



## Efficient treatment of aniline containing wastewater in bipolar membrane microbial electrolysis cell-Fenton system

Li, Xiaohu; Jin, Xiangdan; Zhao, Nannan; Angelidaki, Irini; Zhang, Yifeng

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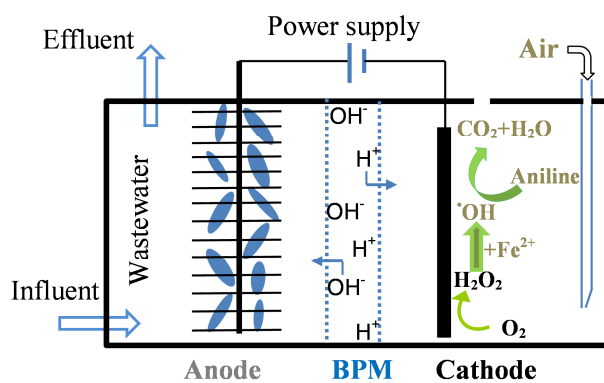
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1 Submission to Water Research

2 **Efficient treatment of aniline containing wastewater in bipolar**  
3 **membrane microbial electrolysis cell-Fenton system**

4 Xiaohu Li, Xiangdan Jin, Nannan Zhao, Irini Angelidaki, Yifeng Zhang\*

5 Department of Environmental Engineering, Technical University of Denmark, DK-2800 Lyngby,

6 Denmark

7

8 \*Corresponding author:

9 Dr. Yifeng Zhang

10 Department of Environmental Engineering, Technical University of Denmark, Denmark

11 Tel: (+45) 45251410.

12 Fax: (+45) 45933850.

13 E-mail address: yifz@env.dtu.dk; yifzmf@gmail.com

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21 **Abstract**

22 Aniline-containing wastewater can cause significant environmental problems and threaten  
23 the humans's life. However, rapid degradation of aniline with cost-efficient methods remains  
24 a challenge. In this work, a novel microbial electrolysis cell with bipolar membrane was  
25 integrated with Fenton reaction (MEC-Fenton) for efficient treatment of real wastewater  
26 containing a high concentration ( $4460 \pm 52 \text{ mg L}^{-1}$ ) of aniline. In this system,  $\text{H}_2\text{O}_2$  was in  
27 situ electro-synthesized from  $\text{O}_2$  reduction on the graphite cathode and was simultaneously  
28 used as source of  $\cdot\text{OH}$  for the oxidation of aniline wastewater under an acidic condition  
29 maintained by the bipolar membrane. The aniline was effectively degraded following first-  
30 order kinetics at a rate constant of  $0.0166 \text{ h}^{-1}$  under an applied voltage of 0.5 V. Meanwhile,  
31 a total organic carbon (TOC) removal efficiency of  $93.1 \pm 1.2\%$  was obtained, revealing  
32 efficient mineralization of aniline. The applicability of bipolar membrane MEC-Fenton  
33 system was successfully demonstrated with actual aniline wastewater. Moreover, energy  
34 balance showed that the system could be a promising technology for removal of  
35 biorefractory organic pollutants from wastewaters.

36 **Keywords:** Microbial electrolysis cell; Fenton reaction; Aniline; Industrial wastewater;  
37 Bipolar membrane;  $\text{H}_2\text{O}_2$

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## 42 1. Introduction

43 Aniline ( $C_6H_5NH_2$ ) has been widely used for various industries producing dyes, pesticides,  
44 rubber chemicals, and pharmaceuticals. Considering the biological accumulation, long term  
45 residue and carcinogenic properties, aniline-contained wastewater is categorized as  
46 hazardous waste (Li et al., 2016a; Wang et al., 2016). Biological methods have been widely  
47 used to treat aniline wastewater at low concentration ( $0-1000\text{ mg L}^{-1}$ ) (Jin et al., 2012; Liu et  
48 al., 2015), during which the aniline can be completely mineralized into  $CO_2$  and  $N_2/NO_x$   
49 (Wang et al., 2016). However, most conventional biological methods cannot treat high  
50 concentration ( $>2000\text{ mg L}^{-1}$ ) aniline wastewater due to the toxicity of aniline (Chen et al.,  
51 2007; Jin et al., 2012). In the past years, advanced oxidation processes especially the Electro-  
52 Fenton process have been recognized as attractive method for aniline degradation due to its  
53 high efficiency (Anotai et al., 2010; Brillas and Casado, 2002). However, there are still  
54 several shortcomings such as high cost electrode materials, high electrical energy  
55 consumption and required thoroughly pH control (at 2-3.5), which hinder industrial  
56 application (Brillas et al., 2009).

