Supporting Information

Application of Carbon Nanotubes Coated Electrodes and Immobilized TiO_2 for Dye Degradation in a Continuous Photocatalytic-Electro-Fenton Process

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Table S1. Studies Using Various Electrodes Modified with CNTs

order	electrode type	modification method	application	ref.
1	pencil graphite (PG)	Immersed into Eppendorf tubes that contained 110 μL of 3000 μg/mL CNTs solution for 1 h in order to form a thin CNTs layer on the PG surface by wet-adsorption	monitoring the interaction between 4-Nonylphenol and DNA	1
2	fluorine doped tin oxide (FTO)	a) Chemical, b) Thermal chemical vapor deposition and c) Plasma enhanced chemical vapor deposition	solar cells	2
3	glassy carbon	Polishing with alumina slurries, washing and sonicating. then 1 mg/mL CNTs:H_2SO_4 was cast on the electrode surface and dried at 200 ˚C for 3 h	NADH detection	3
4	glassy carbon disk (GCD)	Dispersion of CNTs in 1 mL DMF was cast on GCD and dried at room temperature	electrochemical detection of trace insulin	4
5	glassy carbon	Dispersion of CNTs in 1 mL dimethylformamide and dried at room temperature	detection of 2,4,6-trinitrotoluene	5
6	carbon fiber	Dipping the electrode into DMF solution containing 4 mg/mL CNTs for 20 s, drying for 30 min, followed by three, 2 s dippings (with an intermediate 10 min drying)	determination of hydrogen sulfide	6
7	glassy carbon	1 mg CNTs was dispersed in an aqueous 0.1% Nafion solution and was dropped on the GC electrode surface and then the solvent was evaporated under an infrared heat lamp	detection of phenolic estrogenic compounds	7
8	graphite powder	Graphite powder, CNTs and mineral oil were mixed. Then the osmium redox polymer (10 mg/mL) was spread over its surface. The cellular paste was spread on the electrode and it was covered with a dialysis membrane	microbial biosensor	8
9	edge plane pyrolytic graphite	Dropping the dispersion of CNTs onto the polished electrode	determination of paracetamol	9
10	pyrolytic graphite	Carboxylated CNTs dispersion was cast onto the surface of a polished (PG)	reduction of nitrobenzene	10
11	graphite plates	Two components of epoxy conductive adhesive were blended on graphite plates. CNTs and carbon nanopowder were sprayed separately on the electrodes and dried	enhanced power generation	11
12	graphite plates	Ultrasonication of solution containing CNTs/CTAB and further electrodeposition using DC voltage of 17.5 (V)	methylene blue detection and enhanced capacity	12

Table S2. Studies Used Modified Graphite Electrodes for Electrochemical Wastewater Treatment

order	graphite type	used as	modified with	modification method	pollutant	ref.
1	graphite	cathode	polypyrrole/ anthraquinone disulphonate composite film	—	amaranth azo dye	13
2	commercial graphite felt	cathode	—	Anodization in a phosphate buffer (pH=6.88) at 1.8V vs. SCE for 5 min, followed by cathodization at -1.5V vs. SCE for 1 min/ Dipping the graphite in a solution 0.5 M of H_2SO_4 for 30 min	wastewater from the regeneration of ion-exchange resin towers by a chemical industry	14
3	graphite felt	cathode	—	Electrochemically oxidized in H_2SO_4 solution	drinking water	15
4	graphite powder	cathode	polytetrafluoro ethylene (PTFE)	Graphite powder and PTFE dispersion were mixed and ethanol was added as solvent at 80 ˚C. Then it was pulverized to form a sheet which was pressed onto a stainless steel mesh under a pressure of 15 MPa for 3 min. Then it was calcined at 300 ˚C for 2h and immersed in acetone and water five times, followed by air drying	C.I. Acid Red 2	16
5	expanded graphite	anode	Attapulgite	The dispersed acid activated attapulgite was added to expanded graphite and dried at 105 °C for 3 h, and pressed with a performing machine	textile wastewater	17
6	CNTs or graphite	cathode	nitrogen functionalized CNTs	CNTs (or graphite) and PTFE were dispersed. Ethanol was added as solvent and mixed at 80 ˚C. PTFE cake was pulverized to form a thin film and pressed onto stainless steel mesh under the pressure of 15MPa for 3 min	methyl orange	18
7	expanded graphite	working electrode	palygorskite	EG and paraffin and palygorskite were mixed and heated to 65 °C to melt the paraffin. Then it was filled into a glass tube	phenol	19
8	graphite plates	cathode	activated carbon- PTFE or carbon nanotube-PTFE	Activated carbon or carbon nanotube, PTFE (0.84 g), distilled water and n-butanol were mixed, heated at 80 ˚C and sintered at 350 ˚C for 15 min	C.I. Acid Yellow 36	20
9	graphite plate	cathode	CNTs	Ultrasonication of solution containing CNTs/CTAB and further electrodeposition using DC voltage of 17.5 (V)	Acid Red 14, Acid Blue 92	21

