

1 **Synergy of flocculation and flotation for microalgae harvesting using aluminium**
2 **electrolysis**

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14

15 **Abstract**

16 Microalgae are often used as feedstock for renewable biofuel production and as
17 pollutant up-takers for wastewater treatment; however, biomass harvesting still
18 remains a challenge in field applications. In this study, electro-flocculation using
19 aluminium electrolysis was tested as a method to collect *Chlorella vulgaris*. The
20 electrolysis products were positively charged over a wide pH range below 9.5, which
21 gave them a flocculation potential for negatively charged microalgae. As flocculants
22 were in-situ generated and gradually released, microalgae flocs formed in a
23 snowballing mode, resulting in the compaction of large flocs. When higher current
24 density was applied, microalgae could be harvested more rapidly, although there was
25 a trade-off between a higher energy use and more residual aluminium in the culture
26 medium. Benefits of this flocculation method are two-fold: the phosphate decrease in
27 post-harvesting could improve nutrient removal in microalgae based wastewater
28 treatment, while the ammonium increase may favor microalgae recovery for medium
29 recycling.

30 **Keywords:** Microalgae harvesting; Electro-flocculation; Current density; Energy
31 consumption; Phosphate.

32 **1. Introduction**

33 In recent years, the use of microalgae has attracted great interest as a means to produce
34 biofuels and treat wastewater (Baeyens et al., 2015; Kang et al., 2010; Sulzacova et al.,
35 2015). The biofuel yield from microalgae was estimated to be 10 ~ 20 times higher than
36 those from oleaginous seeds and vegetable oils (Chisti, 2007). In microalgae based
37 wastewater treatment, pollutants can be ecologically and safely removed through
38 microalgae assimilation, with the added benefit of biofuel production (Mehrabadi et al.,
39 2016; Tan et al., 2016). However, microalgae harvesting still remains a challenge due to
40 the small cell size, electrical stability and low density in growth media (Cerff et al.,
41 2012). The cost of microalgae harvesting can represent about 60% of the total cost of
42 the final products (Grima et al., 2003).

43 Several methods have been tested to harvest microalgae, including gravity
44 sedimentation (Depraetere et al., 2015), centrifugation (Chen et al., 2015), filtration
45 (Nurra et al., 2014) and chemical flocculation (Reyes and Labra, 2016). Gravity settling
46 is simple but only suitable to harvest microalgae with large size (Park and Craggs,
47 2010). Centrifugation and filtration are rapid and reliable, but require high energy input
48 and large capital investment, making the large-scale implementation economically
49 unfeasible (Kim et al., 2015). Chemical flocculation requires minimal equipment to
50 effectively harvest microalgae; however, the addition of chemical flocculants inevitably
51 introduces large amounts of other undesired anions such as sulfates and chlorides, and
52 thereby leads to operation cost increase and potential negative impacts (Pan et al., 2011).

53 So far, there are few cost-effective and efficient technologies for microalgae harvesting,
54 which limits large-scale applications of microalgae in biofuel production and
55 wastewater treatment.

56 Electro-flocculation is an electrochemical technique for pollutant removal, which is
57 based on the in-situ generation of flocculants during metal electrolysis (Vasudevan et al.,
58 2008). Owing to the advantages of low cost, high efficiency and easy operation,
59 electro-flocculation has been widely applied in wastewater treatment to remove
60 phosphorus (Mores et al., 2016), dyes (Mollah et al., 2010), fluoride (Hu et al., 2005),
61 organic matter (Asselin et al., 2008) and heavy metals (Hanay and Hasar, 2011). Charge
62 neutralization is identified as the main mechanism of electro-flocculation, which creates
63 the sorption affinity for negatively charged pollutants (Vasudevan et al., 2008).
64 Electro-flocculation may act as a potential solution for microalgae harvesting, due to the
65 net negative surface charges on the cells. Dassey and Theegala (2014) observed the
66 limited efficacy of electro-flocculation on the harvesting of *Dunaliella* sp. and
67 *Nannochloris* sp. Xiong et al. (2015) tested the synergy of electro-flocculation and sand
68 particles on the removal of *Dunaliella salina*. In spite of the recent advances,
69 knowledge gaps still exist with respect to the technique's efficacy, especially the
70 mechanisms responsible for flocculation remain poorly understood.