57 Recently, Bio-Electro-Fenton systems such as integrated microbial fuel cell-Fenton  
58 systems (MFC-Fenton) and Microbial Electrolysis Cell-Fenton systems (MEC-Fenton) have  
59 been demonstrated as promising alternative and cost-effective methods to traditional Electro-  
60 Fenton process for degradation of organic pollutants, such as azo dyes (Li et al., 2017b;  
61 Zhang et al., 2015), P-nitrophenol (Tao et al., 2013), Estrone (Xu et al., 2013), Bisphenol A,  
62 Sulfamethazine and Triclocarban (Wang et al., 2017). Though promising, there are still  
63 challenges which need to be addressed and validation is needed before commercial  
64 application. For instance, high mineralization efficiency has only been achieved with

65 synthetic wastewater and/or at low pollutant concentration (Asghar et al., 2014; Xu et al.,  
66 2015). Furthermore, most of the bio-Electro-Fenton systems use cation exchange membrane  
67 (CEM) as a separator, which has difficulty to maintain low catholyte pH and thus may cause  
68 inhibition on the Fenton process. The pH rise could also cause extensive iron precipitation  
69 which in return may damage the CEM and cathode (Ter Heijne et al., 2006). Therefore, a  
70 bio-Electro-Fenton system that can treat real and high concentration wastewater without  
71 causing pH issues is needed.

72 In this study, an innovative bio-Electro-Fenton system using bipolar membrane was  
73 developed to treat real industrial wastewater containing high concentration of aniline. The  
74 bipolar membrane has been shown to be an effective ion separator in previous MFC studies  
75 (Ter Heijne 2010), which could prevent pH elevation in the catholyte and pH drop in the  
76 anolyte (Ter Heijne et al., 2006; Ter Heijne et al., 2010; Zhang and Angelidaki, 2015). To  
77 the best of our knowledge, bipolar membrane has never been applied in bio-Electro-Fenton  
78 system. Furthermore, this is the first time that the MEC-Fenton system was applied for  
79 treatment of real industrial aniline wastewater. To optimize the conditions for the MEC-  
80 Fenton degradation of aniline, the effects of pH value, air flow rate and applied voltage on  
81 aniline degradation were investigated. This work offers an efficient and cost-effective  
82 approach for the removal of biorefractory organic pollutants from industrial wastewaters.

## 83 **2. Material and methods**

### 84 *2.1. Reactor setup*

85 The schematic diagram of the bipolar membrane MEC-Fenton system is shown in Fig. 1.  
86 The MEC consisted of two chambers which were separated by a bipolar membrane (BPM,  
87 fumasep® FBM, FuMA-Tech GmbH, Germany). The membrane was used to maintain low

88 cathode pH and avoid H<sup>+</sup> leakage to the anode (Zhang et al., 2015). The working volume of  
89 anode and cathode chamber was 100 mL (5 cm × 5 cm × 4 cm). The anode electrode was  
90 made of a carbon fiber brush (5.9 cm diameter, 6.9 cm length, Mill-Rose, USA), which was  
91 pretreated at 450 °C for 30 min and then pre-cultivated with mature biofilm in a MFC reactor  
92 before transferring to the MEC (Zhang et al., 2015). The cathode electrode was a graphite  
93 plate (3.5 cm × 4 cm). Cathode potential was measured versus a reference electrode  
94 (Ag/AgCl electrode, +197 mV vs SHE). Titanium wire was used to connect the cathode and  
95 anode electrode to the circuit.

96 **Fig. 1. is here**

## 97 *2.2. Characterization of domestic wastewater and aniline wastewater*

98 Domestic wastewater was collected from primary clarifier (Lyngby Wastewater Treatment  
99 Plant, Copenhagen, Denmark). The characteristics of the wastewater were as following:  
100 chemical oxygen demand (COD) of  $386 \pm 32$  mg L<sup>-1</sup>, pH 8.1, conductivity of 1.7 mS cm<sup>-1</sup>,  
101 stored at 4 °C before use. The aniline wastewater was provided by Vandrens A/S, Denmark  
102 and then stored at 4 °C before use. The characteristics of the aniline wastewater were:  
103 Aniline concentration of  $4460 \pm 52$  mg L<sup>-1</sup>, TOC of  $3360 \pm 80$  mg L<sup>-1</sup>, COD of  $10930 \pm 110$   
104 mg L<sup>-1</sup> and pH = 7.2. The aniline wastewater was amended with 50 mM Na<sub>2</sub>SO<sub>4</sub> and 10 mM  
105 FeSO<sub>4</sub> before each batch run.

