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Water purification from metal ions in the presence of organic matter using electromagnetic radiation-assisted treatment

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Abstract

Organic matter is a widely occurring pollutant in freshwater sources due to the extensive use of synthetic organic compounds in industry and the natural occurrence of organic matter in the Earth's crust. Organic matter causes adverse effects on the performance of water treatment plants, such as accelerated bacterial growth and increased consumption of chemical coagulants. Organic matter also has the ability to chelate some heavy metals forming refractory complexes that resist coagulation. The current research combines electromagnetic radiation (microwave) and electrocoagulation technologies to purify water from OM-heavy metal complexes created using iron and ethylenediaminetetraacetic acid (EDTA). The organic matter-iron solution was introduced to a microwave field to break down the complex, followed by electrolysis of the solution using an aluminum-base EC cell. Microwave power (50.0-300.0 W), temperature (50.0-150.0 °C) and irradiation period (5.0-15.0 minutes) were measured. During the electrolysing stage, initial pH (4.0-8.0), current density (1.0-2.0 mA.cm⁻²) and space between electrodes (5.0-20.0 mm), were examined. The results showed that the electromagnetic radiation-electrocoagulation technology removed up to 92 % of the organic matter-iron complex in comparison to 69.6 % removal using a traditional electrocoagulation method. The best operational conditions were established as follows: 10 minutes of microwave irradiation at 100W at a temperature of 100 °C, followed by 20.0 minutes of electrolysing at an initial pH of 6.0, the space between electrodes 5.0 mm and current density of 1.50 mA.cm⁻².

Keywords: Microwave; electrocoagulation; water; organic matter; heavy metals.

35 1. Introduction

36 Freshwater is crucial for the existence of humankind on planet Earth, playing a major role in the socio-economic
37 development and sustainability of ecosystems (Luo et al., 2020). The majority of the problems humanity has faced
38 in the 21st century have been related to the availability of freshwater (Schwarzenbach et al., 2010; Yin et al., 2020).
39 Water is abundantly available on planet Earth, with more than $1.4 \times 10^8 \text{ m}^3$ water covering more than 70.0 % of
40 the planet, but less than 2.50 % of that water is freshwater suitable for human use, the majority (97.5 %) saline
41 (de Paul and Moore, 2018). 1.60 % of this freshwater is stored within ice and groundwater, leaving only 1.0
42 % ready for human use (Fogden and Wood, 2009; Kuokkanen, 2016). Unfortunately, this very small amount of
43 water is becoming polluted with a wide range of pollutants due to the substantial growth in the global population
44 and industrialisation (Li et al., 2019; Wijesiri et al., 2020). Hashim et al. (2019) found that the concentration of
45 some pollutants has increased in the aquatic environment by 75 % over the last century due to extensive industrial
46 activities and urbanisation. Among these pollutants, organic matter (OM), which contain carbon compounds, is a
47 major concern for water treatment plants due to its adverse effect on the performance of treatment units and the
48 aquatic ecosystem (Hashim, 2017; Pivokonsky et al., 2015). In terms of water treatability, there is evidence that
49 OM inhibits, or significantly minimises, the removal of heavy metals as it tends to chelate heavy metals, forming
50 stubborn complexes that resist coagulation (Gheraout et al., 2009; Pivokonsky et al., 2015). This is of significant
51 concern as heavy metals cannot be biodegraded and tend to accumulate in the tissues of living organisms causing
52 a range of diseases including cancer, Alzheimer and liver failure (Bazrafshan et al., 2015; Yu et al., 2019). OM
53 also maximises operating costs by increasing the consumption of coagulants, this minimising the efficiency of
54 disinfection units in water treatment plants. OM can also react with chlorine producing toxic by-products such as
55 bromodichloromethane and dibromochloromethane (Gheraout et al., 2009; Matilainen et al., 2010). There is a
56 range of innovative methods used to remove organic and inorganic pollutants from water (Pang et al., 2019; Wu
57 et al., 2019), such as the application of hybrid aerogels/perox-monosulfate to remove phenolic compounds, as
58 much as 90 % of p-nitrophenol within 60 minutes (Ren et al., 2018). Metal-organic frameworks have also been
59 used to develop hydrogels and other types of adsorbents (Wang et al., 2019; Wu et al., 2019). However, the cost
60 and skilled labour required to operate these applications have limited their use, especially in poor countries.
61 Interest has recently been to electrocoagulation (EC), which is the *in situ* production of coagulants by the
62 electrification of metal electrodes immersed in the solution being treated as it has several advantages including
63 cost-effectiveness, simplicity, and the need for less chemical additives (de la Fuente et al., 2019; Kuokkanen,
64 2016). EC technology has also proved to be efficient (Bazrafshan et al., 2015; Damaraju et al., 2020), the literature
65 indicating the sensitivity of EC to the presence of OM in water (Khandegar and Saroha, 2013; Mohora et al.,
66 2014). OM can cause more complex problems in EC plants though. It has been known to accelerate the formation
67 of a passive layer on electrodes, this minimising the production of coagulants, which in turn substantially reduces
68 the removal of pollutants and maximises power consumption (Khandegar and Saroha, 2013; Mohora et al., 2014).
69 Research has shown that EC units can minimise arsenic from 150.0 to 6.30 $\mu\text{g/L}$ (96.0 %) within 6 minutes of
70 treatment at a current density of 2.5 A/m^2 and an electrode area/volume water ratio of 0.24 cm^{-1} (Kobyta et al.,
71 2014). However, in the presence of 9.31 mgC/L of OM, the EC unit required 90 minutes and a current density of
72 57.80 A/m^2 to lower arsenic concentrations from 45.0 to 6.20 $\mu\text{g/L}$ (85.0 %) (Mohora et al., 2012).