71 This study explored aluminium (Al) based electro-flocculation to harvest microalgae.
72 The electrolysis products were characterized, and the relationship among harvesting
73 efficiency, surface charge, floc size and floc structure were investigated to reveal the

74 mechanisms. The energy input, Al consumption and culture medium responses were
75 studied for field applications. After microalgae harvesting, the residual Al in the culture
76 medium was also assessed with respect to potential risk.

77 **2. Experimental section**

78 *2.1 Microalgae species and culture*

79 Freshwater *Chlorella vulgaris* (*C. vulgaris*), a commonly used species in biofuel
80 production and microalgae based wastewater treatment (Arbib et al., 2014; de-Bashan
81 et al., 2004), was used in this study. The *C. vulgaris* cells (FACHB-24) were obtained
82 from the Institute of Hydrobiology, Chinese Academy of Sciences, and cultured in
83 BG11 medium according to the instructions. The BG11 medium was composed of 500
84 mg L⁻¹ Bicin, 100 mg L⁻¹ KNO₃, 100 mg L⁻¹ b-C₃H₇O₆PNa₂, 50 mg L⁻¹ NaNO₃, 50 mg
85 L⁻¹ Ca(NO₃)₂•4H₂O, 50 mg L⁻¹ MgCl₂•6H₂O, 40 mg L⁻¹ Na₂SO₄, 20 mg L⁻¹ H₃BO₃, 5
86 mg L⁻¹ Na₂EDTA, 5 mg L⁻¹ MnCl₂•4H₂O, 5 mg L⁻¹ CoCl₂•6H₂O and 0.8 mg L⁻¹
87 Na₂MoO₄•2H₂O, 0.5 mg L⁻¹ FeCl₃•6H₂O and 0.5 mg L⁻¹ ZnCl₂. Microalgae batch
88 cultures (10 L) were maintained at 30 ± 1°C under continuous cool white fluorescent
89 light of 2000 ~ 3000 lux on a 12 h light and 12 h darkness regimen in an illuminating
90 incubator (LRH-250-G, Guangdong Medical Apparatus Co., Ltd., China). The culture
91 was continuously aerated with air at a flow rate of 5 L min⁻¹ using a pump (AC0-001,
92 Sensen Group Co., Ltd., China), and microalgae growth was monitored by counting
93 the cell numbers. The dry cell weight was measured by filtering an aliquot of the
94 culture suspension through pre-weighed GF/C filters (Whatman, England). After

95 rinsed with deionized water, the filters were dried at 105°C for 24 h and re-weighed.

96 2.2 *Electro-flocculation system*

97 The electro-flocculation unit consisted of two Al electrode plates (Jinjia Metal Co.,
98 Ltd., China) and a flat stir paddle (Zhongrun Water Industry Technology Development
99 Co., Ltd., China) for mixing in a 500-ml beaker. The Al electrode plates had a surface
100 area of 3×10 cm and a thickness of 1 cm, and were vertically installed with a gap of 3
101 cm. During electro-flocculation, the electrode plates were partially immersed in the
102 microalgae solution, such that the effective surface area was 22.5 cm². The electric
103 current was supplied by a direct current power supply (DF1730SL5A, Ningbo Zhongce
104 Dftek Electronics Co., Ltd., China). The experimental set-up was schematically
105 presented in Fig. S1 in the supporting information (SI).

106 2.3 *Microalgae electro-flocculation*

107 The exponential growth phase of *C. vulgaris* culture was used in the
108 electro-flocculation experiment. The initial cell concentration was set to 3.63×10^{10}
109 cells L⁻¹. 0.4 L of readily prepared *C. vulgaris* solution was transferred to the
110 electro-flocculation cell, and then stirred at 200 rpm after electric current was supplied.
111 The control was run in the above-mentioned *C. vulgaris* solution, but without electric
112 current. Prior to each run, the electrodes were immersed in 5% HNO₃ solution, and
113 lightly wiped with abrasive paper, and then rinsed with deionized water to remove
114 barrier oxide film on the electrode surface. The flocculation experiments were
115 conducted at raw microalgae solution pH of 8.6. All the flocculation experiments were

116 conducted in triplicates.

117 *2.4 Analytical methods*

118 After 10 min of microalgae electro-flocculation, samples were collected from 5 cm
119 above the bottom to enumerate the cell number using an Axioskop 2 mot plus
120 microscope (Carl ZEISS, Germany). The microalgae harvesting efficiency was
121 calculated as:

$$122 \quad \text{Harvesting efficiency} = (IC-SC)/IC \times 100\% \quad (1)$$

123 where *IC* and *SC* are the initial and sample cell concentration, respectively.