## 106 *2.3. Reactor operation*

107 In this study, the research focused on the performance of aniline degradation in the cathode  
108 chamber. In order to avoid the influence from anode side, the anode chamber was  
109 continuously fed with domestic wastewater amended with sodium acetate (~1.6 g COD L<sup>-1</sup> in

110 total) at  $100 \text{ mL d}^{-1}$ . At the same time the domestic wastewater was recirculated from a feed  
111 reservoir (liquid volume of 300 mL) through anode at a recirculation rate of  $20 \text{ mL min}^{-1}$   
112 using a peristaltic pump (OLE DICH, Instrument makers APS, Denmark). The anode  
113 chamber and reservoir were purged with nitrogen gas before start new batch cycle. The  
114 cathode chamber was filled with 80 mL aniline wastewater and operated in batch mode.  
115 During the treatment process, fresh air was bubbled into the cathode providing oxygen at the  
116 rate of  $16 \text{ mL min}^{-1}$  except otherwise mentioned. All experiments were carried out in  
117 duplicate at ambient temperature ( $20 \pm 2 \text{ }^\circ\text{C}$ ). The cathode and anode were connected to a  
118 battery test system (Neware Battery Testing System TC53, Shenzhen, China), which was  
119 used as a power source (PS) to control the applied voltage and record the current of MEC (Li  
120 et al., 2014).

#### 121 2.4. Analytical methods

122 The samples were taken from the MEC cathode chamber, and then were filtered through  
123  $0.45 \text{ }\mu\text{m}$  filters. The  $\text{H}_2\text{O}_2$  concentration was measured by UV-vis spectrophotometry  
124 (spectronic 20D+, Thermo Scientific) at 400 nm, using potassium titanium (IV) oxalate as  
125 colored indicator (Sellers, 1980). The concentration of aniline was determined by high  
126 performance liquid chromatography (HPLC) (Wang et al., 2011). The pH was measured  
127 using a pH meter (PHM 210 pH meter, Radiometer). Whereafter adding 1 M NaOH solution  
128 in the samples to adjust the pH at 11 to stop the Fenton reaction. Chemical oxygen demand  
129 (COD) was measured according to the standard method (A.W.W.A., 1998). The total organic  
130 carbon (TOC) was measured by Shimadzu TOC 5000 A. Current density was calculated  
131 based on the surface area of cathode. Energy consumption was mainly due to the pumping  
132 system besides power supply. The energy consumption for pumping system was estimated



133 according to previous report (Zhang and Angelidaki, 2015). The calculations of degradation  
134 rate constant of aniline ( $k$ ), COD and TOC removal efficiencies are shown in the  
135 Supplementary data.

### 136 **3. Results and discussion**

#### 137 *3.1. Performance of aniline wastewater treatment in MEC-Fenton*

138 To evaluate the feasibility of this MEC-Fenton system for aniline wastewater treatment,  
139 aniline removal was conducted at 0.5 V, 16 mL min<sup>-1</sup> air flow rate, 10 mM Fe<sup>2+</sup> and initial  
140 pH 3. As shown in Fig. 2, aniline was rapidly degraded with removal efficiency of  $97.1 \pm 1.2\%$   
141 in 6 days, while the removal efficiency was only 8% for the system without Fe<sup>2+</sup> (Control 1)  
142 and 3% for the system without cathodic aeration (Control 2). The results imply that the  
143 bipolar membrane MEC-Fenton system was efficient for aniline degradation.

144 **Fig. 2. is here**

#### 145 *3.2. Effect of initial pH*

146 The electro-Fenton processes are generally performed at low pH to avoid the precipitation  
147 of ferric hydroxides. This requires pH adjustment before and after wastewater treatment. To  
148 study the effect of wastewater pH on the aniline removal, a group of experiments were  
149 conducted under various initial pH values (2, 3, 5 and 7.2) of aniline containing wastewater.  
150 The results are illustrated in Fig. 3. Firstly, experiments were performed without any pH  
151 adjustment at 7.2, which is the native pH value of aniline wastewater. The aniline removal  
152 efficiency just was 8% at this pH value. Comparatively, decrease of the initial pH value from  
153 7.2 to 3 led to a sharp increase in the degradation efficiency of aniline. When the pH was  
154 decreased to 2, the aniline degradation efficiency of  $97.1 \pm 1.2\%$  was obtained (Fig. 3a). The

155 differences observed here may result from the different efficiency of Fenton reaction at  
156 different initial pH values. The results demonstrated the MEC-Fenton system constructed  
157 with bipolar membrane can be used to treat high concentration aniline wastewater efficiently  
158 with initial pH 2-3.