73 The extensive use of organic compounds in industry and agriculture has intensified challenges for the water
74 treatment industry as large quantities of used OM found in industrial effluents, are discharged into freshwater

75 resources. Significant concentrations of OM can be found in the effluents of food, oil refinery, pharmaceutical,
76 and pesticide industries (Lapworth et al., 2012). Natural OM also enters freshwater resources as surface runoff
77 polluted with residuals of plants, animals, and humic substances (Gheraout et al., 2009; Pivokonsky et al., 2015).
78 The present investigation aims to solve one serious OM-related problem: the inhibition or minimisation of the
79 removal of heavy metals by electrocoagulation units. A novel microwave (MW) assisted-EC technology has been
80 used to remove heavy metals from water suffused with OM. MW is a form of electromagnetic waves with a
81 frequency range between 300 MHz and 300 GHz and a wavelength range of between 1m at 300 MHz and 1 mm
82 at 300 GHz (Wong and Gupta, 2015). The ability of MW to heat substances was accidentally discovered during
83 the Second World War by Percy LeBaron Spencer when the latter noticed that his candy had melted while he was
84 testing a radar magnetron (Bijanad et al., 2015; Hayes, 2002). Intensive investigations during the period 1960 to
85 1975 into microwave applications, led to the manufacture of smaller and affordable microwave ovens, by 1978,
86 the first laboratory microwave produced and marketed by CEM (Bassyouni et al., 2011; Wong and Gupta, 2015).
87 Recently, MW technology has attracted substantial interest as an efficient heating tool in chemical medical
88 applications because MW energy couples with the molecules of the substance being heated, meaning the heat is
89 generated from within the mixture without heat transfer. MW heat is independent of the thermal conductivity of
90 the container because it is generated through the direct absorption of MW energy by the targeted substance
91 (Bassyouni et al., 2011; Hayes, 2002). MW results in the rapid superheating of a substance, producing accelerated
92 chemical reactions, something which cannot be produced by traditional heating.

93 MW and EC technology have been used in the present study to develop a new hybrid treatment method able to
94 remove OM-iron complex from water. MW heating is used to decompose the OM-iron complex, which enables
95 the EC unit to remove the heavy metal. Coupling MW and EC technologies in one unit not only facilitates the
96 removal of refractory pollutant, but also helps to minimise the environmental impacts of the treatment process as
97 neither technology requires chemical additives to work, nor do they produce toxic secondary pollutants (Damaraju
98 et al., 2020; Hashim, 2017).

99 **2. Methods**

100 **2.1. Justification of choices**

101 Iron and ethylenediaminetetraacetic acid (EDTA), have been used in the current investigation as the heavy metal
102 and OM models. EDTA has been selected because it is used extensively by various manufacturing industries
103 including paint, electronics, pesticides, and photography, meaning that high concentrations of EDTA will be
104 discharged within their effluents (Babay et al., 2001; Huang et al., 2016). EDTA is also recognised as chelating
105 heavy metals in aquatic environments forming a refractory complex (Chen et al., 2012). Iron was selected as a
106 heavy metal model because it is found in abundance in the Earth's crust (approximately 5.0 % of the chemicals
107 in geological formations) and in the effluents of a wide range of industries, such as fertiliser and surface treatment
108 plants (Babay et al., 2001; Huang et al., 2016). Iron is found occurring in all forms of freshwater resources.
109 Elevated concentrations of both EDTA and iron can also be found together in the effluents of the printed circuit
110 board and herbicide industries (Huang et al., 2016; Lin et al., 2015). Consequently, the presence of EDTA-iron
111 complex is expected in freshwater resources, water treatment plants subject to the negative impacts of said
112 complex.

113 The selection of MW technology as an assistant method for the EC unit is based on the ability of MW irradiation
114 to induce the reaction rate and on the thermal and athermal effects of MW. Typically, a reaction starts with
115 reactants that have an amount of energy less than that required to start the transformation process, these reactants
116 needing extra energy to collide in the geometrically correct orientation and start the transformation (Hayes, 2002;
117 Yin et al., 2020). Once this additional energy, which must be absorbed from the reaction environment becomes
118 available, the reaction starts (Bassyouni et al., 2011). Usually, this energy is driven from external heat sources
119 such as MW or traditional heat sources but MW irradiation provides the required momentum to complete the
120 reaction without influencing the activation energy (Deng et al., 2020; Hayes, 2002). This MW heating process
121 will not decompose thermally unstable compounds that might be present in the reaction environment: the
122 enhancement, or acceleration of chemical reactions by MW irradiation, cannot be explained by rapid heating alone
123 (de la Hoz et al., 2005; Lin et al., 2009). Research investigating the influence of MW on different substances has
124 found that the acceleration in chemical reactions created by MW is a combination of thermal, high bulk
125 temperature and extremely hot microscopic spots and athermal, molecular motion and polarizing fields, effects
126 (Lin et al., 2009; Yang et al., 2014). Thermal effects are related to the dissipation of MW energy into heat as a
127 result of the direct absorption of MW energy by the substance being irradiated (Yu et al., 2010).

128 Athermal effects related to MW refers to any effect that is not associated with temperature increase, i.e. chemical,
129 physical or biochemical changes in the behavior of the substance being microwaved when the system temperature
130 remains constant (Yang et al., 2014; Yu et al., 2010). These effects originate from several sources such as an
131 increase in polarization of the dipolar transition state (Veitía and Ferroud, 2014), and the interaction between the
132 MW field and materials under treatment (de la Hoz et al., 2005). Athermal MW effects increase the vibration and
133 ‘wriggle’ of the polar chains of molecules that lead to the breaking, or weakening, of chemical bonds. This then
134 accelerates the reaction, enhancing the removal of organic and biological pollutants (Tyagi and Lo, 2012).