124 The surface charge of microalgae cells was characterized using a Zetasizer 2000
125 (Malvern Co. United Kingdom). Dynamic size growth of microalgae flocs during
126 electro-flocculation was analyzed using a laser particle size analyzer (Mastersizer 2000,
127 Malvern Co., United Kingdom). The apparatus set-up was described in Fig. S2 in the SI,
128 and the size was denoted by the measured mean diameter ($d_{0.5}$). For the floc image study,
129 the flocs were carefully transferred onto a glass slide and then photographed by an
130 electromotive microscope (ST-CV320, Chongqing UOP Photoelectric Technology Co.,
131 Ltd., China). After microalgae harvesting, phosphate and ammonium in the culture
132 medium were measured according to the Monitoring Analysis Method of Water and
133 Wastewater (Ministry of Environmental Protection of China, 2002). The medium pH
134 and temperature were measured using a Yellow Springs Instruments (Yellow Springs,
135 Ohio, USA). The energy consumption was calculated as:

$$136 \quad \text{Energy consumption (kWh L}^{-1}\text{)} = UIt/v \quad (2)$$

137 Energy consumption (kWh g^{-1} microalgae) = $UIt/v\beta\theta\sigma$ (3)

138 where U is cell voltage (V), I is current intensity (A), t is electrolysis time (s), and v is
139 the volume of microalgae solution (L), β is the initial microalgae concentration, θ is the
140 microalgae harvesting efficiency (%), and σ is the microalgae weight (32×10^{-12} g
141 cell^{-1}).

142 The Al consumption and charge loading were calculated using the Eq. (4) and Eq. (5)
143 according to Faraday's law (Zaied and Bellakhal, 2009),

144 Al consumption = ItM/zFv (4)

145 Charge loading = It/Fv (5)

146 where M is the molecular mass of Al (26.98 g mol^{-1}); z is the number of electrons
147 transferred ($z = 3$); F is Faraday's constant (96487 C mol^{-1}). After electro-flocculation,
148 the residual Al in the medium was analyzed using an Inductively Coupled Plasma
149 Optical Emission Spectrometer (Optima 8300, PerkinElmer, USA).

150 3. Results

151 3.1 Surface charge of Al electrolysis products

152 During Al electrolysis, amorphous-like products were observed. Analysis on surface
153 charge indicated that the products were positively charged. At the current density of
154 $22.2, 44.4$ and 66.7 A m^{-2} , the zeta potential of Al electrolysis products (AEP) ranged
155 between $+6.5$ and $+15.2 \text{ mV}$ within the electrolysis time of 8 min (Fig. 1a). The surface
156 charge of AEP maintained positive in a wide pH range below 9.5, and reached the
157 highest value of $+27.2 \text{ mV}$ under near-neutral pH conditions. In contrast, the zeta

158 potential of *C. vulgaris* cells gradually decreased from -0.2 to -21.8 mV in the pH range
159 of 1.8 ~ 10.5 (Fig. 1b).

160 *3.2 Microalgae floc formation*

161 After Al electrolysis was initiated, microalgae aggregation occurred, thus flocs became
162 larger and more compact along time. At the current density of 44.4 A m^{-2} , the floc size
163 ranged between 2.5 and $316.2 \text{ }\mu\text{m}$ with the mean diameter ($d_{0.5}$) of $99.3 \text{ }\mu\text{m}$ at the
164 electrolysis time of 2 min, and ranged between 70.8 and $562.3 \text{ }\mu\text{m}$ with the mean
165 diameter of $262.3 \text{ }\mu\text{m}$ at 4 min, and ranged between 89.1 and $794.3 \mu\text{m}$ with the mean
166 diameter of $298.1 \text{ }\mu\text{m}$ at 6 min, and ranged between 125.9 and $891.3 \mu\text{m}$ with the mean
167 diameter of $367.6 \text{ }\mu\text{m}$ at 8 min (Fig. 2a). The floc fractal dimension was 1.29, 1.71, 1.96
168 and 2.01 at the electrolysis time of 2, 4, 6, 8 min, respectively (Fig. 2b). Large amounts
169 of tiny gas bubbles were observed on microalgae flocs (Fig. S3 in the SI.). These
170 bubbles carried the flocs to water surface and then broke up.

