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1 **Removing arsenate from water using batch and continuous-flow electrocoagulation with**
2 **diverse power sources**

3
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12
13 **Abstract:**

14 Recently, electrocoagulation (EC) has emerged as a promising method to treat
15 contaminated water. In this study, a novel EC system using stainless steel electrodes was
16 applied to remove arsenate (As(V)) in water. The operation time lasting 5 min, electrical
17 potential difference of 7.5 V, and inter electrode distance of 1 cm were identified as optimal
18 condition for removing As(V) (eliminated 92% of As(V) from 0.1 mg As(V)/L solution). In the
19 batch study using a 9 V rechargeable battery and a small 12 V solar panel, the EC reactor
20 removed 93% and 98% of As(V) from 0.1 mg As(V)/L solution, respectively, and As(V)
21 concentration in treated water was lower than that of the WHO's drinking water guideline (0.01
22 mg/L). In the continuous water flow study, a small cost-effective system (reactor volume of 1
23 L, cost \$15AUD) using a small 12 V solar panel could successfully treat 12 L contaminated
24 water per hour. During 4 hours of continuous testing, the system's efficiency in removing As(V)
25 remained constant at > 91%. The lifespan of the sacrificial anode was more than 500 days in

26 the continuous flow mode. The operation cost for the treatment of As-contaminated water is
27 0.240 A\$/m³ water for the DC electrical system (treated water As(V) 0.008 mg/L) and 0.262
28 A\$/m³ for the solar system (treated water As(V) 0.004 mg/L). In a pilot field study with
29 contaminated groundwater (0.030 mg As/L) in Hanam province, Vietnam, the EC system using
30 solar energy (12 V) was able to reduce As concentration to below the Vietnamese and WHO
31 permissible drinking water limit in 1 min.

32

33 **Keywords:** *Arsenate removal; Electrocoagulation; Stainless steel electrode; Solar energy;*
34 *Water treatment.*

35

36 **1. Introduction**

37 Arsenic (As) is one of the world's most hazardous contaminants. It is estimated that more
38 than 200 million people in 100 countries have been affected by drinking As polluted water [1].
39 As being a potential human carcinogen, causes many other diseases related to skin, lungs, etc.
40 [2]. It is distributed in the environment through natural and anthropogenic processes [3]. The two
41 most toxic forms of As are arsenite (As(III)) and arsenate (As(V)), and their concentrations are
42 controlled mainly by redox conditions and solubility. Whilst As(III) occurs at low redox
43 potential, and its solubility is 10 g/100 mL of water, As(V) appears at a higher redox state and
44 its solubility is up to 66 g/100 mL of water [2]. Although many technologies have been
45 developed and applied to remove As from drinking water [4], As treatment is still a great
46 challenge for the water industry, especially at a small and affordable scale applicable to rural
47 and isolated areas. Among these methods, electrocoagulation (EC) is a promising method,
48 which could eliminate many of the disadvantages of other traditional water treatment techniques
49 [5, 6]. For example, adsorption has the problem of adsorbent regeneration, interference from
50 co-existing anions, and production of toxic exhausted adsorbent waste, while chemical
51 precipitation requires large amounts of chemical coagulants and sludge disposal problem. Some

52 advantages of EC are its simple equipment requirements, easy operation, no use of chemicals,
53 oxidisation of As(III) to As(V) during the process, and less sludge production [6–8]. However,
54 removing As through this method strongly depends on the operational conditions (type of
55 electrodes, voltage, current, inter-electrode distance, operation time, solution pH and initial As
56 concentration) and an electrical source that can be expensive or unavailable in many rural or
57 isolated regions [9].

58 Many kinds of metal can be used as electrodes such as iron (Fe), aluminium (Al), stainless
59 steel, copper (Cu), titanium (Ti), and zinc (Zn) [10]. Among these, Fe electrodes are the most
60 widely used. Ucar et al. (2013) [11] reported that when Fe electrodes in monopolar parallel
61 electrode connection mode were employed, EC could remove up to 95% As(V) from 0.5 mg/L
62 synthetic water at an electrical input of 4.5 mA/cm² in a batch experimental process. The hybrid
63 electrodes with the combination of Fe, Al, or Cu were also tested for removing As [12]. The
64 removal efficiency of the EC technique on treating As(V) with the above electrodes was
65 reported to be 75 to 99% [10]. The application of stainless steel electrode also produced high
66 As removals [13–15]. Though Fe electrodes are the most popular electrodes used in the EC
67 process they can create a problem of water turning into a yellow colour, principally due to the
68 production of fine particles of rust. To overcome this challenge, stainless steel anodes have been
69 used [16].

70 In order to reduce the electricity cost of the EC method, a few studies used solar power
71 in EC to remove different contaminants in water. An EC system utilising solar power and
72 batteries (60 W capacity) was shown to remove 92% of chemical oxygen demand and 49%
73 of total dissolved solids by Al electrodes at a distance apart of 1 cm in 20 min [17]. In another
74 study, an EC system using a 30.2 V solar-photovoltaic module (containing 12 polycrystalline
75 silicon cells) was able to remove 99.9% of lead (Pb) from a solution containing 10 mg Pb/L in
76 10 min [18]. Studies on the application of solar energy in the EC system for removing As from
77 water are limited. Recently, Oh et al. [19] used an EC system with large solar panels (380 to

78 750 W) to remove As from groundwater in Vietnam. Although this system obtained a high As
79 removal efficiency (96.6% from a solution containing 0.376 mg/L in 10 min), this system was
80 large in size, and therefore the initial cost to install it was probably high. Moreover, it can only
81 be installed, operated, and maintained by qualified technicians, and not by local residents.
82 Instead of this type of large centralised solar energy system, a simpler and smaller sized EC
83 unit using smaller solar panels applicable to individual households, as and when clean water is
84 required, is more suitable and affordable for rural areas.

85 In the present study, a new EC process using stainless steel electrodes was utilised to
86 remove As(V) from synthetic water. The objectives of the study were to: firstly, determine the
87 removal efficiency of As(V) from synthetic solution using an EC system with stainless steel
88 electrodes in both batch and continuous flow mode experiments; secondly, determine the
89 influence of many operating conditions of the EC system on As(V) removal, including
90 voltage/current intensity, inter-electrode distance, operation time and solution pH; thirdly,
91 compare different power supply sources (DC, rechargeable battery, and solar power) on As(V)
92 removal and their cost of operation; and fourthly, test the EC system in a pilot field trial with
93 contaminated groundwater collected from a household in Vietnam rural area to evaluate its
94 practical applicability. The pilot trial was conducted in Hanam province, Vietnam while the
95 laboratory studies were conducted in Sydney, Australia. If the small sized solar panel is found
96 to be effective at low operational cost, this could prove to be attractive for decentralised systems
97 in rural or isolated areas used by individual households.

98

99 **2. Material and Methods**

100 **2.1 Feed solution**

101 For the laboratory study, a stock solution was prepared by dissolving 4.165 mg sodium
102 arsenate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$) in 1 L Milli-Q water to obtain a concentration of 1 mg As(V)/L.
103 The stock solution was diluted with distilled water to the desired As(V) concentrations of 0.1,

104 0.25, and 0.5 mg/L for batch studies and 0.1 mg/L for continuous flow mode studies. The ionic
105 strength of the solution was maintained at 1×10^{-3} M NaCl. The initial solution pH was adjusted
106 to 6.0 – 8.0 (the pH range found to occur in groundwater) to study the effect of pH on As(V)
107 removal, and in the other experiments the pH was kept constant at 7.0 ± 0.2 by adding 0.1 M
108 HNO_3 or 0.1 M NaOH.