135 2.2. The EC cell

136 A 1,000 L, rectangular Perspex container was used to manufacture chemically inert and affordable
137 electrocoagulation (EC) (Hashim, 2017) (Figure 1(A)). The EC cell comprised seven perforated plates acting as
138 electrodes, made from aluminium because of its low potential and cost (Chaturvedi and Dave, 2012). The first
139 three electrodes were not supplied with electricity and used as water diffusers (mixers), while the remaining four
140 were supplied with DC power with a total effective area of 280 cm². The DC current was provided using a rectifier

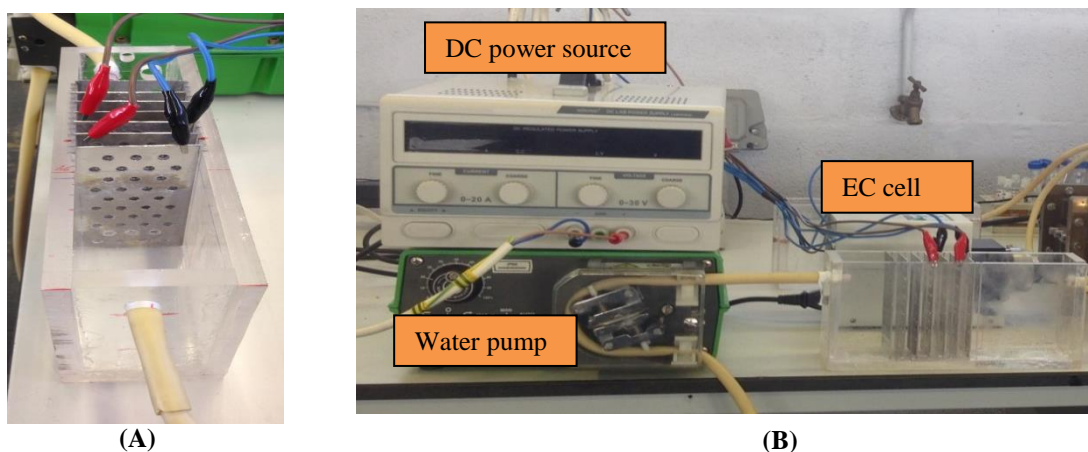


Figure 1: A) the EC cell, B) treatment system.

141 (H-Q Power; 0.0–30.0 Volts). A peristaltic water pump (Watson-Marlow 504U) was used to pump water through
142 the EC cell, as shown in Figure 1(B).

143 **2.3. Solutions**

144 A concentrated iron solution, 300 mg/L, was prepared by dissolving $\text{Fe}_2\text{SO}_4 \cdot 7\text{H}_2\text{O}$ in deionised water. 500 mL
145 samples with a lower concentration of iron, 20 mg/L, were diluted from this stock solution, OM then added to
146 each of these samples. Ethylenediaminetetraacetic acid (EDTA) was used as an organic matter because it has the
147 ability to chelate heavy metal ions to form complexes that resist traditional treatment methods (Chen et al., 2012;
148 Lin et al., 2015). It is also widely used in different industrial and domestic applications such as detergents,
149 painting, fertilisers, printed circuit boards, herbicides, and photography, and surface treatment industries.
150 Industrial effluents often contain EDTA complexes with different metals such as lead, arsenic, and iron (Babay et
151 al., 2001; Huang et al., 2016).

152 The amount of EDTA added to the synthetic water samples ranged from 50 to 500 mg/L, this matching samples
153 were used in other research (Hashim, 2017; Huang et al., 2016).

154 **2.4. Experimental work**

155 **2.4.1. EC experiments**

156 The first set of EC experiments were designed to study the influence of electrode spacing (SBE), current density,
157 and the initial pH on iron removal from water in the absence of EDTA. pH ranged between 4.0-8.0, SBE 5.0-20
158 mm and current density 1.0-2.0 $\text{mA}\cdot\text{cm}^{-2}$. The pH was manipulated using HCl or NaOH, the rest of the parameters
159 were manually adjusted to the required value. The experiments ran for 20 minutes, iron removal was calculated
160 at 5-minute intervals during the electrolysis process by taking 10.0 mL samples from the unit and filtering them
161 with a 0.450 μm filter (supplied by Sigma-Aldrich) to remove sludge (Phadke, 2014). The filtrates were then
162 tested for iron concentrations using an atomic absorption spectrophotometer (Model: ICE-3300).

163 In the second set of experiments, EDTA-iron complex solutions were electrolysed using the same EC cell. To
164 compare iron removal efficiency with and without EDTA, the electrolysis process was carried out using the best
165 operating conditions as established by the previous set of experiments.

166 **2.4.2. MW-EC experiments**

167 Finally, the new method, the MW assisted-EC method, was applied in two steps: MW irradiation followed by
168 the electrolysis process. 500 mL water samples containing EDTA-iron complex, were subject to the MW field
169 at different powers (50 to 300 W), temperatures (50 to 150 °C), and irradiation time (5 to 15 minutes). The
170 irradiation process was carried out using a lab-scale CEM device (model: Discovery – SP W/ACTIVENT). During
171 the microwave process, vials were firmly closed using plastic caps to avoid any spillage. The maximum allowable
172 (emergency) pressure during sample irradiation was set at 120 PSI. After each run, the irradiated sample was left
173 outside to cool down to 20 ± 1 °C, then electrolysed under the best conditions established by the previous sets of
174 experiments.