159 The variation trend of catholyte pH is shown in Fig. 3b. It was observed that pH of aniline  
160 wastewater in the cathode chamber increased slowly to 5.6 from the initial value of 3 after 6  
161 days treatment. The pH increased to 10.7 from the initial values of 5 and 7.2 after 6 days. In  
162 order to investigate the effect of bipolar membrane on the cathodic pH, cation exchange  
163 membrane was used in a MEC as reference experiment, where the obvious removal of  
164 aniline was only observed for three days in MEC-Fenton with cation exchange membrane  
165 (Fig. S1. see Supplementary data). Furthermore, when using cation exchange membrane  
166 instead of bipolar membrane, ferric hydroxide was found in the cathode chamber after three  
167 days. The results demonstrated that the bipolar membrane could be used to help sustaining a  
168 lower catholyte pH without the need of extra acid dosage when the initial pH was 3. On the  
169 other hand, the formation of short-chain carboxylic acids during the mineralization of aniline  
170 such as maleic acid and oxalic acid (Anotai et al., 2006) could also contribute to the acidic  
171 pH. The anodic pH was maintained at 7.3-7.7 without significant changes. These results  
172 further demonstrated that the bipolar membrane is an effective separator in MEC-Fenton  
173 system.

174 **Fig. 3. is here**

### 175 *3.3. Effect of air flow rate*

176 The effect of air flow rate in the cathode on the degradation of aniline was investigated. It  
177 can be seen in Fig. 4, the optimum air flow rate observed was 16 mL min<sup>-1</sup>. It could be due

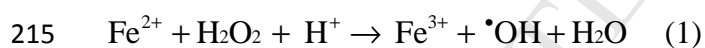
178 to that the increase of dissolved O<sub>2</sub> and mass transfer rate in the aniline wastewater improved  
179 the H<sub>2</sub>O<sub>2</sub> production, and thus promoted the Electro-Fenton process. The decrease of aniline  
180 decay rate at a higher air flow rate can be explained as following. There was a saturated state  
181 for dissolved O<sub>2</sub> in the MEC-Fenton system, thus the accumulations of H<sub>2</sub>O<sub>2</sub> hardly  
182 increased after dissolved O<sub>2</sub> was saturated ( $8.6 \pm 0.2 \text{ mg L}^{-1}$ ). In addition, the resistance of  
183 the aniline wastewater also increased with the excessive mass of O<sub>2</sub> bubble in the cathode  
184 chamber, which could lead the less negative cathode potential (Fig. S2). As a result, slightly  
185 drop in the removal efficiency of aniline was observed at the higher air flow rate. Similar  
186 phenomena were observed in the Electro-Fenton system (Zhou et al., 2013). The trend of  
187 COD and TOC removal efficiency in Fig. 4b was consistent with the evolution of aniline  
188 concentration. The mineralization rate at 4, 8, 16 and 50 mL min<sup>-1</sup> was  $43.5 \pm 2.3\%$ ,  $68.2 \pm$   
189  $1.8\%$ ,  $93.1 \pm 1.2\%$  and  $83.9 \pm 1.9\%$  after 6 days, respectively. Moreover, the air flow rates  
190 could also affect the energy consumption in terms of pumping. These results indicated that  
191 setting an optimum air flow rate in the MEC-Fenton system could not only improve the  
192 treatment efficiency of the aniline wastewater but also reduce treatment cost.

193 **Fig. 4. is here**

#### 194 *3.4. Effect of applied voltage*

195 Applied voltage is a critical parameter affecting the effectiveness of Electro-Fenton process  
196 as it controls the production of hydroxyl radicals. Therefore its influence on the degradation  
197 of aniline in the MEC-Fenton system was investigated under the optimal air flow rate of 16  
198 mL min<sup>-1</sup> and initial pH 3. As shown in Fig. 5, aniline removal efficiency was significantly  
199 enhanced when the applied voltage was increased from 0.3 to 0.5 V. However, further  
200 increase of applied voltage to 0.7 V led significantly in decrease of the aniline removal

201 efficiency, which was probably due to the relatively faster increase of pH in the cathode (Fig.  
202 S3). In addition, the current density increased from  $1.47 \pm 0.03$  to  $3.35 \pm 0.03$  A m<sup>-2</sup> with the  
203 increasing of applied voltage from 0.3 to 0.7 V (Fig. 5b). The cathode potential was  $-0.31 \pm$   
204  $0.01$ ,  $-0.45 \pm 0.01$  and  $-0.60 \pm 0.02$  V at 0.3, 0.5 and 0.7 V (Fig. 5c), respectively. The  
205 corresponding COD removal efficiencies are presented in Fig. 5d. Similar behavior of  
206 aniline removal efficiencies under different applied voltages were observed. The trend was  
207 different with Electro-Fenton processes for aniline wastewater treatment. It could be due to  
208 that the performance of Electro-Fenton for pollutants degradation was highly dependent on  
209 the H<sub>2</sub>O<sub>2</sub> production rate and hydroxyl radical ( $\bullet$ OH) generation from the reaction between  
210 Fe<sup>2+</sup> and H<sub>2</sub>O<sub>2</sub> (Eq. 1). The  $\bullet$ OH generation rate would increase with the increasing of H<sub>2</sub>O<sub>2</sub>  
211 production rate. According to our previous study (Li et al., 2017a), the optimal cathode  
212 potential of the graphite plate for H<sub>2</sub>O<sub>2</sub> production is ranging from -0.4 V to -0.5 V. Thus,  
213 the applied voltage of 0.5 V was the optimal for the aniline degradation in the bipolar  
214 membrane MEC-Fenton system.