171 *3.3 Effect of current density on microalgae harvesting*

172 Using Al electrolysis, a maximum microalgae harvesting efficiency of about 98% was
173 achieved, although different electrolysis time was needed, depending on the current
174 density applied. In general, the higher current density, the shorter electrolysis time is
175 needed to reach the maximum microalgae harvesting. When 22.2, 44.4 and 66.7 A m^{-2}
176 was applied, it took 7, 6 and 4 min to achieve the maximum microalgae harvesting,
177 respectively (Fig. 3a). However, the charge loading holds a similar shape at different
178 current densities. To remove 98% of microalgae cells, the charge loading was about

179 0.75 Faradays m^{-3} (Fig. 3b). The surface charge of microalgae cells as a function of
180 electrolysis time was also investigated during microalgae harvesting. As the
181 electrolysis time increased, an increase was obtained in the cell surface charge, which
182 was enhanced by the higher current density. When 22.2, 44.4 and 66.7 A m^{-2} was
183 applied, the zeta potential of microalgae cells was gradually increased from -14.0 mV
184 to -12.7, -6.2 and -3.9 mV at the electrolysis time of 8 min, respectively (Fig. 3b).

185 *3.4 Energy consumption*

186 When higher current density was applied, more energy consumption was needed to
187 achieve the same microalgae harvesting rate. At the current density of 22.2, 44.4 and
188 66.7 A m^{-2} , the energy consumption was 0.99×10^{-4} , 2.53×10^{-4} and 3.35×10^{-4} kWh
189 L^{-1} , respectively (Fig. 4a). Energy consumption per gram microalgae biomass was
190 calculated and presented in Fig. 4b. It indicated that the energy consumption was the
191 highest at the low microalgae harvesting efficiency. As the harvesting efficiency
192 increased, the energy consumption decreased and kept stable at the harvesting
193 efficiency of $> 80\%$. However, the use of lower charge density generally yielded lower
194 energy consumption per gram biomass for effective microalgae harvesting ($> 80\%$).
195 The energy consumption was 0.87×10^{-4} , 2.22×10^{-4} and 2.94×10^{-4} kWh g^{-1} biomass
196 at the current density of 22.2, 44.4 and 66.7 A m^{-2} , respectively.

197 *3.5 Al consumption and charge loading*

198 Al consumption is calculated and plotted against microalgae harvesting efficiency in
199 Fig. 5a. The data sets take on a similar shape at different current densities. To harvest

200 98% of *C. vulgaris*, 7.23 mg L⁻¹ of Al was consumed from the culture medium.
201 However, the residual Al in the culture medium varied with the current density. The
202 use of higher current density led to higher residual Al. When 22.2, 44.4 and 66.7 A m⁻²
203 was applied, the residual Al was 1.6, 4.2 and 4.9 mg L⁻¹ at the harvesting efficiency of
204 98% (Fig. 5b).

205 *3.6 Microalgae culture medium responses*

206 After microalgae harvesting, there were no significant changes in the medium
207 temperature and pH. When 44.4 A m⁻² was applied, the temperature and pH kept stable
208 throughout the experiments at 21.8°C and 8.6, respectively (Fig. 6a). However,
209 electro-flocculation did lead to chemical changes in the culture medium. Phosphate
210 decrease and ammonium increase were observed during microalgae harvesting. At the
211 current density of 44.4 A m⁻², the phosphate decreased from 3.9 to 3.7 mg L⁻¹ within
212 the initial 1 min, and quickly decreased to 1.8 mg L⁻¹ at 4 min, and then slowly
213 decreased to 0.6 mg L⁻¹ at 8 min; while the ammonium gradually increased from 0.34
214 to 1.22 mg L⁻¹ within the 8 min of electrolysis (Fig. 6b).

215 **4. Discussion**

216 *4.1 Charge neutralization, bridging and bubble flotation*

217 Charge neutralization is an essential step in microalgae flocculation, which decreases
218 energy barrier for microalgae aggregation (Hjorth and Jorgensen, 2012). The AEPs
219 were positively charged over a wide pH range below 9.5, which gave them the
220 flocculation potential for negatively charged microalgae cells (Fig. 1b). With the

221 neutralization, the surface charge of microalgae cells was gradually increased,
222 indicating that positive charge plays a key role in microalgae harvesting using
223 electro-flocculation. It is further supported by the fact that microalgae harvesting
224 efficiency as a function of charge loading holds a similar shape at different current
225 densities (Fig. 3b). However, the higher current density could shorten the electrolysis
226 time of microalgae harvesting (Fig. 3a), due to the higher rate of charge loading (Fig. S4
227 in the SI).