175 It is noteworthy to highlight that the MW field has been used due to its unique merits; its energy couples up with
176 the molecules of the substance being heated, which means heat is produced within the mixture without heat

177 transfer (Bassyouni et al., 2011; Hayes, 2002). MW heat is independent of the thermal conductivity of the
 178 container because it is generated through the direct absorption of MW energy by the targeted substance (Wong
 179 and Gupta, 2015). Another unique and effective heating phenomenon takes place during MW heating: hotspots.
 180 The temperature in these microscopic spots, 0.1 mm in size, is about 100 – 200 °C higher than the bulk temperature
 181 (de la Hoz et al., 2005). Therefore, MW irradiation results in rapid superheating of a substance producing an
 182 enormous acceleration in a range of chemical reactions. This cannot be produced by traditional heating methods.

183 2.4.3. Economic analysis

184 A preliminary operating cost of the MW assisted-EC method has been calculated to provide some indication of
 185 the economic efficiency of this method. For bench-scale treatment units, this estimate only covers the cost of
 186 power and materials (Chopra and Sharma, 2012; Hashim et al., 2017a). The estimate was calculated as follows:

$$187 \text{ Operating cost} = \varphi_1 \times C_{\text{power}} + \varphi_2 \times C_{\text{electrodes}} + \varphi_3 \times C_{\text{chemicals}} \quad (1)$$

188 where C_{power} (kWh/m³) is power, $C_{\text{electrodes}}$ (kg/m³) the electrodes material and $C_{\text{chemicals}}$ (kg/m³) the
 189 chemicals. φ_1 , φ_2 and φ_3 represent the unit price of power, electrodes material, and chemicals.

190 Consumption of power and electrodes material was calculated as follows (Chopra and Sharma, 2012; Hashim et
 191 al., 2017a):

$$192 C_{\text{power}} \text{ (kWh/m}^3\text{)} = \frac{\text{applied electric current (ampere)} \times \text{cell potential (volt)} \times \text{treatment time (hour)}}{1000 \times \text{volume of solution (m}^3\text{)}} \quad (2)$$

$$193 C_{\text{material}} \left(\frac{\text{g}}{\text{m}^3}\right) = \frac{\text{applied electric current (ampere)} \times \text{treatment time (second)} \times \text{molecular weight (g/mol)}}{\text{number of electrons} \times \text{Faraday's constant} \times \text{volume of solution (m}^3\text{)}} \quad (3)$$

194 Quantities of Fe₂SO₄·7H₂O, EDTA, HCl, NaHO, and NaCl were manually measured using a 3-digit accuracy
 195 scale (KERN: PCB-A02). The unit prices of these were estimated according to Iraqi markets in 2020, which were
 196 2.5 cent/kWh of electricity, 1.53 \$/kg of aluminium, 0.32 \$/L of HCl, 0.4 \$/kg of NaOH, and 0.38 \$/kg of NaCl.

197 It should be noted that Eq.1 is not applicable for field-scale treatment units as it does not cover the costs of labour,
 198 maintenance, chemical storage, and sludge management (Chopra and Sharma, 2012).

199 2.4.4. Comparisons between traditional and MW heating

200 Heating using MW is completely different from traditional heating processes as MW energy couples with the
 201 molecules of the substance being heated, the heat generated from within the mixture without heat transfer
 202 (Bassyouni et al., 2011; Hayes, 2002). MW heat is independent of the thermal conductivity of the container
 203 because it is generated through the direct absorption of MW energy by the targeted substance (Wong and Gupta,
 204 2015). MW irradiation results in rapid superheating that produces accelerated chemical reactions, which cannot
 205 be produced by traditional heating making it faster and more efficient than traditional heating processes.

206 Athermal MW effects and the generation of hot spots within the solution being irradiated, contribute to the
 207 decomposition and removal of pollutants from aqueous media (Horikoshi and Serpone, 2014; Yang et al., 2014).

208 To investigate which method of heating is more efficient when removing iron from water in the presence of OM,
209 both were used to heat two groups of water samples containing OM-iron complexes with identical characteristics,
210 to 100 °C for 10 min. The heated samples were left at room temperature to cool down to 20 ± 1 °C before
211 electrolysing them at the same optimum operating conditions obtained in previous experiments.

212 3. Results and discussion

213 3.1. EC experiments

214 This section presents and discusses the results obtained from the EC treatment, including the effect of initial pH,
215 current density, SBE, and OM on iron removal. Initially, the ability of the aluminium-based EC reactor to remove
216 iron ions without organic matter. Then, the effect of organic matter on iron removal was investigated.

217 3.1.1. Iron removal without organic matter

218 The EC experiments were initiated by examining the ability of the aluminium-based EC reactor to remove iron
219 ions from solutions. To attain the best removal efficiency, the effects of key parameters, including the initial pH,
220 SEB, and current density, on the removal of iron were examined.

221 3.1.1.1. Effects of initial pH

222 In order to examine the influence of initial pH on iron removal from water, a series of batch experiments were
223 carried out changing the initial pH in iron solutions (20.0 mg/L of iron) from 4.0 to 8.0. Electrolysis was
224 applied for 20 minutes at a current density of $1.50 \text{ mA}\cdot\text{cm}^{-2}$ and SBE of 5.0 mm.

225 The results indicated that the best iron removal efficiency occurred in slightly acidic pH (between 6 and 7), which
226 is shown in Figure 2. Iron removal efficiency started to increase from 90.1 % to 99 % when the initial pH rose
227 from 4.0 to 6.0 and slightly decreased to 98.9 % when the initial pH reached 7.0. When the initial pH rose to more
228 than 7.0, iron removal decreased. This could be attributed to the predominance of $\text{Al}(\text{OH})_3$ aluminium hydroxides
229 that have a high adsorption capacity for iron ions in natural and slightly acidic ranges, while in acidic and basic
230 ranges, the adsorption capacity of prevailing aluminium-hydroxides for iron is insignificant (Emamjomeh and
231 Sivakumar, 2009; Ghosh et al., 2008).

