

## **Supplementary Materials**

Technological optimization for H<sub>2</sub>O<sub>2</sub> electro-synthesis and economic evaluation on electro-Fenton for treating refractory organic wastewater

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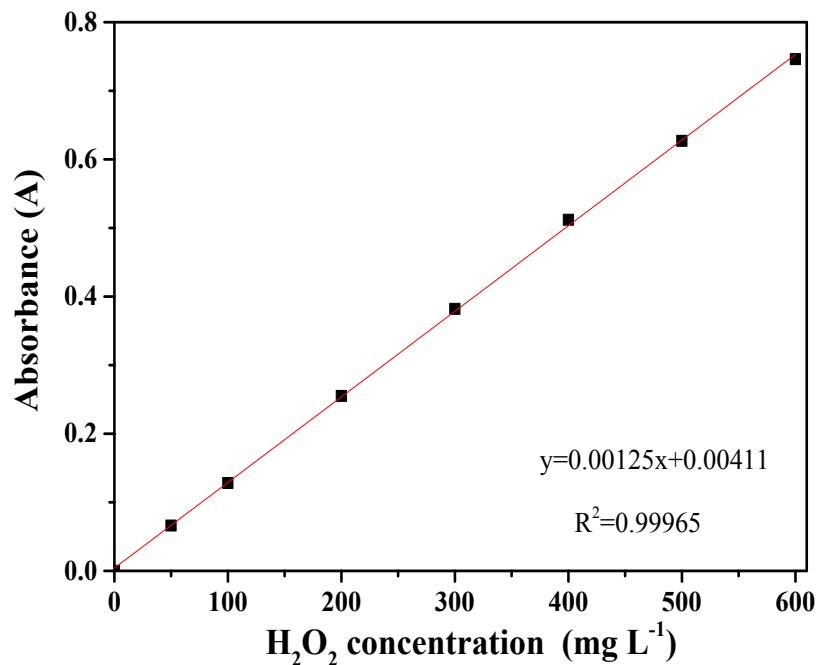