228 With the operation of charge neutralization mechanism alone, the optimum
229 flocculation often occurs at the point of total charge neutralization (Shi et al., 2016).
230 However, in this study, the zeta potential of microalgae cells was negative at the
231 optimum microalgae harvesting (Fig. 3c), which indicated that the optimum flocculation
232 was already achieved before the cell surface charge was totally neutralized. The
233 operation of a potential “bridging mechanism” may favor microalgae flocculation.
234 During Al electrolysis, the generated Al^{3+} and OH^- react spontaneously to produce
235 various monomeric species such as $\text{Al}(\text{OH})^{2+}$, $\text{Al}(\text{OH})_2^+$, $\text{Al}_2(\text{OH})_2^{4+}$, $\text{Al}(\text{OH})_4^-$, and
236 polymeric species such as $\text{Al}_6(\text{OH})_{15}^{3+}$, $\text{Al}_7(\text{OH})_{17}^{4+}$, $\text{Al}_8(\text{OH})_{20}^{4+}$, $\text{Al}_{13}(\text{OH})_{34}^{5+}$ (Ghosh
237 et al., 2008). These freshly amorphous AEPs (Fig. S5 in the SI) have the potential to
238 trap small microalgae flocs and bridge them into large ones (Fig. 2a). Then, H_2 bubbles
239 generated at the cathode entrap into these microalgae flocs (Fig. S3 in the SI), causing
240 them to float to the water surface where they can be easily collected. This “charge
241 neutralization-bridging-flotation” mechanism is illustrated in Fig. S6 in the SI.

242 The floc structure has great influence on flocculation kinetics (Shi et al., 2016; Wyatt
243 et al., 2013). The compact flocs are resistant to breakage and beneficial to the
244 solid-liquid separation. Previous studies reported that large flocs are often fragile (Gibbs,
245 1982); however, in this study, microalgae flocs became not only larger but also denser
246 (Fig. 2a and 3b) as the electrolysis time increased, which may be attributed to the
247 snowballing-mode floc formation. During electro-flocculation, flocculants were in-situ
248 generated and gradually released to form flocs. This layer-by-layer assembly could
249 cause the flocs to become progressively more compact with the continuous addition of
250 flocculants.

251 *4.2 Energy and Al consumption*

252 Economic cost is often a major concern for the practical application of a method,
253 largely driven by energy and material costs (Dassey and Theegala, 2014). In this study,
254 the use of higher current density resulted in quicker microalgae harvesting (Fig. 3a).
255 However, the application of higher current density in an attempt to speed up microalgae
256 harvesting may not be economically efficient, due to the greater energy consumption. To
257 harvest 98% of *C. vulgaris*, the energy consumption at 66.7 A m⁻² was approximately
258 1.32 and 3.38 times higher than those at 44.4 and 22.2 A m⁻², respectively (Fig. 4),
259 which may be attributed to the production of more waste heat at the higher current
260 density (Kobyta and Delipinar, 2008). During electro-flocculation, energy consumption
261 per microalgae biomass exhibited a decreasing trend. It was the most energy-efficient at
262 the harvesting efficiency of > 80% (Fig. 4b). Thus, it is not necessary to collect all the

263 biomass in some fields, such as microalgae based wastewater treatment. The remaining
264 cells may benefit microalgae recovery, possibly aiding further treatment of wastewater.
265 Previous studies demonstrated that electrode distribution and water conductivity may
266 have great influence on energy consumption (Chen, 2004). It was concluded that energy
267 consumption could be minimized by using high conductivity electrolytes (i.e. high salt
268 content) with narrow electrode spacing in a low electric current (Emamjomeh and
269 Sivakumar, 2009). Further studies are needed to optimize the energy efficiency of
270 microalgae harvesting.

271 Charge loading was identified as the key factor of microalgae electro-flocculation
272 (Fig. 3b), leading to the similar Al consumption at different charge densities (Fig. 5a).
273 This is because that the amount of electrochemically dissolved Al is proportional to
274 charge loading according to Faraday's law (Zuo et al., 2008). However, the residual Al
275 in the culture medium varied with the current density. The use of high charge density
276 led to high residual Al in the culture medium (Fig. 5b), which may cause negative
277 impacts due to its potentially toxic nature (Sinha and Mathur, 2016).

278 *4.3 Water quality changes*

279 In the electrolysis process, water pH and temperature are often increased because of the
280 hydroxyl formation and waste heat production (Harif and Adin, 2007). However, due to
281 the low electric power input in this study, there were no significant changes in water pH
282 and temperature in the culture medium after microalgae harvesting (Fig. 6a). Hence, it is
283 possible to balance microalgae harvesting and maintaining acceptable levels of water

284 quality by carefully operating electrolysis, which makes the method sustainable. In the
285 microalgae biofuel industry, medium reuse offers a promising strategy for saving water
286 and nutrients (Castrillo et al., 2013; González-López et al., 2013).