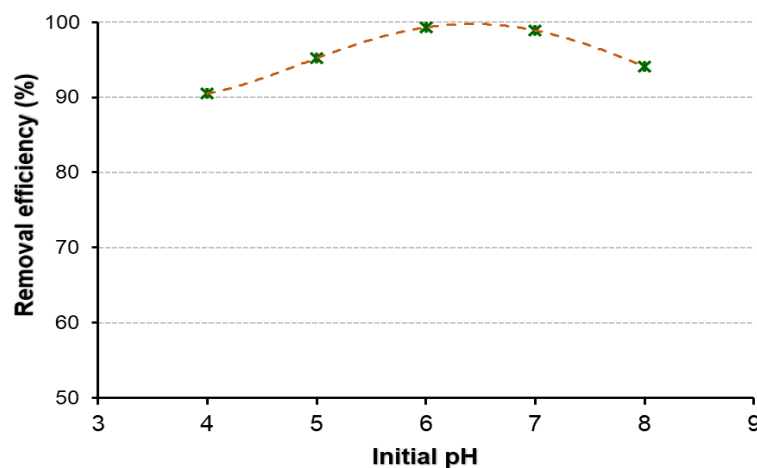


Figure 2: Influence of initial pH on the removal of iron from water.

232 Based on this, the remainder of the experiments were carried out at an initial pH of 6.

233 **3.1.1.2. Influence of current density**

234 Sets of batch experiments were carried out to examine the influence of current density on iron removal from
235 synthetic water samples. Three current densities, 1.0, 1.50, and 2.0 mA.cm⁻², were applied for 20 minutes to treat
236 water samples with 20.0 mg/L of iron at SBE of 5.0 mm and initial pH of 6.0.

237 Figure 3 illustrates iron removal efficiency against electrolysis time for the current densities as above, showing
238 that iron removal is very sensitive to changes in current density. For example, it was noticed that within the first
239 15 minutes of EC treatment, the iron removal rose from 87.70 % to 98.10 %, and 99.10 % as the current density
240 rose from 1.0 to 1.50, and 2.0 mA/cm². After 20 minutes, as much as 99.6 % of the iron was removed at a current
241 density of 1.50 mA.cm⁻², but it decreased to around 93.0 % at a current density of 1.0 mA.cm⁻². This behaviour
242 could be explained by the fact that the higher the current density, the more aluminium ions dissolve from the
243 anode (Vasudevan et al., 2010), which in turn enhances floc formation and iron removal. In terms of power
244 consumption, it was noticed that maximising current density negatively affected the performance of the EC cell:
245 the higher the current density, the higher the power consumption. For instance, power consumption significantly
246 increased from approximately 1.3 to 6.7 kWh/m³ when the current density rose from 1.0 to 2.0 mA/cm². Taking
247 the negative and positive influences of current density on the performance of the EC cell into account, the
248 remainder of the experiments will use a current density of 1.50 mA/cm².

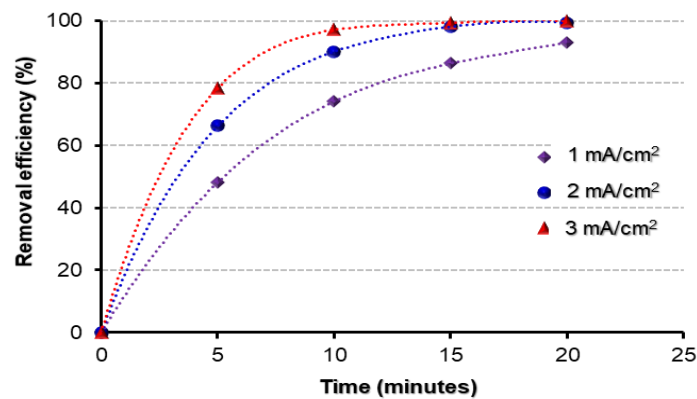


Figure 3: Iron removal at different current densities.

249 **3.1.1.3. Influence of SBE**

250 The influence of SBE on the removal of iron by the aluminium-based EC cell was explored by carrying out a
251 series of batch experiments, using 500 mL samples of synthetic water containing 20.0 mg/L of iron, at different
252 SBE's: 5, 10, and 20 mm. The EC treatment was carried out for 20 minutes at a current density of 1.50 mA.cm⁻²
253 and initial pH of 6.0.

254 The results (Figure 4) revealed that iron removal is inversely proportional to SBE, in that the residual
255 concentrations of iron dropped from 20.0 mg/L to 0.06, 0.5, and 1.05 mg/L as the SBE increased from 5.0 to 10.0
256 and 20.0 mm. This is explained by the formation of a very thin layer of metal hydroxides on the anode at the
257 wider SBE which minimises production coagulants, decreasing removal efficiency (Ghosh et al., 2008; Hashim
258 et al., 2017b). The effects of this metallic layer could be minimised by decreasing the SBE as the evolution of
259 hydrogen gas from the cathodes, removes the accumulated metal hydroxides on the anode surfaces (Hashim et al.,
260 2017b).

261 There was a significant increase in power consumption as SBE increased, increasing from 3.3 to 7.3 kWh/m³
 262 when the SBE expanded from 5.0 to 20.0 mm. An SBE of 5.0 mm was used therefore for the remaining EC
 263 experiments.

