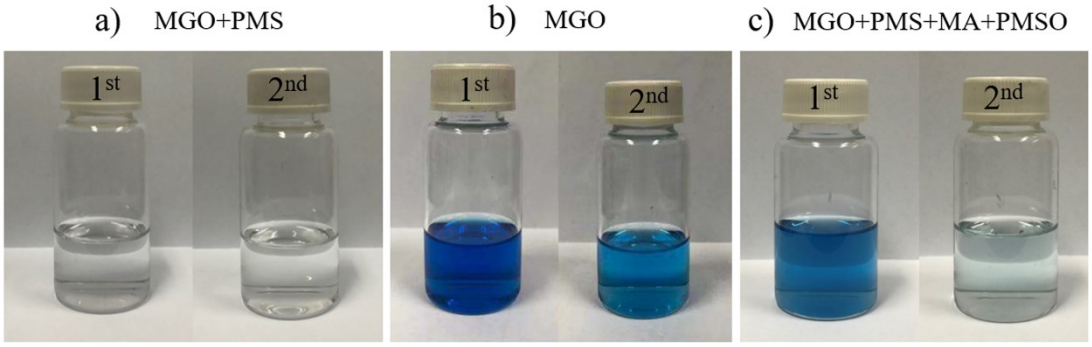

Figure S3 Optical photos of the ethanol samples after two consecutive extractions of spent MGO in different reaction systems

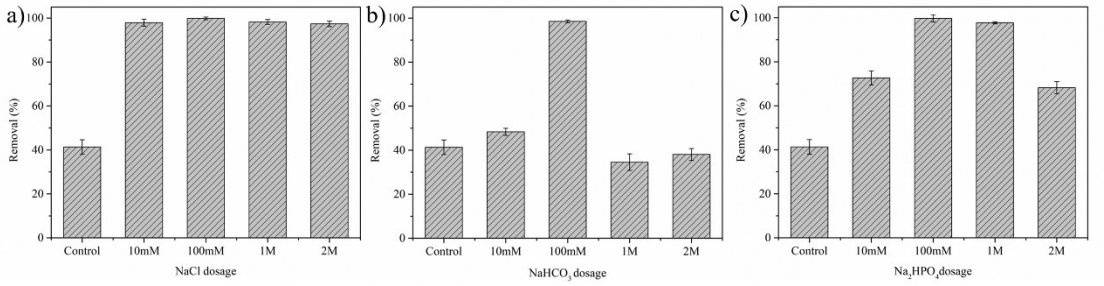

Figure S4 Effects of Cl^- (a), HCO_3^- (b) and HPO_4^{2-} (c) on CBB oxidation with PMS only. Experimental conditions: PMS dosage = 2 mM, initial CBB concentration = 50 mg/L, temperature = 25 °C.