Table S3. Dyes Structures

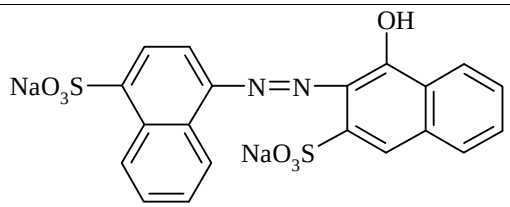
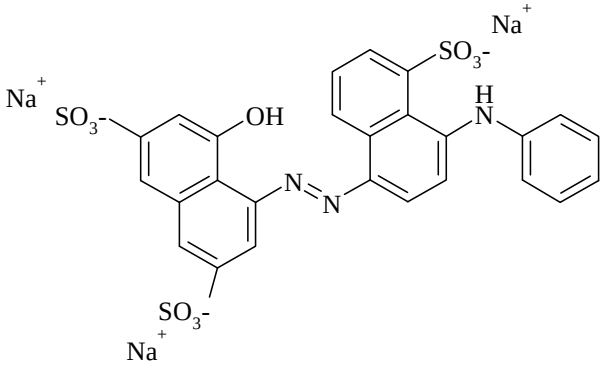
	C.I. Acid Red 14 ($C_{20}H_{12}N_2O_7S_2 \cdot 2Na$) λ_{max}= 515 (nm)
	C.I. Acid Blue 92 ($C_{26}H_{16}N_3O_{10}S_3 . 3Na$) λ_{max}= 572 (nm)