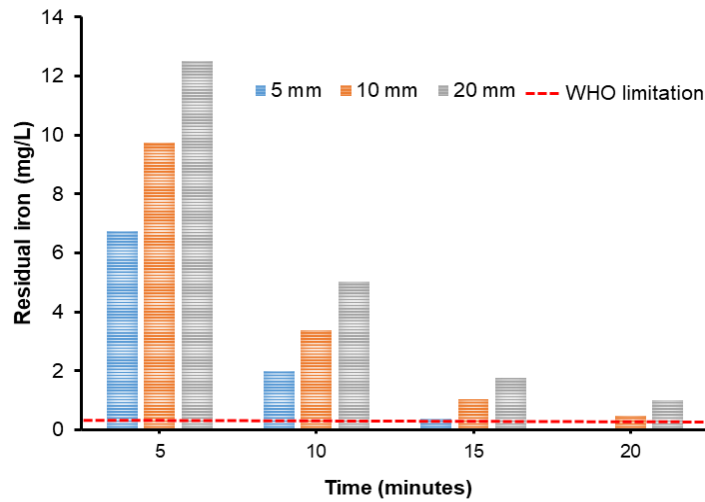


Figure 4: Iron removal at different SBE.

264 3.1.2. The effect of OM on iron removal using the EC method

265 In order to investigate the impact of OM on the removal of iron from water, synthetic water samples containing
 266 EDTA at concentrations ranging from 50 to 500 mg/L, were electrolysed under the best operating conditions as
 267 established in the previous iron removal experiments. It can be clearly observed from Figure 5 that the removal
 268 of iron sharply decreased from 99.6 % to 79.8 %, 69.6 % and 59.5 % as the concentrations of EDTA increased
 269 from 0 to, 50, 100, and 200 mg/L. Adding 500 mg/L of EDTA was enough to disable the iron removal process,
 270 iron removal plateauing close to 25 %.

271 The negative influence of EDTA on the removal of iron could be explained by the ability of EDTA to chelate
 272 iron, forming a complex that is difficult to remove (Chen et al., 2012; Lin et al., 2015), and its ability as an OM,
 273 to compete for the available active sites on flocs, decreasing the removal of the targeted pollutants (Matilainen et
 274 al., 2010; Mohora et al., 2014). OM also promotes the development of a passive layer on the anode's surface that

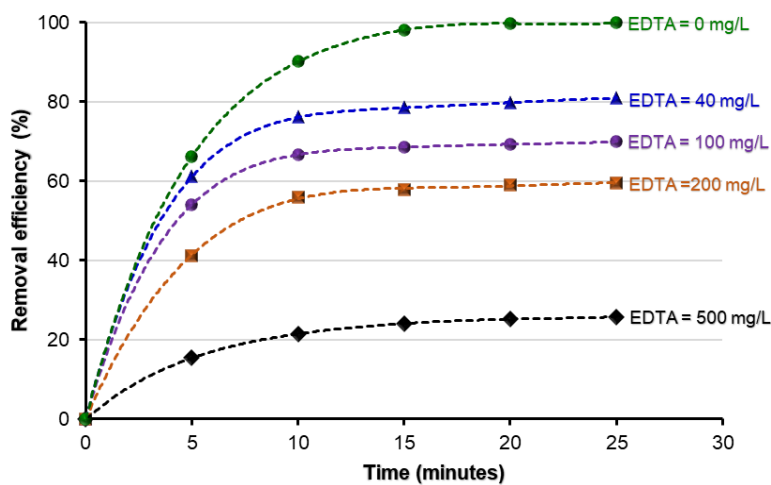


Figure 5: The influence of OM (EDTA) on the removal of iron from water by EC method.

275 decreases the number of liberated ions, consequently decreasing removal efficiency (Khandegar and Saroha, 2013;
276 Mohora et al., 2014).

277 3.2. Application of the MW assisted-EC method to remove iron from water in the presence of OM

278 EDTA-iron complex removal using this hybrid method was carried out in two steps: MW irradiation followed by
279 the electrolysing process. Initially, 500 mL of EDTA-iron complex solution was subjected to an MW field at
280 different powers (50 to 300 W), temperatures (50 to 150 °C), and irradiation times (5 to 15 minutes). The
281 microwaved solutions were then electrolysed at the operating conditions established in the previous experiments.

282 3.2.1. Influence of MW power

283 The influence of MW power on the removal of iron from the water was investigated by irradiating 500 mL of
284 EDTA-iron complex-containing synthetic water samples for 10 minutes at 50, 100, and 300 W, keeping the
285 temperature constant at 50 °C. The irradiated samples were then kept at room temperature to cool down to 20±1
286 °C, electrolysed using the aluminium-based EC cell under the conditions established by the preceding
287 experiments. The results indicated that the performance of the EC cell was enhanced, in terms of iron removal, as
288 MW power increased. It can be seen from Figure 6(A) that microwaving the synthetic samples for 10 minutes at
289 50, 100, and 300W, increased iron removal from 69 % (non-microwaved) to 71.1 %, 74.3 %, and 76.2 %. This
290 can be explained as higher MW power creating rapid heating, this inducing fast molecular motions of ions polar
291 and clusters (Yang et al., 2014). The MW field, specifically the hotspots, quickly provide the required temperature
292 to break chemical bonds without influencing the activation energy (Hayes, 2002). Although, it has been found
293 that the higher the MW power the better the iron removal, increasing the MW power negatively influenced energy
294 consumption. Figure 6(B) shows that increasing MW power from 50 to 100 and 300 W, dramatically increased
295 power consumption from 0.01 to 0.018 and 0.05 kWh. As such, an MW power of 100W was used to run the
296 remaining experiments.