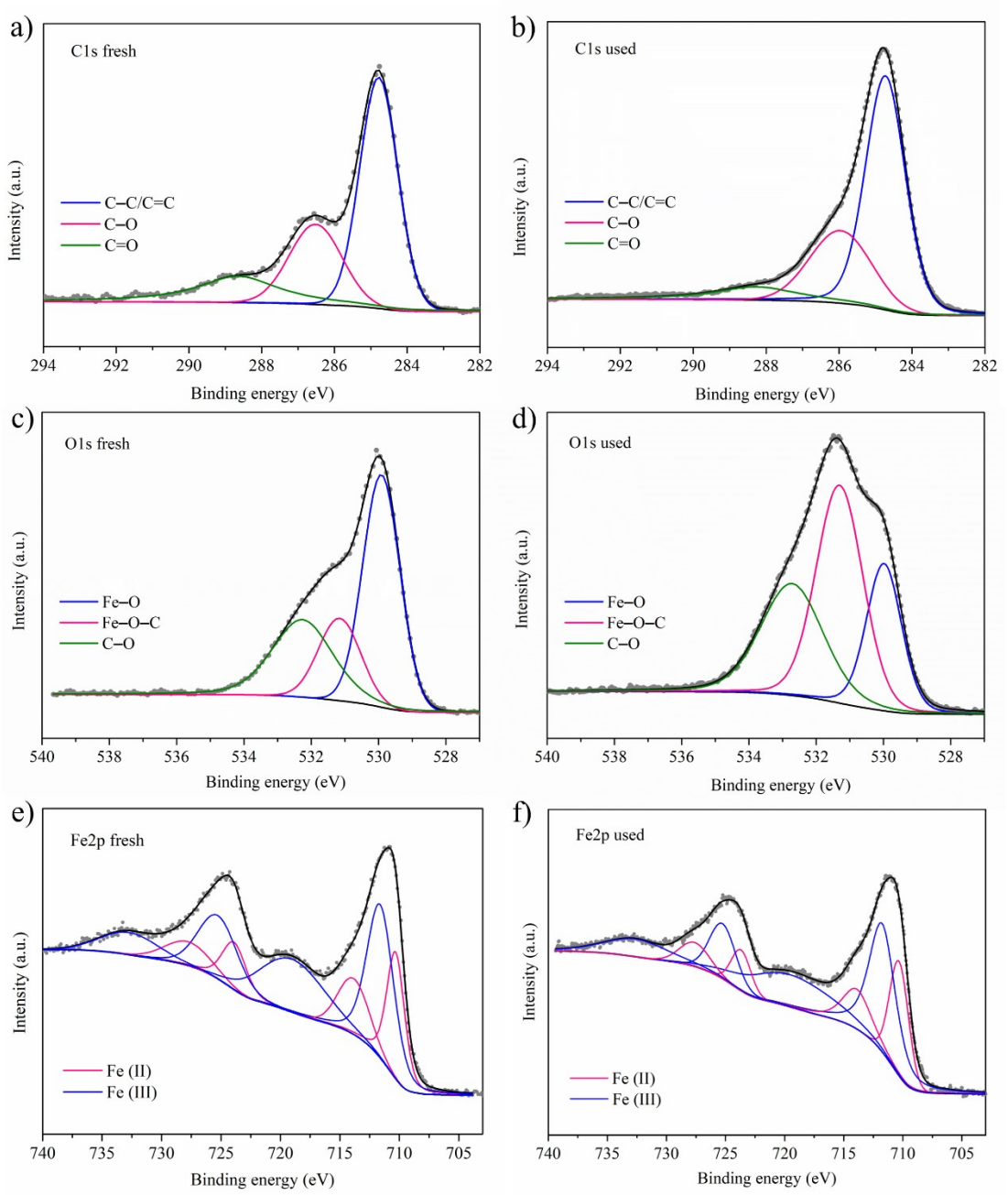

Figure S5 Deconvolution of C1s (a, b), O1s (c, d) and Fe2p (e, f) spectra for fresh and used MGO