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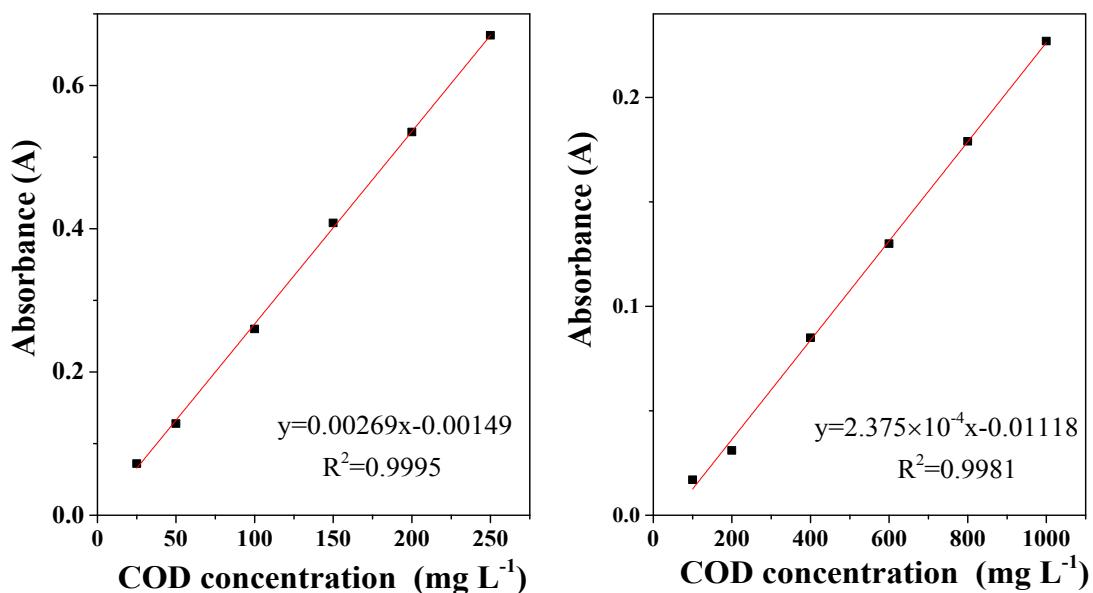
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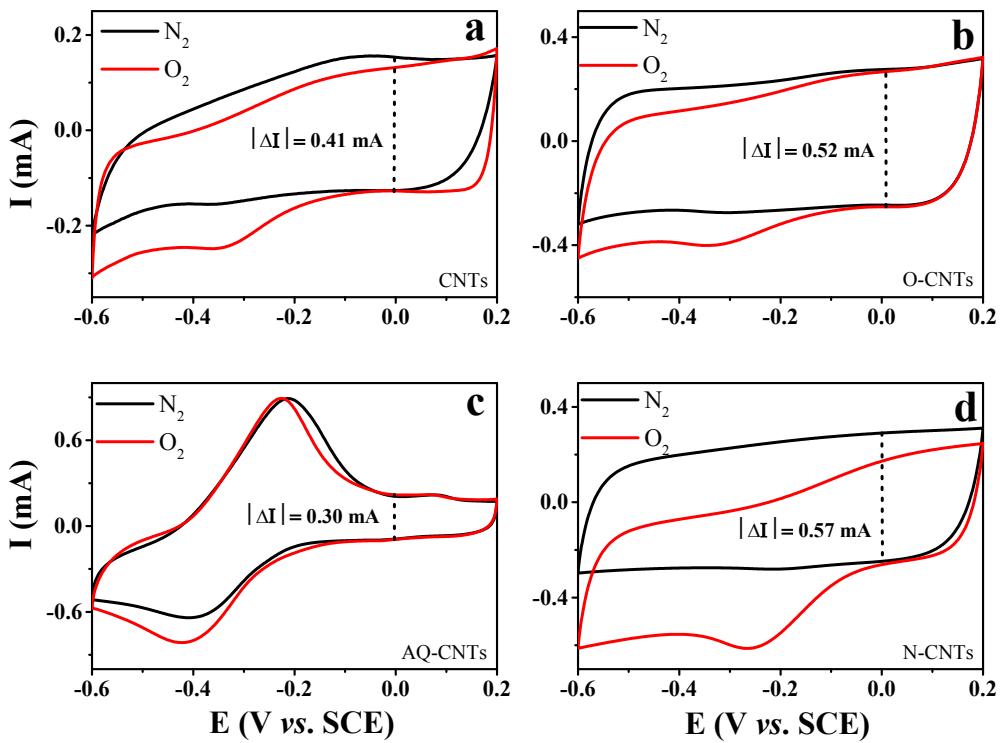
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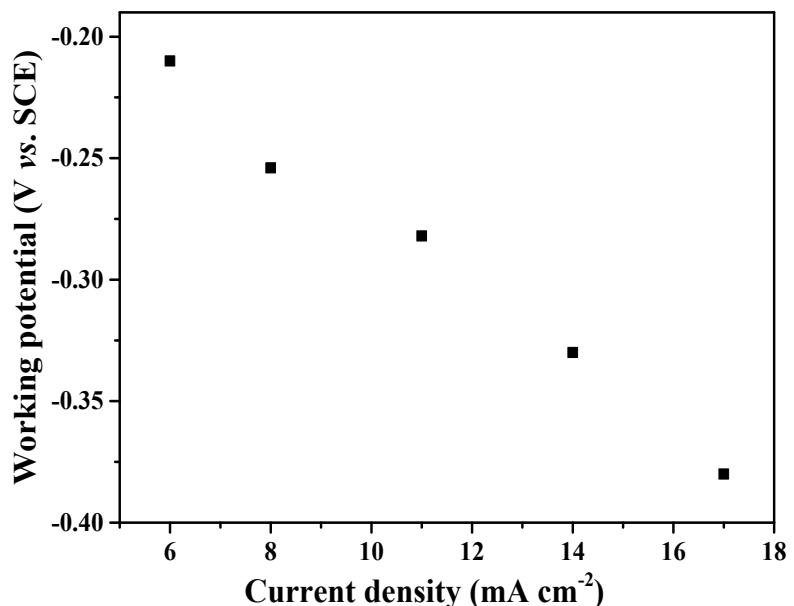
**Figure S1.** Standard curve of  $\text{H}_2\text{O}_2$  concentration and absorbance.