287 In addition to biofuel production, microalgae are also widely used in wastewater
288 treatment (Sulzacova et al., 2015; Tan et al., 2016). In microalgae based wastewater
289 treatment, phosphorus and nitrogen are assimilated by microalgae as nutrients for
290 growth, and are subsequently removed through biomass harvesting (Tan et al., 2016).
291 Following microalgae collection using electro-flocculation in this study, residual
292 phosphate in the medium was significantly decreased (Fig. 6b), which potentially
293 enhanced nutrient removal in wastewater treatment. Ammonium as a nitrogen source is
294 generally favored by microalgae (Kim et al., 2013); as seen in this study, a
295 post-harvesting increase in ammonium may benefit microalgae recovery for future
296 medium recycling. During electrolysis, nitrate reduction ($\text{NO}_3^- + 10 \text{H}^+ + 8 \text{e}^- = \text{NH}_4^+ +$
297 $3\text{H}_2\text{O}$) can occur at the cathode, which potentially contributes to the ammonium
298 increase in the culture medium (Peel et al., 2003).

299 *4.4 Recommendations for future applications*

300 Microalgae harvesting is a crucial step but still remains a challenge for biomass
301 engineering or environmental applications. In this study, electro-flocculation proved to
302 be a rapid and efficient way to harvest microalgae. The in-situ generation of
303 flocculants can be easily controlled by an electrical switch, which offers the prospect
304 of applications in continuous systems (Fig. S7 in the SI). Many studies have conducted

305 the life cycle assessment (LCA) of biofuel production from microalgae and confirmed
306 the potential of microalgae as an energy source (Lardon et al., 2009; Yang et al., 2011).
307 In this study, the cost of microalgae harvesting using Al electrolysis was estimated to
308 be 1.47×10^{-3} US\$ g⁻¹ biomass, most of which was born on the material use (Table S1).
309 Further studies are needed to optimize operation conditions to increase the electrode
310 utilization efficiency.

311 Despite the fact that Al electrolysis is an effective microalgae harvesting technique
312 for most engineering applications, it is not recommended for cases where the biomass
313 is to be used for food or animal feed. The excess Al could enter the food chain and
314 induce bond and brain diseases in human beings (Douichene et al., 2016). The synergy
315 of edible macromolecular flocculants (flocculation) and insert electrodes (flotation)
316 may provide a promising strategy to harvesting microalgae for food use.

317 **5. Conclusions**

318 The use of Al electrolysis allowed feasible microalgae harvesting (~ 98%) with the
319 operation of charge neutralization, bridging and bubble flotation mechanisms.
320 Microalgae floc formation followed a snowballing mode, with the flocs becoming larger
321 and more compact through time. When the higher current density of 66.7 A m⁻² was
322 applied, microalgae harvesting was achieved in a shorter time of 4 min, but at the cost
323 of higher energy consumption of 3.35×10^{-4} kWh L⁻¹ and more residual Al of 4.9 mg
324 L⁻¹. Using electro-flocculation, the phosphate removal can be a side benefit for
325 microalgae based wastewater treatment.

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460

461 **Figure Captions**

462 **Fig.1.** The surface charge properties of AEP. (a) Effect of electrolysis time; (b) Effect of
463 pH. Error bars indicate standard deviations.

464 **Fig. 2.** The microalgae floc formation during electro-flocculation. (a) The floc size
465 distribution at different electrolysis time; (b) The floc fractal dimension at different
466 electrolysis time. The current density was set to 44.4 A m^{-2} . Error bars indicate
467 standard deviations.

468 **Fig. 3.** The microalgae harvesting efficiency (a), charge loading (b) and cell surface
469 charge (c) at different current densities. Error bars indicate standard deviations.