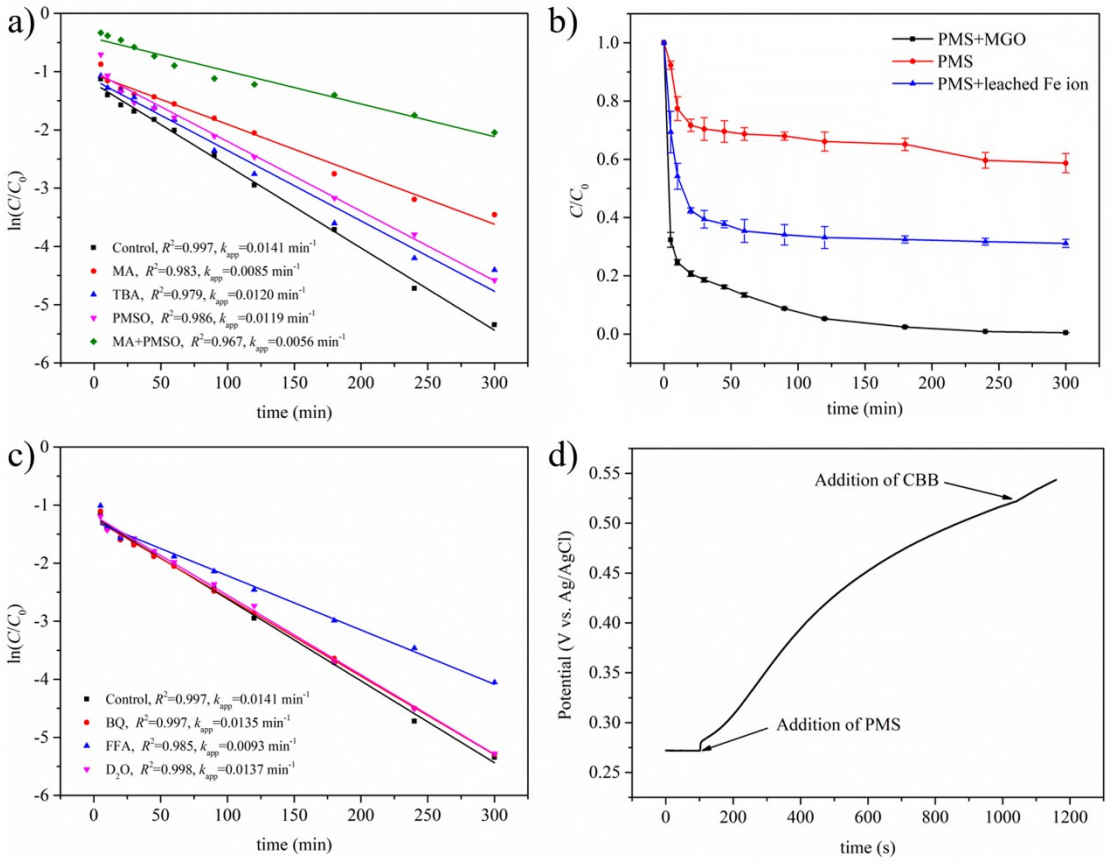

Figure S6 Pseudo-first-order model fitting of CBB removal in the MGO/PMS system with MA, TBA and PMSO (a), effect of leached ions on CBB removal (b), pseudo-first-order model fitting of CBB removal with BQ, FFA and D_2O (c), and the open circuit potential-time curve on MGO-GCE (d).