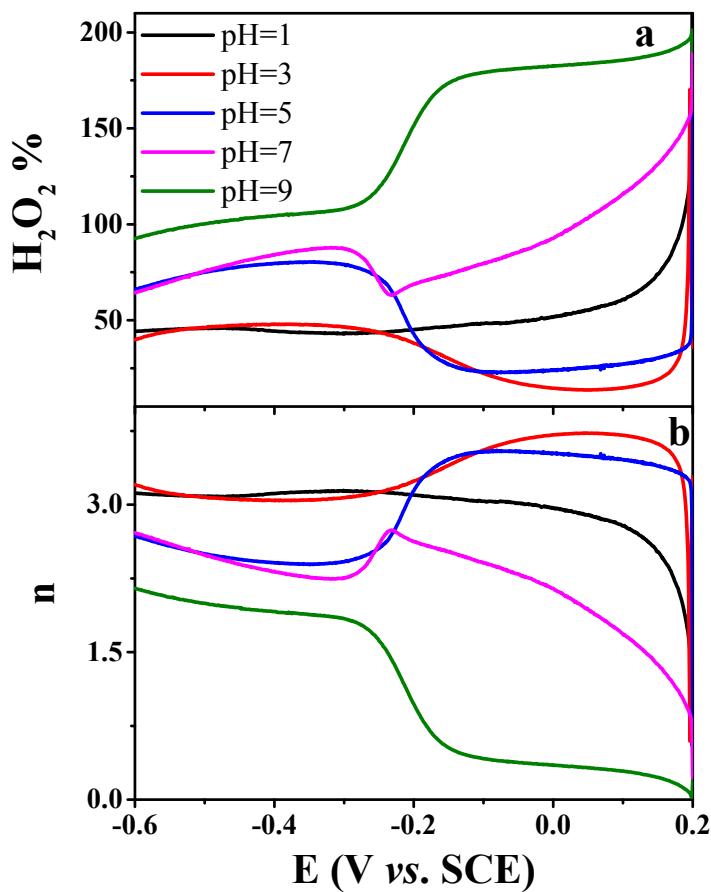
**Figure S2.** Standard curve of COD concentration and absorbance of (a) Low range ( $15\sim 250 \text{ mg L}^{-1}$ ), (b) High range ( $100\sim 1000 \text{ mg L}^{-1}$ ).



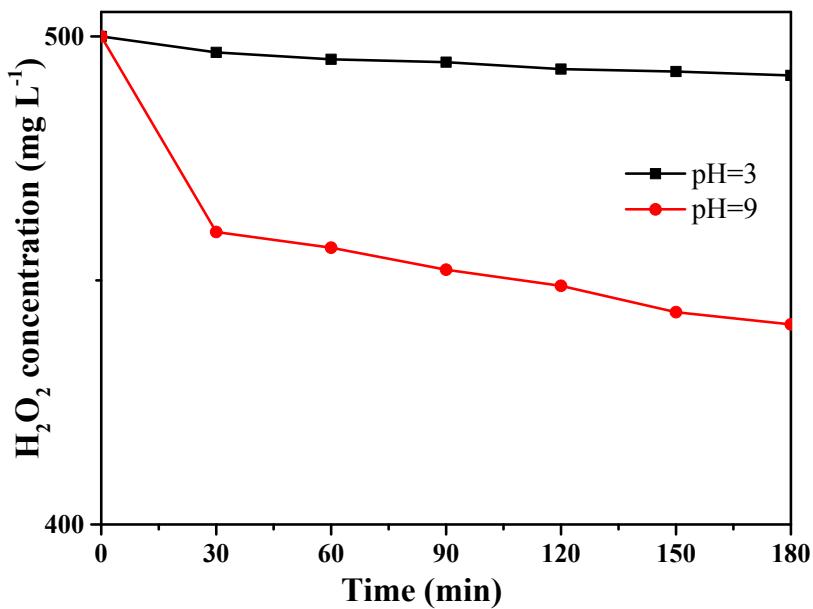
**Figure S3.** Cyclic voltammograms of (a) CNTs, (b) O-CNTs, (c) AQ-CNTs, and (d) N-CNTs performed in  $\text{N}_2$ -saturated (in black) and  $\text{O}_2$ -saturated (in red) 0.1 M  $\text{Na}_2\text{SO}_4$  (pH: 3) at 50 mV s<sup>-1</sup>.