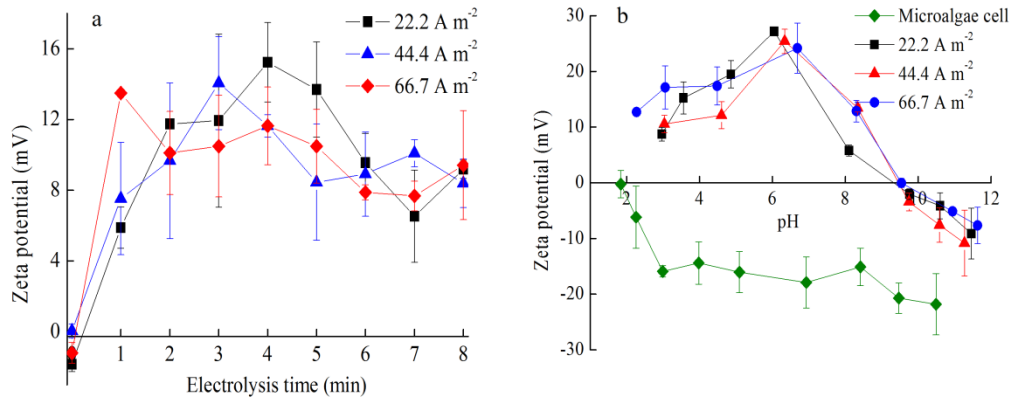
470 **Fig. 4.** The energy consumption during microalgae harvesting using
471 electro-flocculation. (a) Energy consumption per liter; (b) Energy consumption per
472 gram microalgae biomass. Error bars indicate standard deviations.

473 **Fig. 5.** The Al consumption (a) and residual Al (b) at different current densities. Error
474 bars indicate standard deviations.

475 **Fig. 6.** The responses of microalgae culture medium to electro-flocculation using Al
476 electrodes. (a) Temperature and pH, (b) Phosphate and ammonium. The current density
477 was set to 44.4 A m^{-2} . Error bars indicate standard deviations.

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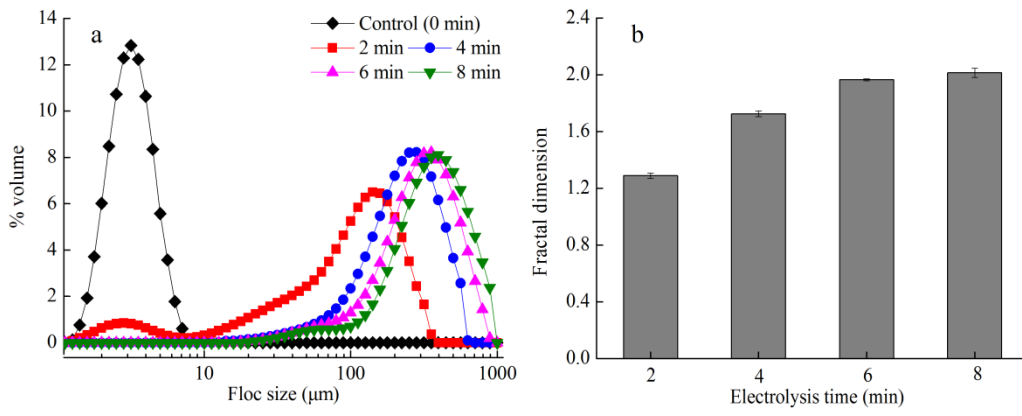
479 **Fig.1**



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482 **Fig. 2**



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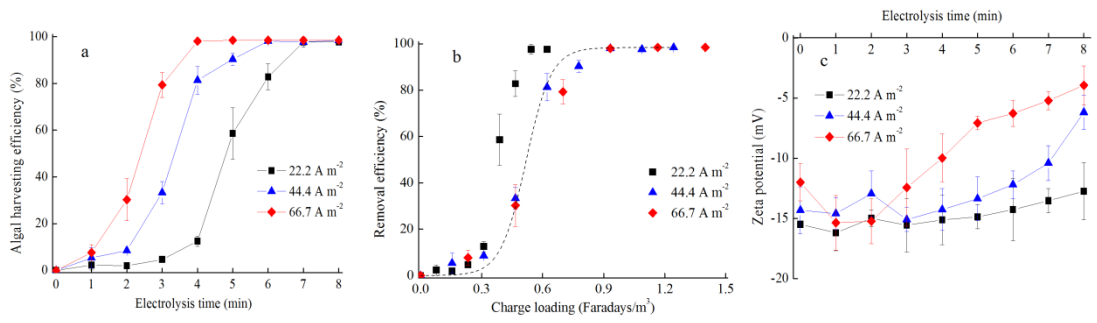
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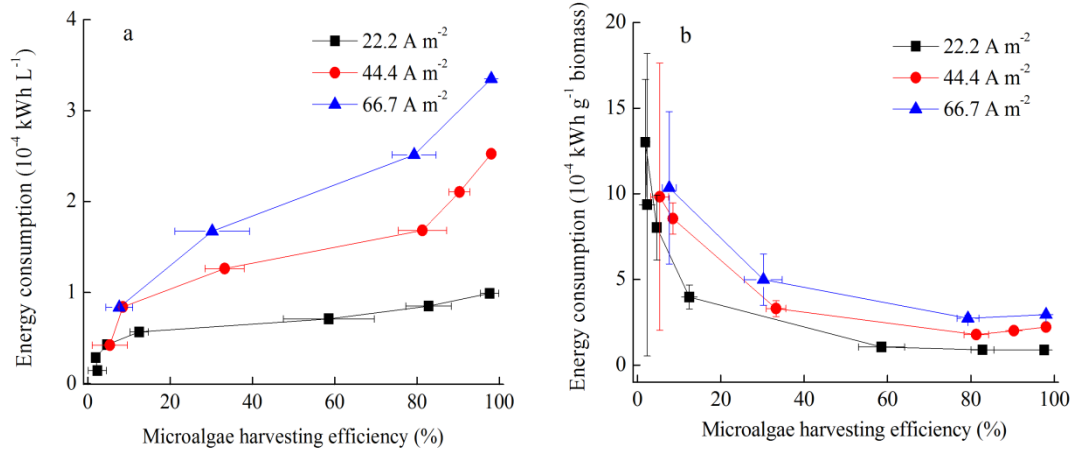
499 **Fig. 3**