109 For the pilot study, a contaminated groundwater sample collected from a household in Ly
110 Nhan rural district, Ha Nam province, Vietnam was used. Hanam province is the most As-
111 polluted area in the country. According to Huy et al. [20] and Nguyen et al. [20, 21], more than
112 52% of groundwater in this province is contaminated by As. This water had pH and As, Fe
113 concentrations of 7.2, 0.030 mg/L and 0.27 mg/L, respectively.

114

115 **2.2 EC system**

116 **2.2.1 Laboratory study**

117 A couple of commercial stainless-steel plates of grade 316 (11 cm \times 6 cm \times 0.09 cm)
118 were used as anode and cathode. The electrodes were hung inside a transparent polypropylene
119 tank (size of 10.2 \times 10.2 \times 20 cm) by a glass stick in a monopolar parallel connection mode, so
120 that the submerged part of the electrodes inside the solution was 8 cm \times 6 cm \times 0.09 cm. The
121 solution volume was 1L. To maintain a uniform concentration, the solution was mixed using a
122 magnetic stirrer rotated at 120 rpm at the bottom of the tank. Batch and continuous-mode
123 experiments with DC electricity and rechargeable battery were conducted using this set-up at
124 room temperature of 25 ± 1 °C.

125 In the batch studies, a regulated laboratory DC power supply system (model MP-3840)
126 was used to supply an electrical current with predetermined voltages to the electrodes (Fig. 1a).
127 The voltages and generated electrical current supplied by this DC system could be varied
128 through its converter. The performance of the EC system on As(V) removal was investigated
129 at different conditions: electric potential difference 3.0 – 10 V, operation time 5 – 120 min,

130 initial As(V) concentration 0.1 – 0.5 mg/L, the distance between electrodes 0.5 – 4.0 cm, and
131 solution pH 6 – 8. In each experiment, samples were periodically collected, filtered through
132 0.45 µm filters, and the filtrates were analysed for As using an ICP-MS instrument (Agilent
133 Technologies 7900). The experimental scenario, which produced an As(V) concentration
134 smaller than the WHO's permissible limit for As in drinking water with the lowest energy
135 consumption, was considered to be the optimal treatment condition. The initial and final pH of
136 the solution in all experiments were measured using a pH meter (model HQ40D) to investigate
137 whether the pH of the solution changed and, if so, to what extent.

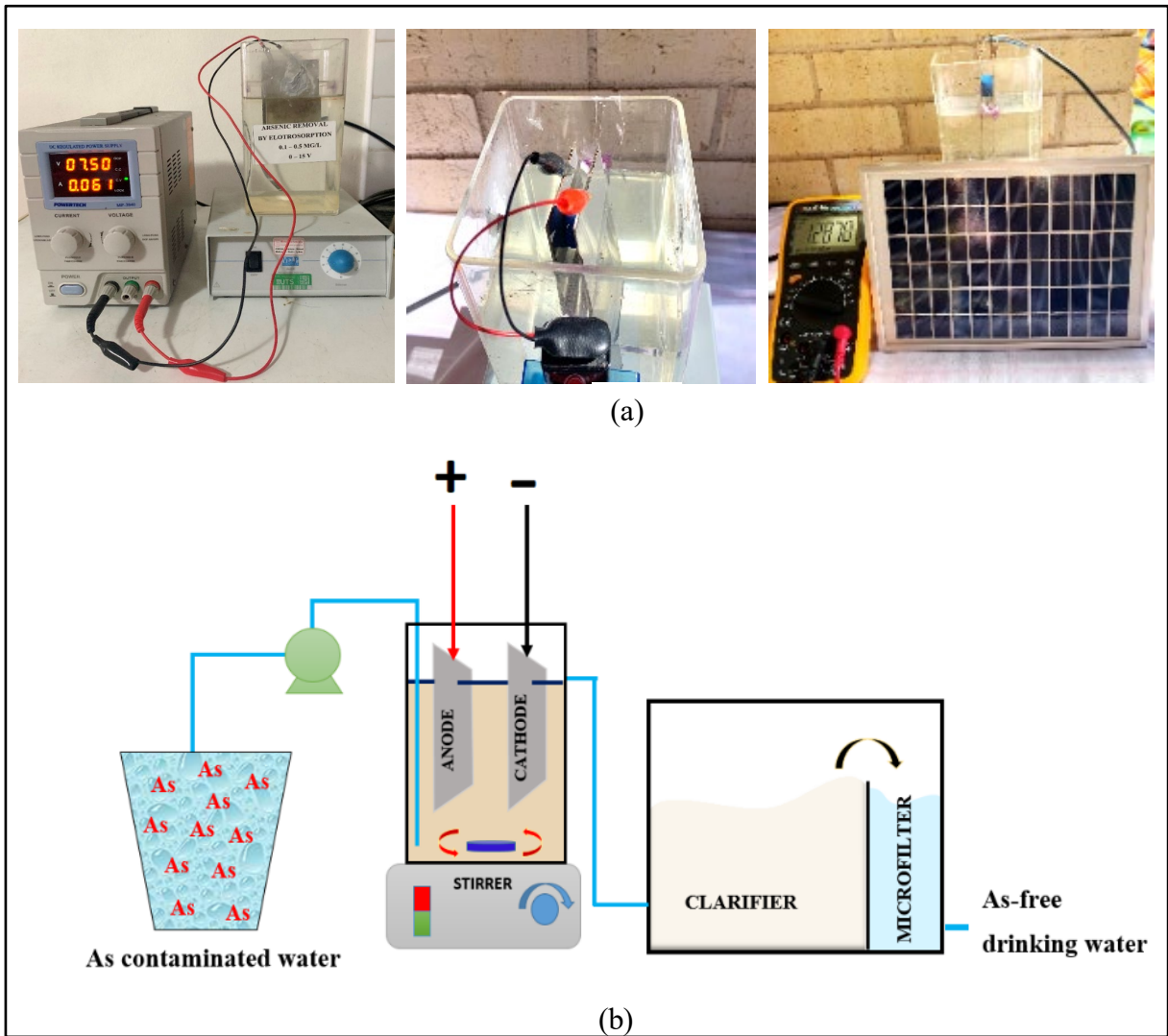
138 In order to evaluate the feasibility of using renewable energies, two other experiments
139 were carried out at the optimal condition of the EC system determined in the initial experiments,
140 using a 9 V rechargeable battery and a 12 V solar panel separately instead of the DC source of
141 electricity (Fig. 1a). The 9 V rechargeable battery was simply connected to electrodes by using
142 clips. The small 12 V solar panel (Powertech) was designed with 36 multi-crystalline silicon
143 solar cells and a maximum power of 5 watts. The size of the solar panel was 25.1 cm × 20.5 cm
144 × 1.8 cm with a weight of 0.7 kg. It was fitted with a blocking diode making it suitable for direct
145 connection to the EC system by means of clips without using any intermediate equipment. The
146 experiments with the solar panel were conducted outdoors and in the winter season from 10.00
147 am to 14.00 pm (July and August 2020, Australia). The voltage and current density of the solar
148 panel were measured using a Multimeter Voltage Sensor (model QM1529). It demonstrated
149 that the solar energy during this period was enough for the EC system's operation.