216 Mineralization of organic pollutants with fast kinetics is highly desirable for  
217 contamination control. Here, the TOC removal efficiency was tested to evaluate the  
218 performance of MEC-Fenton for aniline mineralization (Fig. 5d). The TOC removal  
219 efficiency was  $66.8 \pm 3.1$ ,  $93.1 \pm 1.2$ ,  $51.2 \pm 1.9\%$  at 0.3, 0.5 and 0.7 V after 6 days,  
220 respectively. The higher mineralization rate of aniline at 0.5 V could be due to the faster  
221 H<sub>2</sub>O<sub>2</sub> production rate which is dependent mainly on the cathode electrode potential regulated  
222 by the external applied voltage. The results are similar with the trend of aniline removal  
223 efficiency. The removal rate constant of aniline degradation was 0.0097, 0.0166 and 0.0066

224  $\text{h}^{-1}$  at 0.3, 0.5 and 0.7 V, respectively (Fig. S4). These results imply that aniline can be  
225 efficiently mineralized by the MEC-Fenton technology at 0.5 V. This behavior can be  
226 ascribed to the greater production rate of  $\text{H}_2\text{O}_2$  at 0.5 V. The residual  $\text{H}_2\text{O}_2$  in the treated  
227 aniline wastewater at different applied voltage were also measured (Fig. S5). The residual  
228  $\text{H}_2\text{O}_2$  concentration after MEC-Fenton treatment was less than  $10 \text{ mg L}^{-1}$ . The results also  
229 demonstrated the feasibility of the bipolar membrane MEC-Fenton system for efficient  
230 control of residual  $\text{H}_2\text{O}_2$  level during aniline wastewater treatment.

231 **Fig. 5. is here**

### 232 *3.5. Energy efficiency for aniline wastewater treatment*

233 Energy consumption is one of the major concerns for wastewater treatment using Electro-  
234 Fenton technology, especially for recalcitrant pollutant degradation. In this bipolar  
235 membrane MEC-Fenton process, the optimal external voltage for aniline wastewater  
236 treatment was 0.5 V, which was much lower than that required for conventional Electro-  
237 Fenton process. The costs of the MEC-Fenton system mainly include the capital costs and  
238 the operating costs. The bipolar membrane MEC reactor capital costs are approx.  $5544 \text{ €m}^{-3}$   
239 (in Denmark) (Zhang and Angelidaki, 2016). The operating costs mainly include reagent  
240 costs and energy consumption of the external power supply. The MEC-Fenton system  
241 degrade aniline only required energy consumption of  $0.728 \text{ kWh kg}^{-1}$ -aniline from the  
242 external power over a fed batch cycle, which was much lower than classical Electro-Fenton  
243 process treat aniline with a cost of  $74 \text{ kWh kg}^{-1}$ -aniline (Brillas and Casado, 2002). The  
244 energy consumption for pumping would be  $0.374 \text{ kWh kg}^{-1}$ -aniline. Meanwhile our  
245 estimates were based on small laboratory-scale reactor and did not include reagent, e.g.,

246  $\text{Na}_2\text{SO}_4$ ,  $\text{FeSO}_4$ . Nevertheless the above results suggest that the bipolar membrane MEC-  
247 Fenton system was a cost-effective method for aniline wastewater treatment.