**Figure S4.** The working potential of GDE measured at different current density.



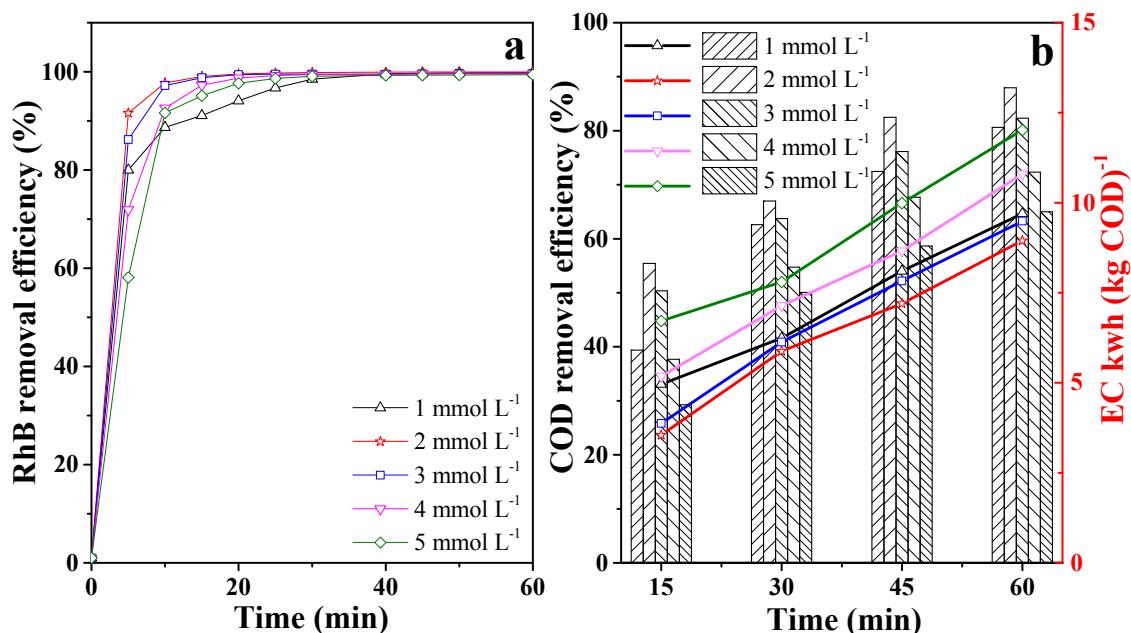
**Figure S5.** (a)  $H_2O_2$  selectivity and (b) number of transferred electrons ( $n$ ) for oxygen reduction reaction on O-CNTs measured on RRDE (1600 rpm) during linear scanning voltammetry (LSV) scanning at  $5 \text{ mV s}^{-1}$  in  $0.1 \text{ M Na}_2\text{SO}_4$  at different pH.