150 The operation of the solar panel, in general, depends on the weather conditions. On days
151 when the sunlight is insufficient, it is recommended that a simple and low-cost rechargeable
152 battery be employed for storing solar energy captured during bright sunny periods and then
153 supply this stored power for the EC system on days with insufficient sunlight.

154 To evaluate the sacrifice (dissolution) rate of the anode, in addition to the calculations
155 using Faraday's Law (see Section 3.1.2), a chemical analysis was conducted to measure the Fe

156 concentration in the EC reactor. 1 mL of suspension in the EC reactor at different operation
157 times and currents was sampled and mixed with 5 mL each of 2M HNO₃ and 1M HCl. The
158 mixture was then shaken at 50 rpm at room temperature for 1 hour, filtered and analysed for Fe
159 using an ICP-MS instrument (Agilent Technologies 7900). This is a modification of the method
160 utilised by Kalaruban et al. [5].

161 In the continuous flow mode study, both the DC power supply system and the small 12
162 V solar panel were utilised separately with a small EC reactor (containing 1L of As-
163 contaminated water) to investigate the As(V) removal efficiency. The system was operated at
164 different flow velocities of 12 L/h, 6 L/h, and 3 L/h corresponding to hydraulic retention times
165 (HRT) of 5, 10, and 20 min [HRT (min) = reactor volume (L)/flow velocity (L/h) × 60 (min)].
166 A schematic diagram of the continuous flow reactor is presented in Fig. 1b. The synthetic feed
167 solution containing 0.1 mg As(V)/L was pumped continuously by a dosing pump (Masterflex
168 L/S) into the 1 L reactor at the optimal operating condition determined in the batch study (U =
169 7.5 V, I = 0.030 A, pH = 7.0 ± 0.2, electrodes distance 1 cm). The treated solution then
170 automatically flows into the 2 L clarifier. The effluent samples in the clarifier were collected
171 after 1, 2, and 4 h, then they were filtered and analysed for As, Fe, nickel (Ni) and chromium
172 (Cr) using an ICP-MS instrument (Agilent Technologies 7900). Ni and Cr were analysed, as
173 they are also components of stainless steel.



174

175 **Fig. 1.** Installation of EC system with (a) DC power supply, 9 V rechargeable battery and 12
 176 V solar panel in batch EC reactor and (b) the schematic diagram of the EC continuous flow
 177 reactor

178

179 **2.2.2. Pilot trial in the field with contaminated ground water in Vietnam**

180 The pilot trial was conducted in Hanam province, Vietnam. A small 1.15 L acrylic tank
 181 equipped with two commercial 316 stainless-steel plates (size of 11 cm × 10 cm × 0.03 cm) and
 182 a small solar panel (12 V) was used as the EC reactor. As-contaminated groundwater from a
 183 household was treated by the same method as used in the laboratory batch study. The trial was
 184 carried out at different operation times of 1, 5, 10, 15 and 20 min and the inter-electrode distance
 185 was kept constant at 1.0 cm. Effluent from the EC system was filtered through 0.45 μm filter,

186 and the filtrate was analysed for As and Fe using an ICP-MS instrument (Perkin Elmer Elan
187 DRC 9000 ICP-MS).

188

189 **3. Results and discussion**

190 **3.1 Batch study**

191 **3.1.1 Effect of initial concentration and operation time**

192 Effect of initial As(V) concentration (C_o) was investigated at three concentrations, 0.1,
193 0.25 and 0.5 mg/L. In this experiment, the distance between electrodes, initial pH, and electrical
194 potential difference (U) were 1 cm, 7.0, and 3 V, respectively. Under these conditions, the
195 current produced was very small, only 0.007 A. Samples from the reactor container were
196 collected at 5, 10, 15, 30, 45, 60, 90, and 120 min, and analysed for As, pH, and Fe. Fig. 2a
197 depicts the effect of C_o on the residual As(V) concentration in water samples as a function of
198 operation time (t_{EC} (min)). As can be seen from this figure, the WHO permissible limit for As
199 in drinking water ($C_{WHO} = 10 \mu\text{g/L}$, [22]) was attained after 60 min for C_o of 0.1 mg/L and 90
200 min for C_o of 0.25 mg/L. At C_o of 0.5 mg/L, the C_{WHO} could not be obtained within the 120
201 min tested in the experiment. Since As(V) concentration of around 0.1 mg/L is the typical level
202 reported for As-contaminated groundwater in many countries [23], it was chosen for use in the
203 subsequent studies.

204 Fig. 2a also shows that the As(V) removal rate was fast during the first hour and then
205 decreased slowly with time for all As(V) concentrations as reported by others [8, 13]. This
206 decline in the rate of As(V) removal is due to the decrease in solution concentration of As(V),
207 which was in equilibrium with the As(V) adsorbed on the hydrous iron oxide/hydroxide
208 precipitates formed from Fe dissolution derived from the anode during the EC process. When
209 the As(V) concentration drops, so does the maximum loading of As(V) per mg of Fe precipitates
210 in equilibrium [24]. These precipitates were formed by the reaction of Fe with the OH^- ions
211 generated in the cathode at the neutral pH of the solution [7, 13]. It is also possible that at longer

212 EC times the As(V) concentrations became very small to produce any additional Fe/As co-
213 precipitation, mainly because the solubility products of Fe/As precipitates would not have
214 reached at these low As(V) concentrations.

215

216 3.1.2 Effect of electrical current and anode dissolution

217 The effect of electrical current was studied by keeping the distance between electrodes,
218 C_0 , and pH constant at 1 cm, 0.1 mg/L, and 7.0, respectively. The voltage of power supply
219 source (U) was adjusted to 3 V, 5 V, 7.5 V, and 10 V, and the corresponding values of the
220 electrical current (I) were measured as 0.007 A, 0.018 A, 0.030 A, and 0.061 A, respectively.
221 The experiment remained in operation for up to 120 min. Results showed that C_{WHO} could be
222 reached in 15 min at 0.018 A (U = 5 V), which is much faster than that at 0.007 A (U = 3 V)
223 (Fig. 2b). At the higher currents of 0.030 A (U = 7.5 V) and 0.061 A (U = 10 V), C_{WHO} was
224 reached extremely quickly (within 5 min). Consistent with these data, the increase in the
225 electrical current led to rising As(V) removal efficiency. For example, at t_{EC} of 5 min the
226 removal efficiency (E%) increased from 24% to 58.5%, 92%, and 96%, corresponding to the
227 rise of the current values from 0.007 A to 0.018 A, 0.030 A, and 0.061 A (current densities of
228 0.11, 0.27, 0.45, and 0.92 mA/cm²), respectively. The possible reason for this is that at a higher
229 electrical current, a larger amount of Fe from the anode was dissolved resulting in greater
230 amounts of hydrous iron oxides/hydroxides production [7, 13, 24]. This led to the removal of a
231 larger percentage of As(V) due to As(V) replacing the increasing number of hydroxyl groups
232 in the iron precipitates [7]. It is also possible that at high concentrations of Fe, larger amount of
233 As is removed by co-precipitation with Fe [25]. Other studies also reported that As removal
234 efficiency was proportional to the current density [7, 15, 24].