### 248 *3.6. Perspectives*

249 The results in this study demonstrated that the bipolar membrane MEC-Fenton system was  
250 environment-friendly, efficient and low cost compared to conventional Electro-Fenton  
251 system. In this process, the MEC besides treating domestic wastewater in the anode chamber  
252 (the COD removal efficiency reached  $80.5 \pm 2.2\%$  under 0.5 V), also mineralizes aniline  
253 from wastewater in the cathode chamber. It was proven that the operation of bipolar  
254 membrane MEC-Fenton greatly enhanced the treatment of aniline wastewater. Compared to  
255 other bio-Electro-Fenton system such as MFC-Fenton system, the bipolar membrane MEC-  
256 Fenton system has its own merits. Firstly, the degradation efficiency was greatly improved  
257 by adding low applied voltage (0.5 V) compared to MFC (Zhang et al., 2015). Secondly, the  
258 MEC-Fenton reactor with bipolar membrane requires lower dose of acid to adjust and  
259 control the pH of the aniline wastewater. Thirdly, the energy consumption was only 1.423  
260 kWh  $\text{kg}^{-1}$ -TOC under optimal operation condition, which was much lower than that in  
261 Electro-Fenton process (45.8 kWh  $\text{kg}^{-1}$ -TOC) (Gao et al., 2015). In addition, compared with  
262 other methods for aniline removal (see table 1), the MEC-Fenton system has relative high  
263 removal rate, especially higher than that of the biodegradation method. All these advantages  
264 together suggest that the MEC-Fenton system has potential for cost-effective and efficient  
265 degradation of recalcitrant organic pollutants. Finally, this system also can be extended to  
266 treat other industrial wastewater such as pharmaceuticals wastewaters. Though promising,  
267 more efforts should be made to accelerate the industrial application, such as development of  
268 large scale system with continues-flow operation. Future work also should focus on the

269 development of low cost cathode electrode with large surface such as three dimensional  
270 electrode, which may improve the H<sub>2</sub>O<sub>2</sub> production rate and further enhance the aniline  
271 removal rate.

#### 272 **4. Conclusions**

273 This study demonstrated that the MEC-Fenton system is an effective and environmentally  
274 friendly technology for aniline containing wastewater treatment. In such system, high  
275 concentration ( $4460 \pm 52 \text{ mg L}^{-1}$ ) aniline was not only effectively degraded with removal  
276 rate of  $30.1 \pm 0.4 \text{ mg L}^{-1} \text{ h}^{-1}$ , but also highly mineralized with TOC removal efficiency of  
277  $93.1 \pm 1.2\%$  and  $k$  of  $0.0166 \text{ h}^{-1}$  at initial pH 3. Notably the energy consumption was only  
278  $1.423 \text{ kWh kg}^{-1}\text{-TOC}$ . This work provides a cost-effective method for aniline degradation,  
279 which is also attractive and applicable for efficient treatment of industrial wastewater.

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Table 1. Performance of aniline removal using different technologies.

Method	Concentration (mg L <sup>-1</sup> )	Removal efficiency	Removal rate (mg L <sup>-1</sup> h <sup>-1</sup> )	Energy consumption kWh kg <sup>-1</sup> -aniline	Reference
Fenton	930	85.9%	798.9	-	(Anotai et al., 2006)
Electro-Fenton	1000	63%	315	74	(Brillas and Casado, 2002)
Biodegradation	300	87%	2.175	-	(Jin et al., 2012)
Fluidized-bed Fenton	930	97%	1804.2	-	(Anotai et al., 2010)
MFC-biodegradation	260.4±9.3	91.2±2.2%	1.65±0.04	-	(Cheng et al., 2015)
Electrocatalytic	3500	97.7%	683.9	36.2	(Li et al., 2016b)
Electrodialysis	1000	100%	6792.4	2.86	(Wang et al., 2016)
MEC-Fenton	4460±52	97.1±1.2%	30.1±0.4	1.10	This study

390 -: no report the energy consumption.

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410 **Figure Captions**

411 **Fig. 1.** Schematic illustration of the MEC-Fenton reactor with bipolar membrane (BPM).

412 **Fig. 2.** The performance of bipolar membrane MEC-Fenton system on the aniline  
413 degradation. Conditions:  $E = 0.5$  V, initial pH = 3 and air flow rate of  $16 \text{ mL min}^{-1}$ . (Control  
414 1: without  $\text{Fe}^{2+}$ ; Control 2: without cathodic aeration)

415 **Fig. 3.** The effect of initial pH on the performance of bipolar membrane MEC-Fenton  
416 system. Conditions:  $E = 0.5$  V, air flow rate of  $16 \text{ mL min}^{-1}$ .

417 **Fig. 4.** The effect of air flow rate on the performance of bipolar membrane MEC-Fenton  
418 system. Conditions:  $E = 0.5$  V, initial pH = 3.

419 **Fig. 5.** The effect of applied voltage on the bipolar membrane MEC-Fenton degradation of  
420 aniline.