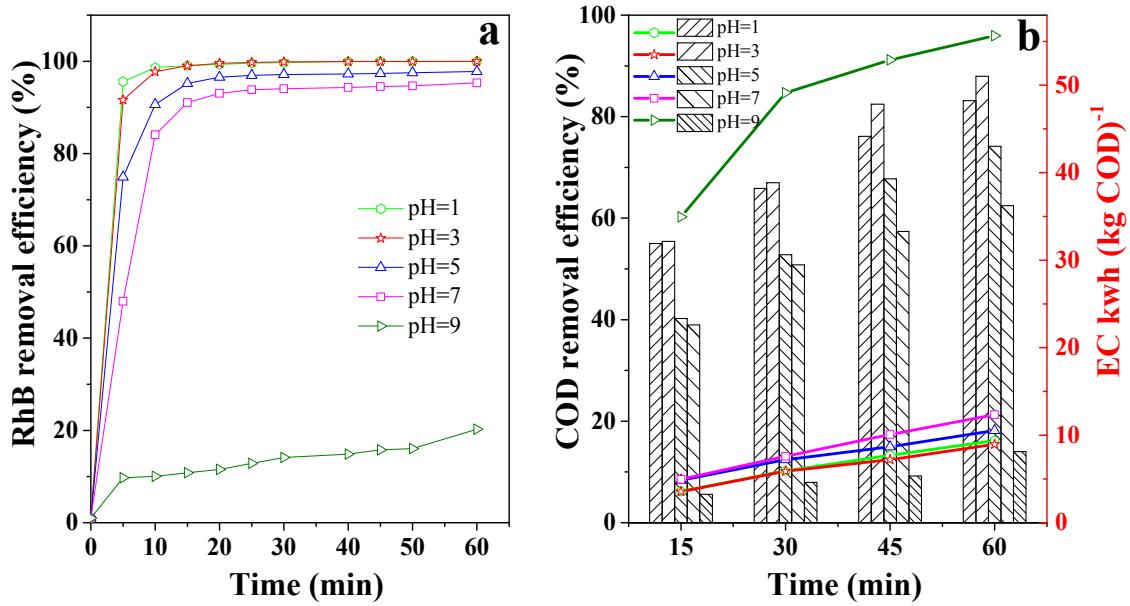
**Figure S6.**  $H_2O_2$  concentration varying with time at pH 3 and 9.

**Table S1.**  $\text{H}_2\text{O}_2$  yield collected from published paper.

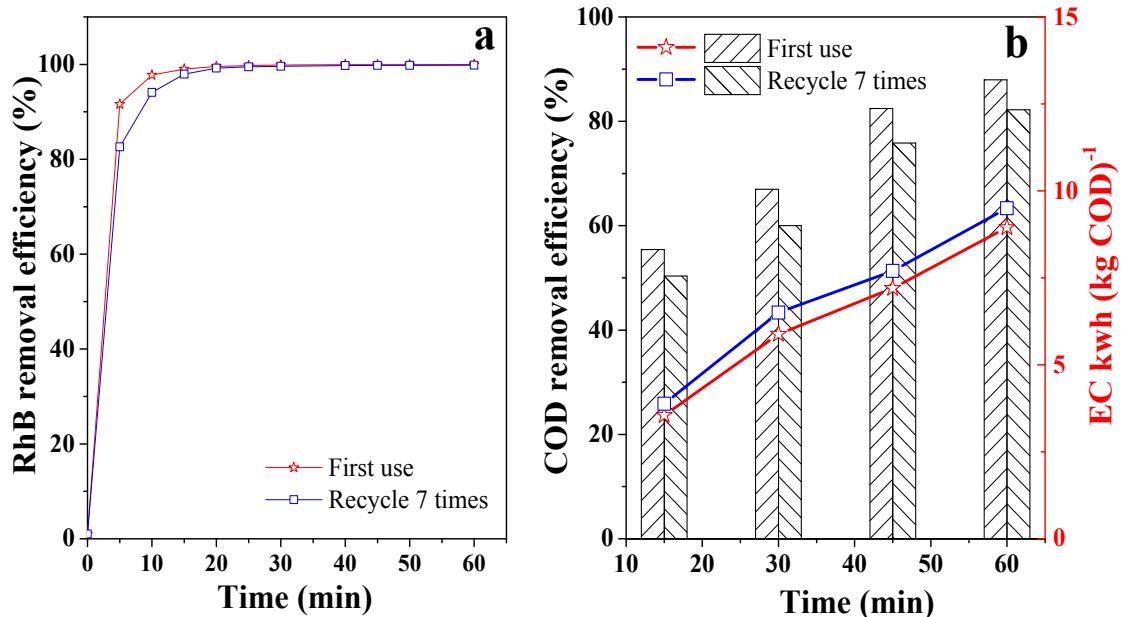
Cathode material	pH	j or E	Gas flow rate ( $\text{L min}^{-1}$ )	$\text{H}_2\text{O}_2$ yield ( $\text{mg L}^{-1} \text{h}^{-1}$ )	$\text{H}_2\text{O}_2$ yield ( $\text{mg h}^{-1} \text{cm}^{-2}$ )	Ref.
MC	3	$150 \text{ mA cm}^{-2}$	-	92.2	14.3	1
TBAQ-CNT	3	$7.1 \text{ mA cm}^{-2}$	0.4 (air)	100.4	4.24	2
CNTs	3	-0.5 V vs. SCE	0.4 ( $\text{O}_2$ )	24	0.6	3
TBAQ-CB	-	-1.0 V vs. SCE	-	200	4	4
CoPc	3	-0.4V vs. Ag/AgCl	-	220	4.97	5
Ta <sub>2</sub> O <sub>5</sub> -C	2	-0.1V vs. Ag/AgCl	0.6 (air)	14	1.64	6
N-G@CNT	3	-0.2 V vs. SCE	-	10.5	0.26	7
FePc	-	-1.0 V vs. Ag/AgCl	-	160	3.6	8
O-CNTs	3	$14 \text{ mA cm}^{-2}$	0.6 ( $\text{O}_2$ )	304	6.7	This work