235 Fig. 2b also shows that the As(V) removal efficiency significantly increased when t_{EC}
236 increased from 5 min to 120 min. For example, at the current value of 0.018 A (U = 5 V), As(V)
237 removal efficiency at 5 min was only 58.8%. However, it increased to 79.7% after 10 min and

238 reached almost 99% at 30 min. The increase of t_{EC} elevated the amount of dissolved Fe as
 239 observed for the effect of an increase in current. The higher amounts of dissolved Fe would
 240 have produced abundant Fe hydrous oxides/hydroxides, leading to a greater amount of As(V)
 241 being removed by adsorption. Greater amounts of As(V) are also removed by co-precipitation
 242 with Fe at high concentrations of Fe [25].

243 In order to evaluate the sacrifice (dissolution) rate of the anode, the mass of Fe generated
 244 at the anode m_{Fe} (mg), and the electrode consumption concentration $C_{electrode}$ (mg/L)
 245 (calculation based on m_{Fe}) for the As removal from water were calculated using chemical
 246 analysis and Faraday's Law. The Faraday's Law equations are presented below [7]:

$$247 \quad m_{Fe} = \frac{I \times t_{EC} \times M_{Fe}}{z \times F} \quad [1]$$

$$248 \quad C_{electrode} = \frac{I \times t_{EC} \times M_{Fe}}{z \times F \times v} \quad [2]$$

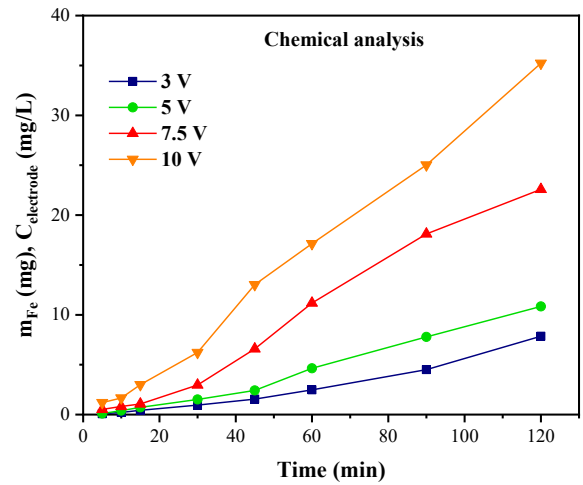
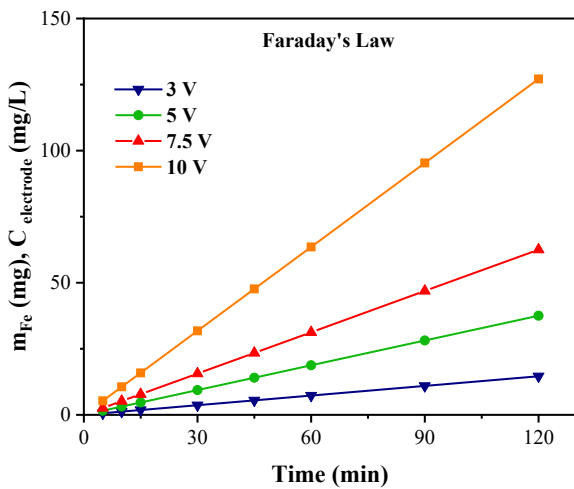
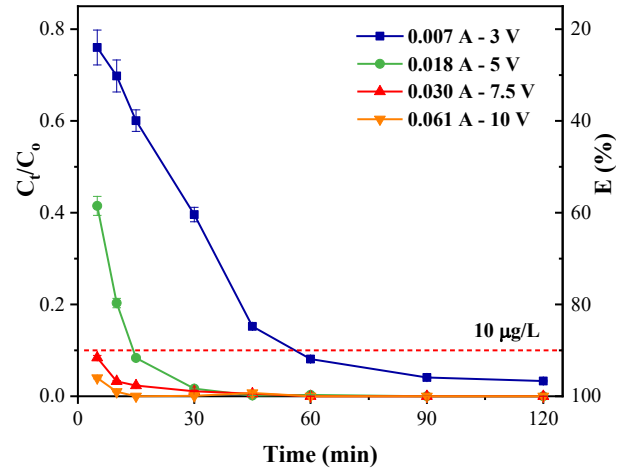
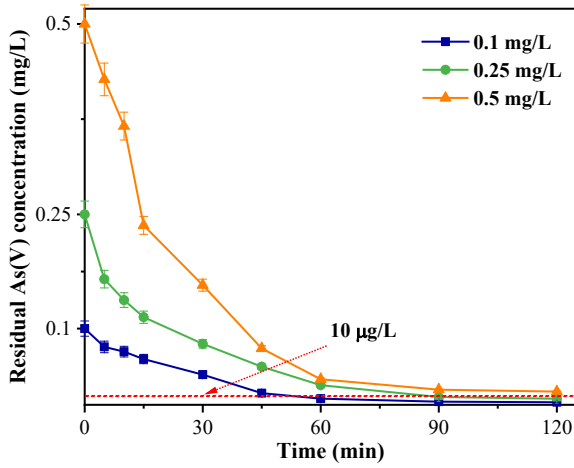
249 where I is the current value (A), M_{Fe} is the molecular weight of iron (55,850 mg/mol), z is the
 250 number of electrons involved in the oxidation/reduction reaction ($z_{Fe} = 2$), F is the Faraday's
 251 constant (1F = 1608.06 A.min/mol), t_{EC} is EC operation time (min), v is the solution volume (v
 252 = 1 L).

253 Fig. 2c depicts that the calculated amounts of Fe ions generated as well as the weight of
 254 the anode lost increased with I and t_{EC} . For example, the increase of I from 0.007 A to 0.061 A
 255 led to a rapid increase in the sacrifice rate of the anode, from 5.2 mg/L to 127.1 mg/L after 120
 256 min reaction. The Fe concentrations calculated from this equation at the end of the experiment
 257 ($t_{EC} = 120$ min), at I = 0.007, 0.018, 0.030 and 0.061 A were 14.6, 37.5, 62.5 and 127.1 mg/L,
 258 respectively. However, the corresponding concentrations of Fe dissolved from the anode
 259 measured by chemical analysis were much smaller, i.e. 7.9, 10.8, 22.6 and 35.2 mg/L,
 260 respectively (Fig. 2d). A possible reason for the difference in the chemical analysis and
 261 calculated values is that Faraday's law is valid only when all the electrons in the system
 262 participate solely in the metal-dissolution reaction at the anode, which seldom happens. Another
 263 reason for the lower measured value is that the anode used does not consist of pure Fe; it is

264 stainless steel, which contains other metals such as Cr, in addition to Fe. This is not taken into
265 consideration in Faraday's equation.

266 The initial solution pH of 7.0 increased slightly up to a maximum of 8.0 during the EC
267 process. The precipitation of Fe dissolved from the anode would have reduced pH, but the
268 amounts of OH⁻ generated at the cathode and OH⁻ ions released during As adsorption on the
269 hydrous Fe oxide/hydroxides would have been more than the H⁺ released during Fe
270 precipitation. These reactions are possible reasons for the slight increase in solution pH.

