

## Supporting Information

# A Highly Active Based Graphene Cathode for Electro-Fenton Reaction

*Thi Xuan Huong Le, Mikhael Bechelany, Joffrey Champavert, Marc Cretin\**

<sup>1</sup> Institut Européen des Membranes, UMR 5635 CNRS Université Montpellier, Place Eugene Bataillon, F-34095 Montpellier cedex 5, France

\* Corresponding author: Marc.Cretin@univ-montp2.fr

## 1. Materials and method

### 1.1. Chemicals

The carbon felt was purchased from A Johnson Matthey Co., Germany. AO7 (Orange II sodium salt) was obtained from Sigma-Aldrich. Sulfuric acid (ACS reagent, 95.0 - 98.0%), sodium sulphate (anhydrous, 99.0 – 100.5%) and iron (II) sulphate hepta-hydrate (99%), were used at the purest grade.

### 1.2. Preparation of graphene/carbon felt cathode

The commercial carbon felt with a dimension of 5.0 cm x 1.0 cm x 1.27 cm was first cleaned in an ultrasonic bath with acetone for 2h to remove adsorbed contaminations, followed by thorough rinse with deionized water and dried at 60°C for 24h. This pretreated carbon felt was denoted as raw-CF. Graphite oxide was synthesized from graphite powder by modified Hummers method [13]. GO suspension was prepared by adding in water at a concentration of 1.5 mg/mL, followed by ultrasonication under the ambient condition (Bransonic 3510E-MT, 130 W) for 5h to obtain a homogeneous dispersion. All solutions were prepared in MilliQ water.

Graphene oxide was deposited on the raw-CF surface via an electrophoretic deposition in a two-electrodes cell with a platinum foil as the cathode and the raw carbon felt as the anode where negative particles of GO were pushed to adhere on the surface of CF. After that in the same cell, the reduction was conducted by reversing electrodes, so GO was reduced by a cathodic current on the surface of CF. The essential factors affecting the modification such as current density value, time of GO deposition on the CF surface and conditions of electrochemical reduction to form rGO were investigated. In this method, GO could be served also as weak supporting electrolyte so sodium sulfate or potassium chloride were not necessary to add in the solution. The modified electrode was noted as ECR-CF (Electrochemical Reduction-Carbon Felt).

### 1.3. Material characterization

Chemical and structural characterizations have been performed using a scanning electron microscopy (SEM, Hitachi S-4800), X-ray diffraction (PANalytical Xpert-PRO diffractometer equipped with a X'celerator detector using Ni-filtered Cu-radiation), AFM (AFM NANOMAN 5 from Veeco instrument controlled with a Nanoscope V software) and XPS (ESCALAB 250

Thermal Electron) with AlK $\alpha$  (1486.6 eV). Binding energies were calibrated by using the containment carbon (C1s = 284.4 eV).

Electrochemical characterization was investigated relying on cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS). Experiments were conducted in a solution of 10 mM K<sub>3</sub>[Fe(CN)<sub>6</sub>] and 1.0 M KCl by using the  $\mu$ 3AUT70466 Autolab system (Eco Chemie BV, Netherlands) and Bio-Logic SP-150 in a three-electrodes cell including a working electrode (modified and unmodified CF), a counter electrode (Pt foil), and a reference electrode (Saturated Calomel Electrode, SCE). The impedance spectra were recorded at the open circuit voltage (OCV) with frequency range from 50 kHz to 100 mHz and a voltage amplitude of 10 mV.

#### 1.4. Decolorization of Acid Orange 7 by EF process

The electro-Fenton treatment of AO7 aqueous solution was carried out in a 75 mL undivided two-electrodes cell using a power supply (Lambda Electronique, USA) to control the applied current. The prepared cathode (2 cm<sup>2</sup>) was used as working electrode and a platinum sheet as counter electrode. The distance between the working electrode and the counter electrode was 3 cm. The 30 mL aqueous solution of AO7 (0.1 mM), Na<sub>2</sub>SO<sub>4</sub> (50 mM) as supporting electrolyte and FeSO<sub>4</sub>·7H<sub>2</sub>O (0.2 mM) as the catalyst were prepared by adjusting pH at 3. Prior to the electrolysis, oxygen was bubbled to saturate the solutions which were stirred magnetically in a rate of 800 rpm. The decomposition of AO7 was monitored by measuring the absorbance of dye at the selected single wavelength  $\lambda = 485$  nm specified for azo bond, using Spectrophotometer Jenway 6300 (Barioworld Scientific Ltd, Dunmow UK). The absorbance of this bond was proportional to the AO7 concentration, according to the Beer-Lambert Law.

## 2. Optimization for preparation ECR-CF

Table S2 – Anodic current (I) and potential peak (E) of redox wave [Fe(CN)<sub>6</sub>]<sup>3-</sup>/[Fe(CN)<sub>6</sub>]<sup>4-</sup> as the function of current density and time deposition or reduction.

	Electrophoretic deposition								
	Current density (mA/cm <sup>2</sup> )				Time (min)				
	0.75	1.5	3	6	5	10	20	30	40
<b>I (mA)</b>	11	26	23.2	22.4	15	26	26	21	20.2
<b>E(V vs. SCE)</b>	0.37	0.35	0.42	0.47	0.41	0.35	0.43	0.40	0.42
	Electrochemical reduction								
<b>I (mA)</b>	20	26	23	17.8	25	26	22	14.8	13.4
<b>E(V vs. SCE)</b>	0.46	0.35	0.54	0.54	0.57	0.35	0.35	0.46	0.47