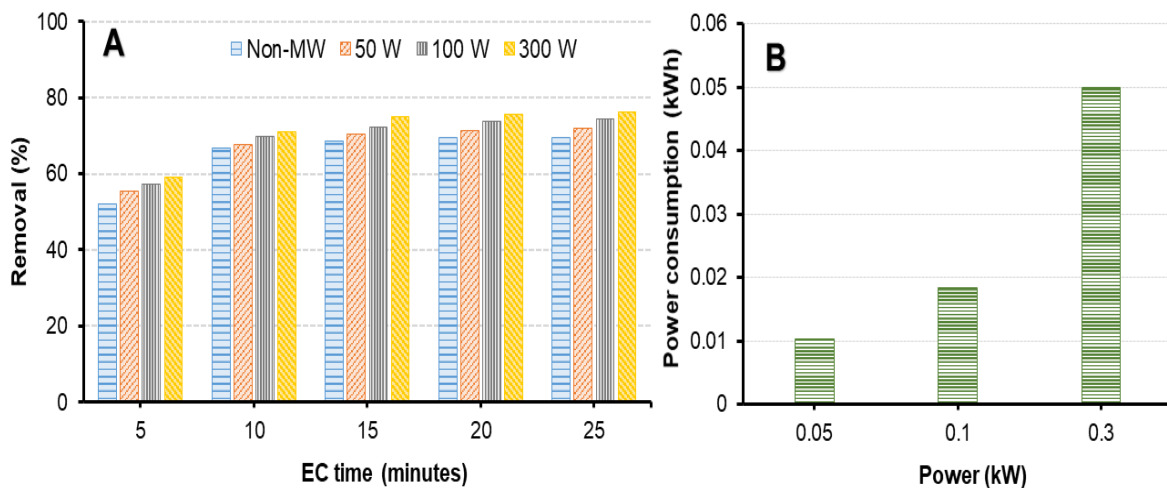


Figure 6: Influence of MW power on: (A) iron removal using EC method, (B) power consumption.

297 3.2.2. Influence of MW temperature

298 In order to examine the influence of MW heating on the performance of the EC method in terms of iron removal,
299 sets of 500 mL of EDTA-iron complex-containing synthetic water samples were microwaved for 10 minutes at

300 different temperatures (50, 100, and 150 °C), keeping the MW power constant at 100W. These samples were then
301 left at room temperature to cool down to 20 ± 1 °C, and electrolysed at the optimum conditions for iron removal.

302 Figure 7(A) shows that the EC method performed better with an increase in MW temperature. The rate of iron
303 removal increased from 69.6 % (for non-microwaved samples) to 74.3 %, 92 %, and 95.6 % as the microwaving
304 temperature increased from 50 to 100 and 150 °C. This increase in iron removal can mainly be attributed to the
305 unique thermal effects of the MW field. If the MW temperature increases, MW energy interacting with the
306 molecules of the substance being heated and the temperature of the hotspots, will also increase. Therefore, rapid
307 superheating of the substance takes place, the decomposition of the EDTA-iron complex likewise increasing as
308 does iron removal.

309 However, the results show that increasing MW temperature increases the length of time of application, this
310 negatively influencing the operational cost of the unit. Figure 7(B) illustrates that raising the temperature of the
311 solution from 50 to 100 and 150 °C, increased power consumption from 0.018 to 0.023 and 0.03 kWh. Therefore,
312 an MW temperature of 100 °C will be used to perform the remainder of the experiments.

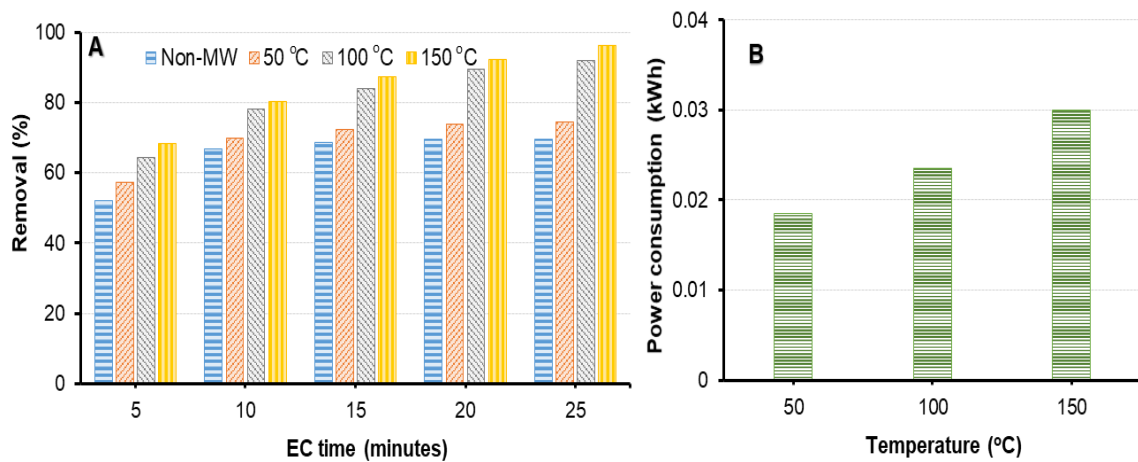


Figure 7: Influence of MW temperature on: (A) Iron removal using EC method, (B) power consumption.

313 3.2.3. Influence of MW duration

314 In order to examine the effects of irradiation time on iron removal, a series of batch experiments were conducted
315 by microwaving 500 mL of EDTA-iron complex- containing synthetic water samples for different lengths of
316 time: 5.0, 10.0, and 15.0 minutes. The microwaving process was carried out at a constant MW power of 100W and
317 a constant temperature of 100 °C.

318 Figure 8(A) shows that iron removal efficiency was enhanced with an increase in microwave time: the removal
319 of iron rose from 83 % to 92 % and 93.2 % as the microwaving time increased from 5.0 to 10.0 and 15.0 min. The
320 explanation for this is that longer irradiation time allows for the transfer of more heat to the solution being
321 microwaved (Yang et al., 2014). However, Figure 8(B) shows that increasing microwave time significantly

322 increases power consumption: increasing the microwaving time from 5.0 to 10.0 and 15.0 minutes, increased
323 power consumption from 0.015 to 0.023, and 0.032 kWh.