271 According to the above results, $t_{EC} = 5$ min and the potential of 7.5 V were identified as
272 optimal conditions for As(V) removal. In these conditions, C_{WHO} can be reached quickly (only
273 5 min compared to 15 and 60 min at the potentials of 3 V and 5 V, respectively) while ensuring
274 less energy usage than that at the potential value of 10 V. Therefore, these conditions (5 min,
275 7.5 V) were chosen for analysis in the subsequent studies.



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281 **Fig. 2.** The effect of (a) As(V) initial concentration at 3V and (b) electrical current on As(V)
282 removal; The sacrifice rate of anode in the EC process as (c) calculated using Faraday's Law
283 and (d) measured using chemical analysis, at $C_o = 0.1$ mg/L, electrodes distance apart = 1 cm,
284 $pH_{initial} = 7.0$. The 10 μ g/L horizontal line within Fig. 2 (a) and (b) indicates As concentration
285 limit in the WHO drinking water guideline.

286

287 3.1.3 Effect of distance between electrodes

288 This study was conducted at C_o of 0.1 mg/L, pH of 7.0, t_{EC} of 5 min, U of 7.5 V, and
289 varying electrodes distance apart of 0.5, 1.0, 2.0, 3.0, and 4.0 cm. As can be seen from Fig. 3a,
290 As(V) removal efficiency and current values increased when the distance between electrodes
291 was reduced. At electrodes distance apart of 0.5 cm, the current intensity and As(V) removal
292 efficiency reached 0.087 A and 99.5%, respectively. These values fell to 0.030 A and 92%,

293 respectively, at an electrodes distance apart of 1 cm. When the electrodes distance apart
294 increased to 2, 3, and 4 cm, the EC system increasingly proved to be inefficient for removing
295 As(V), reaching values of less than 20%. This decline in removal efficiency can be explained
296 as being caused by the increase in the space between the electrodes, which in turn decreased
297 the current generated (down to 0.003 A). The end result was less Fe dissolution from the anode
298 for its interaction with As(V) ions [5]. Similar outcomes were reported for the removal of other
299 contaminants from water via the EC process, such as removing water hardness using Fe-rod
300 electrodes [26], mercury using Al and Fe electrodes [27], and indium ions using Fe electrodes
301 [28].

302 Although the inter-electrode distance of 0.5 cm produced the best As(V) removal
303 efficiency, the cost of electrical power (voltage x current) consumed is high due to the high
304 current generated. Therefore, the next shortest inter-electrode distance of 1 cm was selected as
305 the optimal distance and applied in subsequent studies. Moreover, this distance is more practical
306 to maintain in the EC installation set-up.

307

308 **3.1.4 Effect of solution pH**

309 The solution pH is one of the important factors influencing contaminants removal in the
310 water treatment process. Investigation of the effect of solution pH was conducted at pH varying
311 from 6 to 8 because it is a common pH range in most As-contaminated groundwater [8]. Fig.
312 3b shows that As(V) removal efficiency slightly increased when pH was increased from 6 to 7
313 and then decreased as the pH is further increased to 8. The decrease in As(V) removal at lower
314 pH is possibly due to the dissolution of iron hydroxide precipitates that were responsible for
315 adsorbing the added As(V) [13]. The reduction in As removal at the higher pH of 8 could be
316 due to reduced adsorption of the more negatively charged As species on the negatively charged
317 iron hydroxide precipitates [23]. It is well known that the negative charges on the surface of Fe
318 oxide/hydroxide [29] and on the As species increase (H_2AsO_4^- to HAsO_4^{2-}) with pH [30].

319 Based on the results of all the batch studies conducted, it can be concluded that the
 320 optimum As(V) removal at C_o 0.1 mg/L could be achieved at pH of 7, t_{EC} of 5 min, electric
 321 potential of 7.5 V (corresponding to the current value of 0.030 A), and electrodes distance apart
 322 of 1 cm. These conditions were therefore considered as the optimal conditions for the EC
 323 process and employed in the subsequent studies on the use of alternative power sources and
 324 continuous flow mode process.

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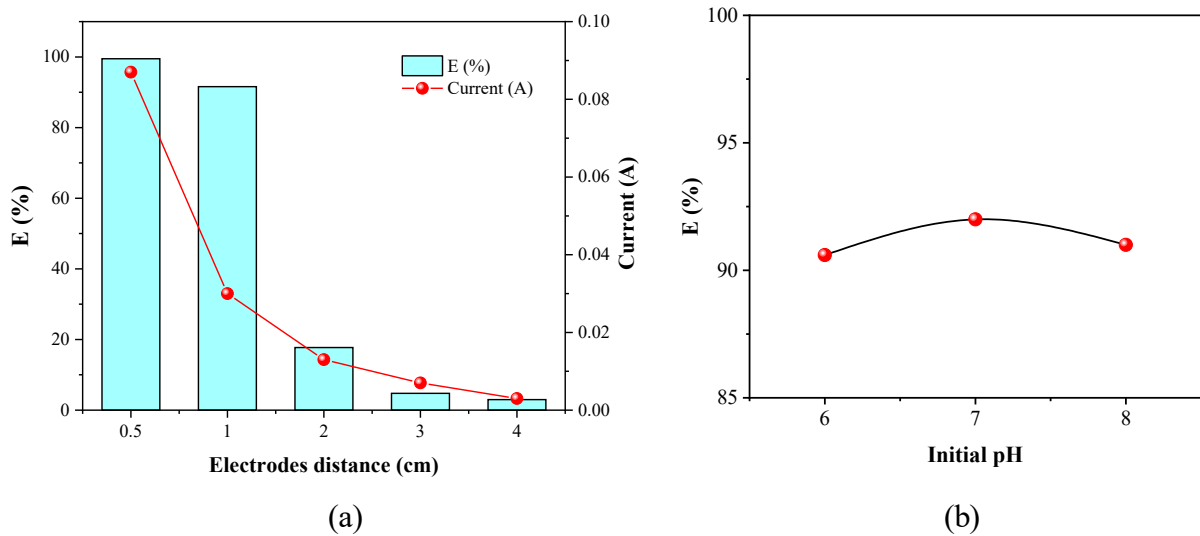
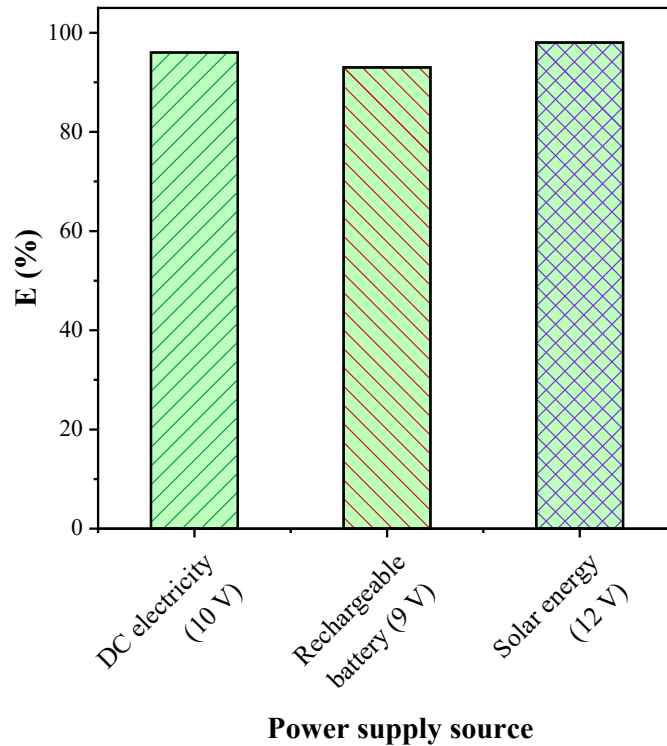