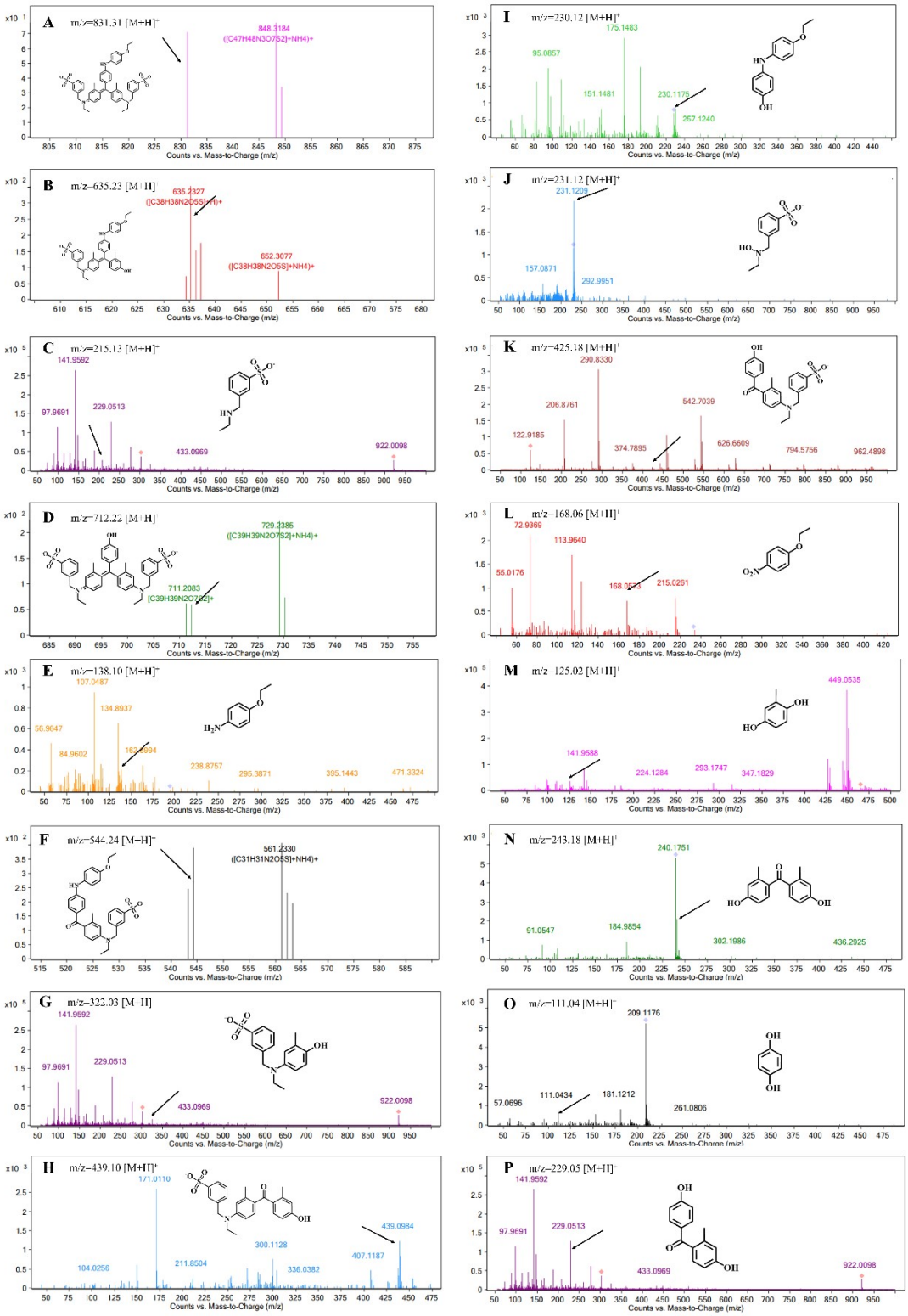

Figure S7 MS data of the CBB sample after treatment with the MGO/PMS system

Table S1 Properties of the water matrices employed in this work.

	pH	UV254 (AU/cm)	Conductivity (mS/cm)
DI water	6.51	ND	$<10^{-4}$
Tap water	7.69	0.038	0.234
Lake water	7.41	0.081	0.701
Sea water	7.97	0.024	24.7

Table S2 XPS deconvolution results of C1s, O1s and Fe2p spectra for fresh and used MGO

Sample	C1s			O1s			Fe2p	
	C–C/C=C (%)	C–O (%)	C=O (%)	Fe-O (%)	Fe-O-C (%)	C-O (%)	Fe(II) (%)	Fe(III) (%)
fresh	54.7	25.8	19.5	49.6	20.0	30.4	34.8	65.2
used	62.6	27.5	9.9	23.3	44.7	32.0	33.7	66.3

Table S3 Condensed Fukui indexes of f_{OW}^- and f_{OW}^0 in CBB

Atom index	f_{OW}^-	f_{OW}^0	Atom index	f_{OW}^-	f_{OW}^0
1(C)	0.01419	0.04283	55(S)	0.01374	0.00918
2(C)	0.01614	0.01978	56(O)	0.03632	0.01923
3(C)	0.01192	0.02179	57(O)	0.03448	0.01796
4(C)	0.01210	0.02086	58(O)	0.03365	0.01733
5(C)	0.01627	0.01813	59(C)	0.00373	0.00398
7(C)	0.01572	0.01813	63(N)	0.02845	0.02137
9(C)	0.01087	0.02040	64(C)	0.00467	0.00337
12(C)	0.01918	0.02030	67(C)	0.00417	0.00279
13(C)	0.01403	0.01984	71(C)	0.00577	0.00516
14(C)	0.01418	0.01988	74(C)	0.01518	0.01664
15(C)	0.01945	0.01800	75(C)	0.01406	0.01721
17(C)	0.02021	0.01842	76(C)	0.01510	0.01747
18(C)	0.01165	0.01757	77(C)	0.01355	0.01602
21(C)	0.01855	0.02065	79(C)	0.01570	0.01763
22(C)	0.01320	0.02059	81(C)	0.01461	0.01663
23(C)	0.01379	0.02031	84(S)	0.01385	0.00914
24(C)	0.01919	0.01803	85(O)	0.03614	0.01901
26(C)	0.01983	0.01882	86(O)	0.03293	0.01687
27(C)	0.01078	0.01793	87(O)	0.03595	0.01879
30(C)	0.00402	0.00409	88(N)	0.02233	0.01954
34(N)	0.02748	0.02245	90(C)	0.01689	0.01531
35(C)	0.00439	0.00340	91(C)	0.01617	0.01726
38(C)	0.00384	0.00263	92(C)	0.01543	0.01667
42(C)	0.00511	0.00508	93(C)	0.01670	0.01595
45(C)	0.01449	0.01707	95(C)	0.01698	0.01645
46(C)	0.01420	0.01811	97(C)	0.01549	0.01453
47(C)	0.01301	0.01684	100(O)	0.02002	0.01175
48(C)	0.01517	0.01770	101(C)	0.00356	0.00211
50(C)	0.01299	0.01627	104(C)	0.00303	0.00178
52(C)	0.01368	0.01713			