Table S4. CCD Matrix for the PEF-TiO_2 Process

run order	parameters				Na_2SO_4 as electrolyte				NaCl as electrolyte			
	time (X_1, min)	$[Fe^{3+}]$ (X_2, mM)	CD (X_3, mA/cm^2)	$[C_0]$ (X_4, mg/L)	decolorization (%)		COD removal (%)	EEC (kWh/g_{COD})	decolorization (%)		COD removal (%)	EEC (kWh/g_{COD})
					AR14	AB92			AR14	AB92		
1	120	0.15	12.95	50	72.18	60.30	62.88	0.426	79.68	85.31	70.71	0.378
2	100	0.2	15.30	70	67.51	35.44	53.77	0.320	67.10	68.89	58.56	0.294
3	80	0.05	12.95	50	47.64	50.70	40.27	0.443	57.00	68.54	50.93	0.350
4	80	0.15	12.95	50	69.04	58.79	55.76	0.320	70.42	82.96	64.59	0.276
5	60	0.2	10.60	70	43.15	40.65	37.01	0.246	53.35	58.01	48.02	0.190
6	100	0.2	10.60	30	73.27	71.59	63.99	0.526	79.01	70.24	69.84	0.482
7	60	0.1	10.60	70	45.51	28.01	37.09	0.246	44.66	45.42	38.57	0.236
8	60	0.2	15.30	70	60.70	23.68	49.71	0.207	57.67	60.08	50.30	0.205
9	40	0.15	12.95	50	62.48	33.93	50.99	0.175	57.48	47.10	52.13	0.171
10	80	0.15	8.25	50	56.63	36.16	46.85	0.333	55.07	73.13	52.89	0.295
11	60	0.1	10.60	30	69.64	27.11	43.78	0.461	58.52	71.59	53.37	0.378
12	80	0.15	12.95	10	79.89	65.64	77.32	0.900	80.50	92.92	87.35	0.758
13	80	0.15	17.65	50	62.57	19.74	52.54	0.381	65.46	78.64	58.42	0.343
14	80	0.15	12.95	50	69.94	56.79	54.73	0.326	71.55	83.09	63.87	0.279
15	80	0.15	12.95	50	71.24	56.05	55.57	0.321	69.54	81.44	63.01	0.283
16	80	0.15	12.95	50	68.84	57.93	54.89	0.325	72.83	83.68	65.65	0.272
17	80	0.15	12.95	50	70.58	55.99	56.51	0.316	70.90	82.53	64.93	0.275
18	60	0.2	15.30	30	67.24	28.25	53.77	0.425	84.18	56.37	75.96	0.301
19	100	0.1	15.30	30	62.78	55.69	52.69	0.723	63.89	98.26	60.03	0.634
20	80	0.25	12.95	50	52.13	64.96	47.35	0.377	72.68	49.85	65.98	0.270
21	80	0.15	12.95	50	71.22	59.29	56.53	0.316	70.42	82.96	64.17	0.278
22	60	0.1	15.30	70	55.11	20.78	41.71	0.247	47.64	45.24	41.47	0.249
23	100	0.2	15.30	30	77.33	40.93	67.51	0.574	87.67	68.79	78.32	0.486
24	100	0.2	10.60	70	64.18	62.56	54.50	0.279	63.62	69.05	57.58	0.264
25	60	0.2	10.60	30	49.98	32.09	41.51	0.486	74.66	54.16	66.74	0.303
26	80	0.15	12.95	90	58.19	45.54	38.25	0.316	57.50	69.49	38.57	0.329
27	60	0.1	15.30	30	63.23	44.27	51.99	0.439	58.47	86.56	54.75	0.417
28	100	0.1	10.60	70	54.53	46.06	44.73	0.340	55.63	62.73	49.84	0.305
29	80	0.15	12.95	50	69.19	57.32	54.75	0.326	70.81	81.56	64.63	0.276
30	100	0.1	10.60	30	73.15	43.24	58.68	0.573	62.37	96.97	60.24	0.559
31	100	0.1	15.30	70	56.49	27.43	44.84	0.383	59.38	66.33	53.51	0.321

Table S5. Coefficients of Significant Factors to the Response Functions

		Na_2SO_4 as electrolyte				NaCl as electrolyte			
		decolorization (%)		COD removal (%)	EEC (kWh/g_{COD})	decolorization (%)		COD removal (%)	EEC (kWh/g_{COD})
		AR14	AB92			AR14	AB92		
1	constant	70.0098	57.449	55.5354	0.321289	70.9244	82.606	64.4081	0.277022
2	t	3.9193	7.952	4.4963	0.060836	4.3298	8.3434	3.9963	0.061671
3	$[Fe^{3+}]$	1.3279	2.964	2.5169	-0.02007	6.1699	-4.3704	5.1527	-0.03067
4	CD	2.0362	-4.487	1.9206	0.010682	2.2897	1.3907	1.6565	0.011899
5	$[C_0]$	-5.5355	-4.114	-6.1948	-0.1295	-6.9051	-7.2509	-9.1237	-0.09811
6	t^2	-0.6666*	-3.898	-0.0215*	-0.00761*	-0.9296*	-4.6345	-1.1315*	-0.00229*
7	$[Fe^{3+}]^2$	-5.0272	-1.219*	-3.302	0.019797	-1.8638	-6.387	-1.8723	0.006594
8	CD^2	-2.5977	-8.689	-1.8309	0.006632*	-3.0085	-2.2137	-2.5721	0.008762
9	$[C_0]^2$	-0.2389*	-1.779*	0.1916*	0.069324	-0.8236*	-0.8836*	-0.7458*	0.064813
10	$t.[Fe^{3+}]$	2.9847	2.1*	1.9627	-0.01826	-0.278*	-1.6945	-0.7612*	-0.0007*
11	t.CD	-2.4391	-3.317	-2.5549	0.025156	0.0393*	-0.9881*	-0.1788*	0.003836*
12	$t.[C_0]$	0.1129*	-1.335*	-1.218*	-0.0131	1.5816	-0.4572*	1.469*	-0.02865
13	$[Fe^{3+}].CD$	2.9634	-5.145	1.2996*	-0.01149*	1.1102*	-1.0628*	0.8275*	-0.006*
14	$[Fe^{3+}].[C_0]$	1.5555	2.343*	0.4361*	0.001357*	-2.9897	8.758	-1.9636	0.016154
15	$CD.[C_0]$	1.7446	-4.067	-0.082*	-0.00416*	-0.3201*	-0.7311*	-0.5638*	-0.00272*