Fig. 3. The effect of (a) distance between electrodes on As(V) removal efficiency and current generated at pH 7 and (b) initial pH on As(V) removal efficiency at electrodes distance apart = 1 cm, $C_o = 0.1$ mg/L, $U = 7.5$ V, $t_{EC} = 5$ min

3.1.5. Use of alternative power supply sources

334 Batch experiments with alternative power supply sources were conducted using either a
 335 12 V solar panel or 9 V rechargeable battery and the As(V) removal efficiencies were compared
 336 with 10 V DC electricity source. These experiments can provide vital information on whether
 337 renewable energy sources can replace the DC power supply used to treat As(V) in the EC
 338 system. Fig. 4 shows that the As(V) removal efficiency of both renewable energy sources was
 339 approximately the same as that of the DC power supply (93% for rechargeable battery and 98%
 340 for solar panel compared with 96% for the DC electrical source, the slight differences are due

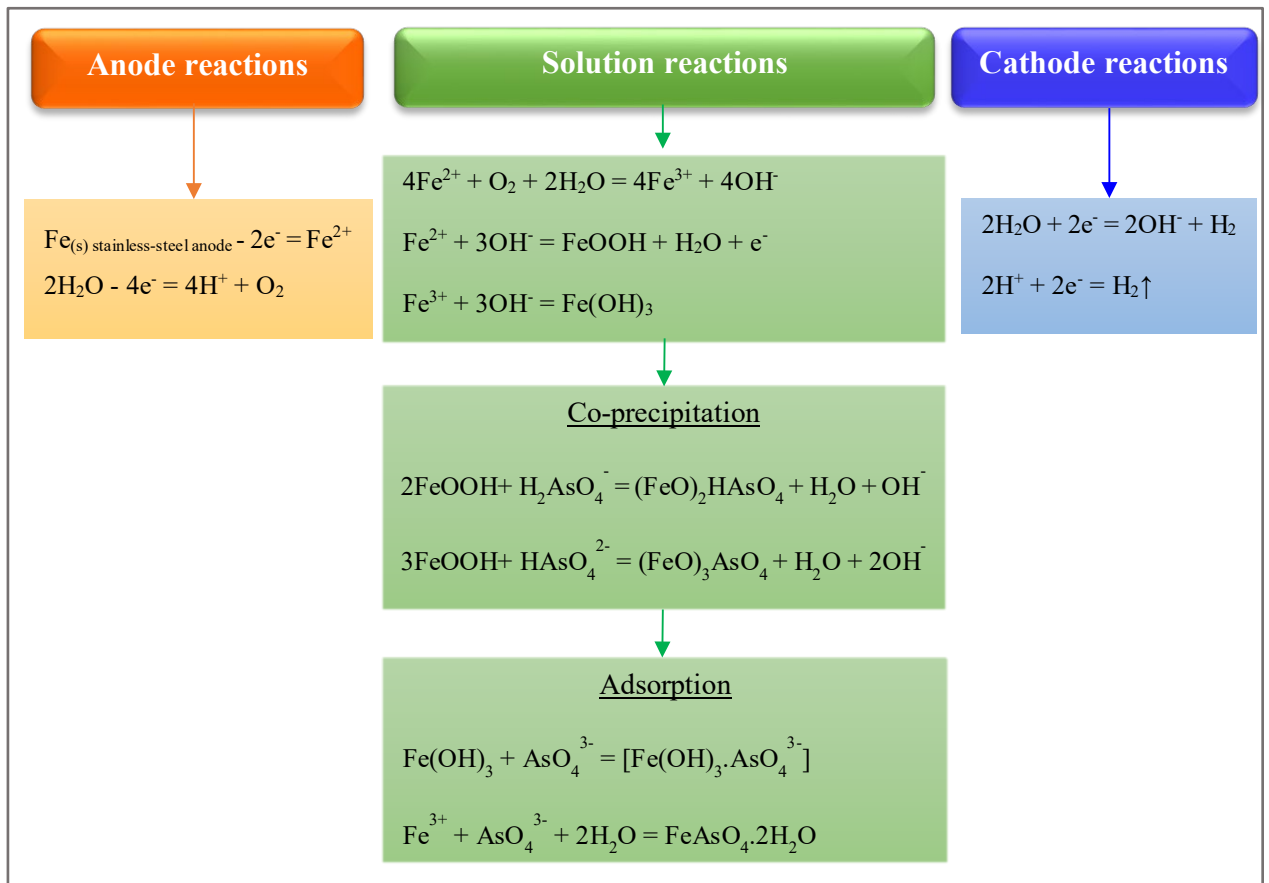
341 to the differences in voltage). This result indicates that renewable energy sources such as a
342 rechargeable battery or a small-scale solar energy system can remove As(V) effectively in
343 miniature portable EC systems that are suitable and affordable for rural households.
344



345
346
347 **Fig. 4.** The As(V) removal efficiency of EC system using different energy sources in batch
348 study, $C_o = 0.1$ mg/L, pH = 7, $t_{EC} = 5$ min and electrodes distance apart = 1 cm
349

350 3.2. Mechanisms of As(V) removal in the EC process

351 The main chemical reactions in the EC process and the reactions involving As(V) removal
352 from the solution are presented in Fig. 5 [31]. The two prime mechanisms of As(V) removal in
353 the EC process are co-precipitation of As(V) with the various Fe ion species generated in the
354 EC process and adsorption of As(V) on the hydroxide precipitates of Fe^{2+} and Fe^{3+} . These are
355 depicted in the last four equations in the solution compartment of Fig. 5 [32].
356