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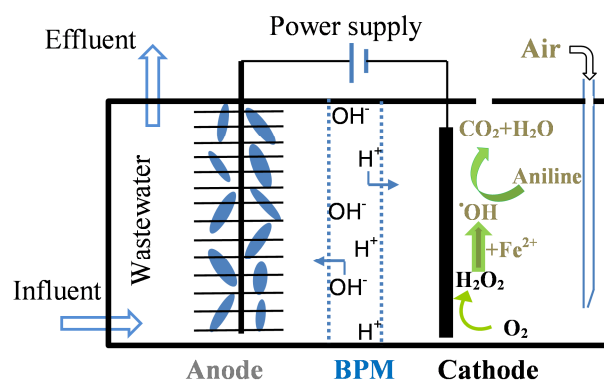
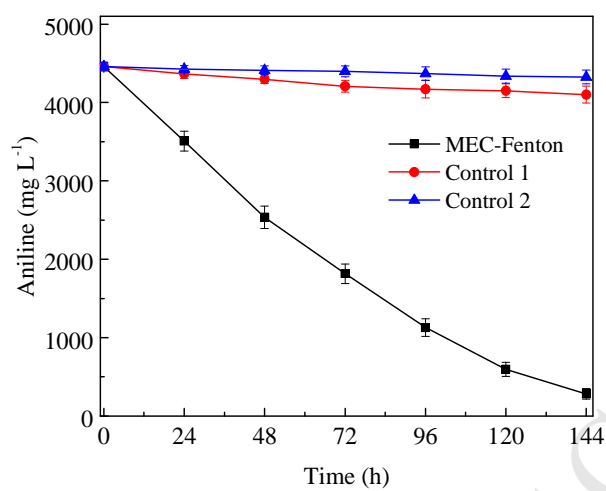


Fig. 1.

**Fig. 2.**

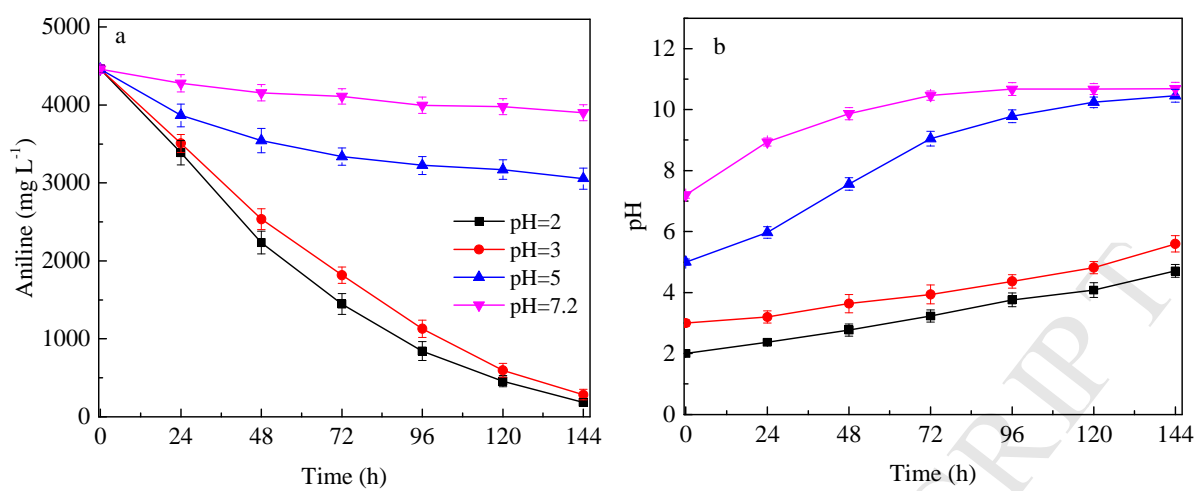


Fig. 3.

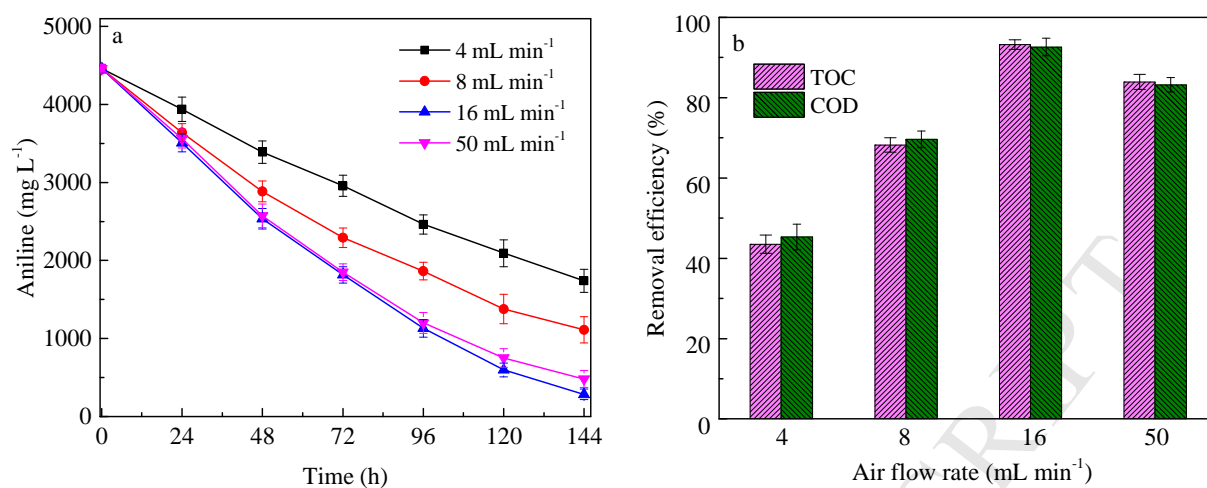


Fig. 4.