* The insignificant parameters

Table S6. Comparison of the Present Study with Other Researches

	treatment method	pollutant	reactor	electrode material	electrolyte	flow rate or time	CD (mA/cm^2)	decolorization (%)	ref.
1	PEF-TiO_2	Acid Red 14 (6 mg/L)	batch (1 L)	anode: Pt sheet cathode: graphite-CNTs/ immobilized TiO_2 on sand blasted glass plates by sol–gel dip-coating	Na_2SO_4 0.05 M/ Fe^{3+} 0.2 mM	120 min	2.77	99	22
2	Electro-Fenton	simulated industrial textile wastewater	continuous (1.8 L)	anode: iron cathode: stainless steel	NaCl 25 g/L	0.33 L/h	123.97	99.3	23
3	PEF-TiO_2	Direct Yellow 12 (50 mg/L)	batch (2.5 L)	anode: Pt sheet cathode: CNT-PTFE/ immobilized ZnO nanoparticles on glass plates	Na_2SO_4 0.05 M/ Fe^{3+} 0.2 mM	70 min	34.78	97	24
4	Electro-Fenton	Lissamine Green B (8.5 mg/L)	continuous bubble reactor (0.675 L)	anode, cathode: graphite bars (surface area of 1.27 cm^2)	Na_2SO_4 0.01 M/ Fe^{2+} 150 mg/L	0.048 L/h	potential difference (15V)	80	25
5	PEF-TiO_2	Acid Red 14 and Acid Blue 92 (48.70 mg/L)	continuous bubble reactor (1 L)	anode: graphite cathode: graphite-CNTs/ immobilized TiO_2 on UV resistant silicone polymer	NaCl 1 g/L/ Fe^{3+} 0.15 mM	0.825 L/h	12.95	AR14: 94.47 AB92: 98.98	this study

For the binary solutions, the extension of Beer-Lambert law (eq S1) was used:[26,27]

$$A_{\lambda_j} = \sum_{i=1}^{2} \varepsilon_{\lambda_j}^{i} C^{i} L \qquad \text{(S1)}$$

in which, A_{λ_j} is the absorbance of a mixture of 2 components at the *j*th wavelength, C^i is the concentration of the *i*th component, $\varepsilon_{\lambda_j}^{i}$ is the extinction coefficient of the *i*th component at the *j*th wavelength, and L is the cell path length. According to this expression, two equations and two readings of absorbance at two different wavelengths are needed to determine the concentration of two dyes in a mixture. Finally, the involving of the resolution of a matrix system is expressed as eq S2 (assuming L = 1 cm): [26,27]

$$\begin{bmatrix} A_{\lambda_R} \\ A_{\lambda_B} \end{bmatrix} = \begin{bmatrix} \varepsilon_{\lambda_R}^{R} & \varepsilon_{\lambda_R}^{B} \\ \varepsilon_{\lambda_B}^{R} & \varepsilon_{\lambda_B}^{B} \end{bmatrix} \times \begin{bmatrix} C^R \\ C^B \end{bmatrix} \qquad \text{(S2)}$$

The resolution of this two-equation system gives the expressions (eq S3) for the calculation of the concentrations of AR14 (C^R) and AB92 (C^B) in the mixture: [26,27]

$$C^B = \frac{A_{\lambda_B}\varepsilon_{\lambda_R}^{R} - A_{\lambda_R}\varepsilon_{\lambda_B}^{R}}{\varepsilon_{\lambda_B}^{B}\varepsilon_{\lambda_R}^{R} - \varepsilon_{\lambda_R}^{B}\varepsilon_{\lambda_B}^{R}}; \quad C^R = \frac{A_{\lambda_R}\varepsilon_{\lambda_B}^{B} - A_{\lambda_B}\varepsilon_{\lambda_R}^{B}}{\varepsilon_{\lambda_B}^{B}\varepsilon_{\lambda_R}^{R} - \varepsilon_{\lambda_R}^{B}\varepsilon_{\lambda_B}^{R}} \qquad \text{(S3)}$$

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