357

358 **Fig. 5.** Chemical reactions in the EC process and As removal mechanisms

359

360 3.3. Continuous flow mode study

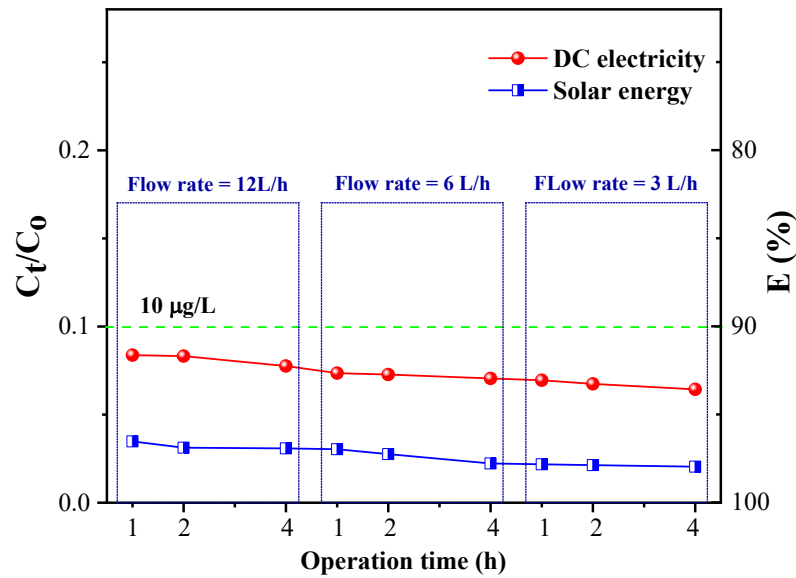
361 This study was conducted in two EC systems. The first EC system was operated with the
 362 7.5 V DC electrical source, which was the optimal voltage found in earlier experiments (section
 363 3.1.2), and the second one was with the small 12 V solar panel. Solution containing 0.1 mg
 364 As(V)/L was continuously supplied to the EC reaction chamber at three different flow rates (12,
 365 6, 3 L/h, corresponding to HRT of 5, 10 and 20 min). Fig. 6 shows that the As(V) removal
 366 efficiency in both systems was maintained at more than 91% during 4 h operation at all 3 flow
 367 rates. The residual As(V) concentration in the effluent was always lower than that of the WHO
 368 drinking water guideline. The solar energy system's As(V) treatment efficiency (> 96%) was
 369 higher than that of the electrical system (> 91%) at all flow rates due to the higher voltage of
 370 the solar panel. Results also show that the Fe concentration was below the WHO guideline
 371 value and the Vietnam's water quality standard of 0.3 mg/L for drinking water [33, 34]. The

372 effluent concentration of Ni and Cr was 0.0132 and 0.0274 mg/L, respectively. These values
373 were much lower than that of the WHO's drinking water guideline for these metals (0.07 mg/L
374 for Ni and 0.05 mg/L for Cr [35]).

375 Fig. 6 also shows that a decrease of flow rate (an increase of HRT) increases the efficiency
376 of As(V) removal. This is because at higher HRT, As(V) had longer time to interact with Fe
377 hydroxides leading to greater adsorption and co-precipitation.

378 The results showed that the solar-EC system with a flow rate of 12 L/h could be chosen
379 for use in decentralised systems in rural or isolated areas to remove As(V). Though lower flow
380 rates (3 L/h and 6 L/h) can remove higher percentages of As(V), the volume of water treated
381 would be lower. However, for contaminated water containing very high As concentrations,
382 lower flow rates (higher HRTs) may be necessary. At the flow rate of 12 L/h, an EC reactor of
383 a small volume of 1 L could provide 12 L clean water per hour. This flow rate is nearly the
384 same as that of many commercial household filters currently being used. On a normal day, the
385 system can produce 48 L of As(V)-free water during a 4-hour operation, which is enough to
386 meet the drinking water demand of the average family.

387 Although the solar energy system has many advantages such as simplicity, cost-
388 effectiveness, and non-electrical appliance requirements, its operation depends on the weather
389 conditions. The recommended solution for this issue, as stated in section 2.2, is using an
390 appropriate rechargeable battery for storing solar energy during favourable weather conditions
391 and using it when solar energy is limited.



392

393

394 **Fig. 6.** The As(V) removal efficiency of EC system using DC electricity (7.5 V) and solar
 395 energy (12 V) at various flow rates, feed solution concentration $C_o = 0.1$ mg/L, pH = 7,
 396 electrodes distance apart = 1 cm. C_t (mg/L) is As concentration in the treated solution at time t
 397 (h). The 10 µg/L horizontal line within the figure represents As concentration limit in the WHO
 398 drinking water guideline.

399

400 3.4. EC operation cost

401 Operation cost is one of the most important factors in water treatment technologies
 402 because it decides the applicability of the treatment method in real-life situations, which
 403 primarily depends on users' budgets. The operation cost of the EC system in continuous flow
 404 mode was calculated for DC and solar power sources according to Eq. 3 and presented in Table
 405 1. Here the price of Australian electricity was assumed to be A\$0.30/kWh [36]:

406

$$407 \text{ Treatment cost/m}^3 \text{ water} = \text{Electricity cost/m}^3 + \text{Electrode cost/m}^3 + \text{Reactor cost/m}^3 \quad [3]$$

408

409 The lifespan of the anode for the small-scale reactor was determined by measuring the
 410 amount of Fe in solution. For a given reactor of 1L containing an initial As(V) concentration of
 411 0.1 mg/L operated at the optimal conditions (pH 7.0, electrode distance 1 cm, flow rate 12 L/h,
 412 $U = 7.5$ V for DC power or 12 V for solar energy), the sacrifice rate of anode $C_{\text{electrode}}$ was

413 measured to be 0.516 mg/L for the DC electrical EC system and 0.664 mg/L for the solar energy
414 EC system in 5 min. In one day, the EC system could treat 48 L water for 4 hours (1 m³ in 20.83
415 days at a flow rate of 12 L/h; see Section 3.3 for calculation) and consume 24.77 and 31.87 mg
416 of the anode for DC electricity and solar energy system, respectively. The weight of the
417 submerged part of the selected anode in this reactor was measured as 32,000 mg. Assuming
418 that the anode can be effectively used until 50% of it is dissolved, the anode could be used for
419 645 (32,000 mg/24.77 mg/day x 50%) days using DC electricity source (at U = 7.5 V) or 502
420 (32,000 mg/31.87 mg/day x 50%) days using solar energy (at U = 12 V). The total volume of
421 treated water produced per lifetime of the anode is calculated as 31.0 m³ (645 days/20.83
422 days/m³ water) using the DC electricity source and 24.1 m³ (502 days/20.83 days/m³ water)
423 using a solar panel source.

424 **Table 1.** Cost calculation for EC system operation using DC electricity and solar energy (based
 425 on flow rate 12 L/h, operation time 4 h/day, and treatment capacity 0.048 m³ water/system.day
 426 or 20.83 day/m³ water)

	Formula	DC system	Solar energy system
Voltage (V)	U	7.5	12
Current (A)	I	0.03	0.08
Power (W)	$U \times I$	0.225	0.960
Operation time in 1 day (h)	H	4	4
Energy consumption in 1 day (kWh)	$W \times h \times 10^{-3}$	0.0009	0
Energy cost in 1 day (A\$)	$A\$0.30/\text{kWh} \times \text{kWh}$	0.00027	0
Energy cost for 1 m ³ treated water (A\$/m ³)	$A\$0.30 \times \text{kWh} \times 20.8 \text{ d}/\text{m}^3$	0.00563 (1)	0 (1)
Electrodes cost (A\$)		3.0	3.0
Mass of electrode dissolved (C _{electrode} , mg/day)		24.77	31.87
Electrodes lifespan (day) (up to 50% dissolution)	$= 32,000 \text{ mg}/(C_{\text{electrode}} \text{ mg}/\text{m}^3) \times 50/100$	645	502
Electrode cost/m ³ treated water (A\$/m ³)	$= A\$3/(\text{lifespan d} \times 0.048 \text{ m}^3/\text{d})$	0.097 (2)	0.125 (2)
Capital cost (Power supply and accessory in a reactor) (A\$) ^a		12	12
Lifetime of power supply (year) ^b		5	5
Reactor/m ³ treated water (A\$/m ³)	$= A\$12/(\text{lifetime y} \times 365 \text{ d} \times 0.048 \text{ m}^3/\text{d})$	0.137 (3)	0.137 (3)
Treatment cost (A\$/m ³): (1) + (2) + (3)		0.240	0.262

427 Note: ^a Cost of the EC reactor, including power supply system DC or solar panel and accessory
 428 like buckets, is A\$12.