**Figure S7.** (a) RhB removal efficiency and (b) COD removal efficiency and energy consumption at different  $\text{Fe}^{2+}$  concentration. Conditions:  $0.1 \text{ mol L}^{-1} \text{ Na}_2\text{SO}_4$ , pH=3, [RhB]:  $200 \text{ mg L}^{-1}$ , current density:  $14 \text{ mA cm}^{-2}$ ,  $\text{O}_2$  flow rate: $0.6 \text{ L min}^{-1}$ .



**Figure S8.** (a) RhB removal efficiency and (b) COD removal efficiency and energy consumption at different solution pH. Conditions:  $0.1 \text{ mol L}^{-1} \text{ Na}_2\text{SO}_4$ ,  $[\text{Fe}^{2+}]$ : 2 mM,  $[\text{RhB}]$ :  $200 \text{ mg L}^{-1}$ , current density:  $14 \text{ mA cm}^{-2}$ ,  $\text{O}_2$  flow rate:  $0.6 \text{ L min}^{-1}$ .



**Figure S9.** The reusability of GDE evaluated after 7-time runs in electro-Fenton treatment of RhB wastewater. Conditions:  $0.1 \text{ mol L}^{-1} \text{ Na}_2\text{SO}_4$ ,  $[\text{Fe}^{2+}]$ : 2 mM, pH=3,  $[\text{RhB}]$ :  $200 \text{ mg L}^{-1}$ , current density:  $14 \text{ mA cm}^{-2}$ ,  $\text{O}_2$  flow rate:  $0.6 \text{ L min}^{-1}$ .

**Table S2. The necessary data for calculation of energy consumption.**

Reaction time (min)	slot voltage (V)	Current (A)	Initial COD (mg L <sup>-1</sup> )	COD at time t(mg L <sup>-1</sup> )	Solution volume (L)
15				117.7	
30	3.3	0.25	264	87.2	0.4
45				50	
60				31.8	

**Table S3. Data comparison for energy consumption and COD removal efficiency.**

wastewater	j (mA cm <sup>-2</sup> )	pH	[Fe <sup>2+</sup> ]	COD removal efficiency(%)	EC (kWh kg <sub>COD</sub> <sup>-1</sup> )	Ref.
DMP	-	3	-	-	20~40	9
RhB	20	3	0.3 M	79	80	10
landfill leachate	15	3	560 mg L <sup>-1</sup>	71±6	207±20	11
RhB	-	6.2	3 mM	-	17	12
RhB	14	3	2 mM	88	9	This work

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