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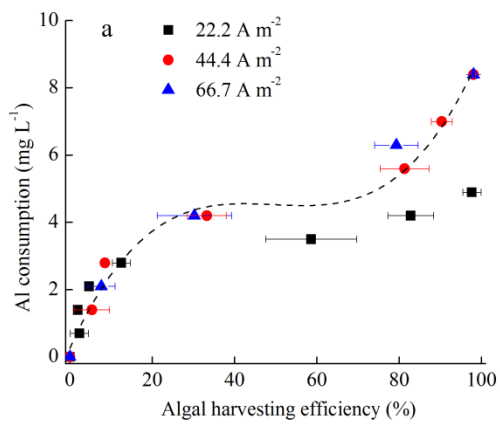
502 **Fig. 4**



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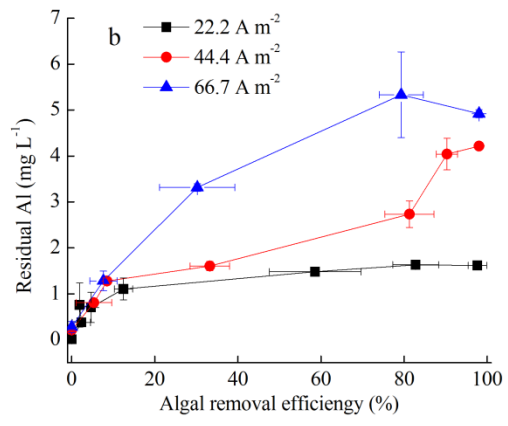
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505 **Fig. 5**

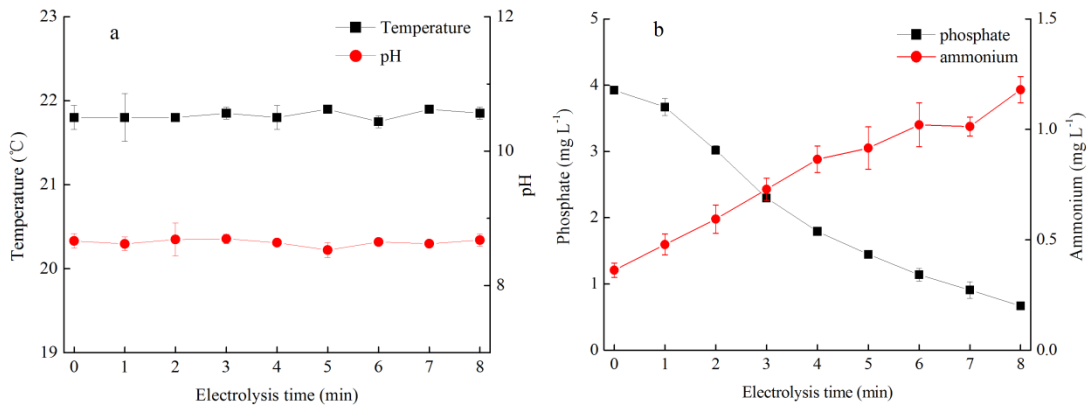


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508 **Fig. 6**



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