429 ^b Assuming that the lifetime of the power supply source is 5 years. According to the
 430 supplier, the lifetime of the solar panel could be 20 years, and at least 10 years for DC
 431 power supply system.

432 The calculation shows that the DC electricity cost in this study is considerably low,
433 0.00563 A\$/m³ of treated water while the cost of the solar energy system is zero. The cost for
434 the treatment of As-contaminated water is 0.240 A\$/m³ water for the DC electrical EC system
435 and 0.262 A\$/m³ for the solar EC system. The slightly higher cost of the solar energy system is
436 due to its higher voltage, which dissolved larger amounts of the anode. This resulted in a higher
437 percentage of As removal resulting in As concentration in treated water (0.004 mg/L) of almost
438 half the value obtained with DC electrical system (0.008 mg/L) (Fig. 6). Such a high percentage
439 of removal is not required for the solution of As concentration 0.1 mg/L used here because the
440 final As concentration is well below the WHO limit (0.01 mg/L). However, for waters
441 containing higher initial As concentrations, this is an advantage.

442 In comparison with the estimated cost reported in other EC studies, the cost estimated in
443 this study is lower. This is primarily because other studies used much higher initial
444 concentration of As which required a very high electrical power to bring down the concentration
445 to below 10 µg/L. For example, Şik et al. [37] reported the electricity and electrode cost as
446 0.546 US\$/m³ (A\$0.746/m³) to reduce the As(V) concentration of 200 µg/L to <10 µg/L in 12
447 min using Fe ball anode with 0.3 A current when the inter electrode distance was kept at 7.5
448 mm. Thakur and Mondal [30] reported treatment cost of 0.357 US\$/m³ (A\$0.488/m³) to reduce
449 the initial As(III) concentration of 550 µg/L to 8 µg/L using Al electrodes at 1 cm distance
450 apart with 10 A/m² current. Their treatment cost included cost of electrode + chemicals +
451 electricity + sludge disposal + fixed cost. Both the above two studies used DC electricity for
452 supplying the power, whereas in the current study the cost was separately estimated for both
453 DC and solar energy systems.

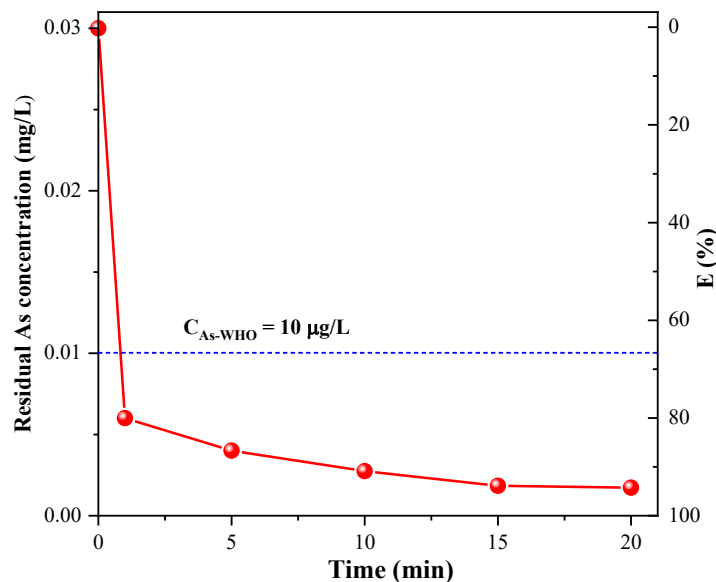
454

455 **3.5. Pilot trial in the field**

456 The pilot trial study was conducted under five different t_{EC} of 1, 5, 10, 15 and 20 min.
457 The results of As removal by the 12 V solar panel EC system are presented in Fig. 7. At the

458 initial As concentration in groundwater of 0.030 mg/L and $t_{EC} = 1$ min, the As was removed
 459 efficiently (80.0% removal) and the treated water had As concentration below the Vietnamese
 460 and WHO permissible limit for As in drinking water (0.010 mg/L) [22, 33]. The As removal
 461 efficiency increased to 86.7, 90.0, 93.9 and 94.2% when the t_{EC} increased to 5, 10, 15 and 20
 462 min, respectively. However, at the shortest time of $t_{EC} = 1$ min the power usage is the lowest
 463 and therefore the cost of operation is most economical. On the other hand, if the initial As
 464 concentration is high, a longer time of operation is required to bring the As concentration in the
 465 treated water to below the WHO limit. Compared to the results of the laboratory batch study,
 466 in the pilot trial a shorter time was found to be sufficient to produce As concentration below the
 467 safety level. This phenomenon can be explained by the lower As concentration in the raw groundwater
 468 in comparison with the initial As concentration used in the laboratory study (0.10 mg/L).

469 The Fe concentration in the EC treated water varied slightly (0.18 – 0.28 mg/L) in
 470 comparison with that in the raw groundwater (0.27 mg/L). However, the Fe concentration in
 471 the treated water was well below the Vietnam and WHO permissible limit for Fe in drinking
 472 water (0.3 mg/L) [33, 34].



473
 474
 475
 476
 477

Fig. 7. As removal efficiency of EC system in a pilot trial study, $C_o = 0.03$ mg/L, . The 10 μ g/L horizontal line within the figure represents As concentration limit in the WHO drinking water guidelines

478 **4. Conclusions**

479 The new small-scale (household) EC system developed with DC power supply using
480 stainless steel electrodes could successfully remove As(V) from contaminated water and
481 maintain the As concentration below the WHO recommended safety level. In the batch study,
482 the ideal scenario for the EC system's operation was determined as pH 7.0, electrodes distance
483 apart of 1 cm, voltage of 7.5 V and current intensity of 0.030 A for initial As(V) concentration
484 of 0.1 mg/L. The batch study using a rechargeable battery and solar energy confirmed that these
485 renewable energy sources could be used instead of the DC source to remove As(V) effectively
486 in the EC system.

487 In the continuous water flow study, both DC electricity and solar energy sources were
488 able to supply the required power for the successful operation of the EC system with a low
489 capital cost of A\$15 and running cost of 0.240 - 0.262 A\$/m³ treated water. The system with
490 both power sources could remove As(V) from water at very high rate of efficiency, more than
491 91-96%. A small volume EC reactor of 1 L could supply enough drinking water daily for a
492 household, and the anode replacement only needs to be done after more than 500 days at a 4
493 h/day of operation. The low-cost EC system can be used with or without a DC electricity source,
494 so it can be implemented in all areas. The laboratory study was repeated in a pilot field trial
495 with a solar panel (12 V) using groundwater in Vietnam (0.030 mg As/L) and it was found to
496 successfully remove As concentration below the Vietnamese and WHO permissible limit in 1
497 min operation.

498

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