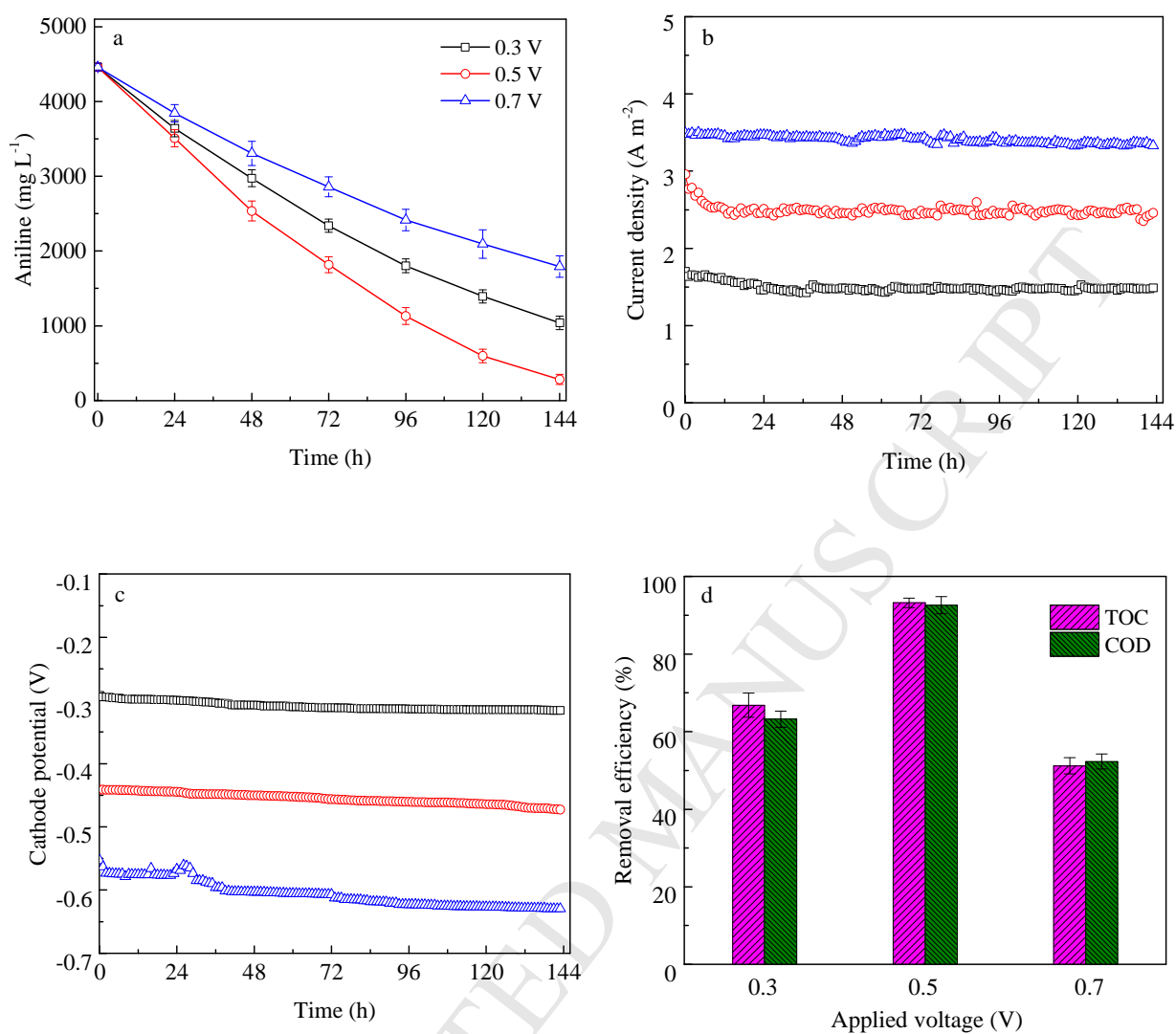


Fig. 5.

**Highlights**

- Novel MEC-Fenton process for the treatment of real aniline-contained wastewater.
- The bipolar membrane was an effective pH separator in MEC-Fenton process.
- High removal efficiency was achieved at relatively higher aniline concentration.
- Identified key factors affecting the aniline degradation in MEC-Fenton system.
- Efficient removal of aniline with low energy consumption in MEC-Fenton system.

ACCEPTED MANUSCRIPT