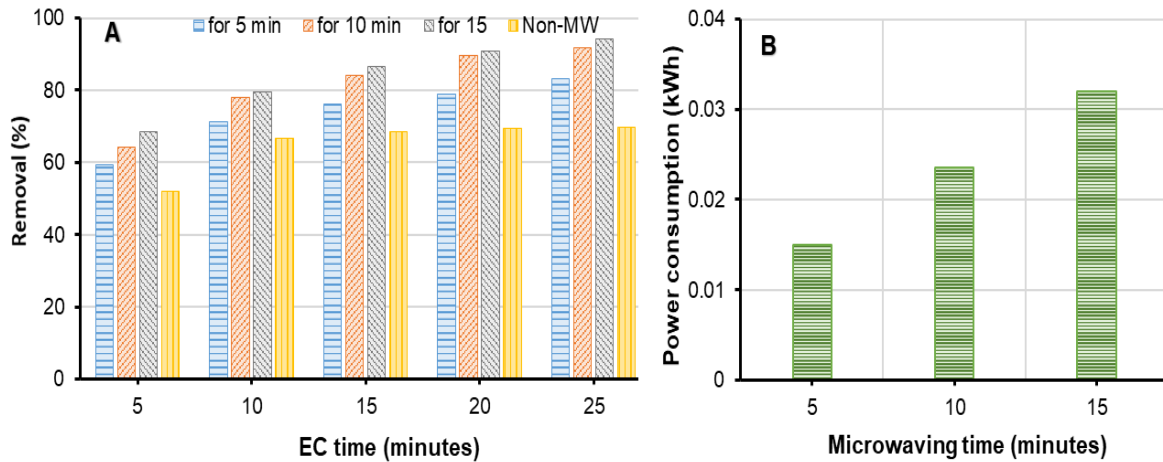


Figure 8: Influence of MW time on: (A) iron removal, (B) power consumption.

324 The outcomes of the present study have evidenced that the application of an MW field could significantly enhance
325 the removal of heavy metals using the EC method even when elevated concentrations of OM present in the water
326 are treated. This enhancement can mainly be attributed to the thermal and athermal effects of the MW field on the
327 stability of EDTA-iron complexes. Raising the temperature of water to 100 °C, results in the generation of hotspots
328 within the solution, the temperature in these microscopic spots (0.1 mm in size), is about 100 – 200 °C higher than
329 the bulk temperature (de la Hoz et al., 2005). The temperature of hot spots in the present study ranged between
330 200 and 300 °C. Within this range of temperature, the OM (EDTA) degraded into simple acids, such as
331 dimethylamine, and carbon dioxide (Chen et al., 2006; Motekaitis et al., 1982), releasing iron ions into the solution
332 which were easily removed by the EC unit.

333 In summary, the results of the present study have evidenced the ability of the MW assisted-EC method for the
334 removal of OM-metal complexes from aqueous media, which makes it a promising alternative to treat the effluents
335 of industries that use large amounts of heavy metals and OM

336 3.2.4. Economic analysis

337 The preliminary operating cost of the MW assisted-EC method was calculated at the optimum operating
338 conditions for OM-iron complex removal, which were 10 minutes of microwaving, 20 minutes of electrolysis at
339 a current density of 1.5 mA/cm², initial pH of 6, a gap between electrodes of 5 mm and water temperature of 20
340 °C. According to equations 1-3, the minimum operating cost was 0.679 USA \$/m³.

341 Although this is slightly higher than that of some more conventional treatment methods such as biological methods
342 (Yehya et al., 2014), it is still cheaper than many other advanced methods such as membrane filtration (0.94 \$/m³)
343 and reverse osmosis (0.63 \$/m³) for power alone (Alzahrani and Mohammad, 2014; Sagle and Freeman, 2004).
344 More importantly, the new MW assisted-EC method efficiently removes refractory heavy metal-OM complex,
345 something which cannot be achieved using conventional methods. This method also possesses many attractive
346 advantages: it is simple to install and operate, it is safe as it requires no chemical handling and it requires a
347 relatively small area.

348 **3.2.5. Comparison between traditional and MW heating**

349 Traditional and MW heating methods were used to heat two groups of OM-iron complex-containing water
350 samples with identical characteristics, to 100 °C for 10 mins. They were cooled down to 20±1 °C before
351 electrolysing.

352 The results showed that MW heating was superior to traditional heating in terms of iron removal and power
353 consumption. Figure 9(A) shows that the iron removal from the microwaved samples was 11 % better than for
354 traditionally heated samples. This can be explained by the unique characteristics of the MW heating process.
355 Firstly, MW energy couples with the molecules of the substance being heated, the heat generated from within the
356 mixture without heat transfer (Bassyouni et al., 2011; Hayes, 2002). Secondly, MW heat is independent of the
357 thermal conductivity of the container because it is generated through the direct absorption of MW energy by the
358 targeted substance (Wong and Gupta, 2015). Thirdly, the formation of super-hot microscopic spots (hotspots)
359 within the solution being microwaved produces accelerated chemical reactions that cannot be produced by
360 traditional heating.

361 Figure 9(B) shows the significant difference between MW and traditional heating methods in terms of power
362 consumption. MW heating requires about 0.33 kWh for 10 minutes at 100 W, to increase the temperature of water
363 to 100 °C. Traditional heating requires 0.675 kWh, 2 times the power consumed by MW. This difference in power
364 consumption is mainly due to the difference between MW and traditional heating mechanisms. Traditional heating
365 processes transfer heat from an external source, passing through the walls of the container, to the targeted
366 substance (Wong and Gupta, 2015). This makes the temperature of the container higher than that of the substance
367 until they attain a state of equilibrium, which in turn maximises power consumption. MW heat, as mentioned
368 above, is generated through the direct absorption of MW energy by the targeted substance, MW heat coupling
369 with the molecules of the substance being heated.

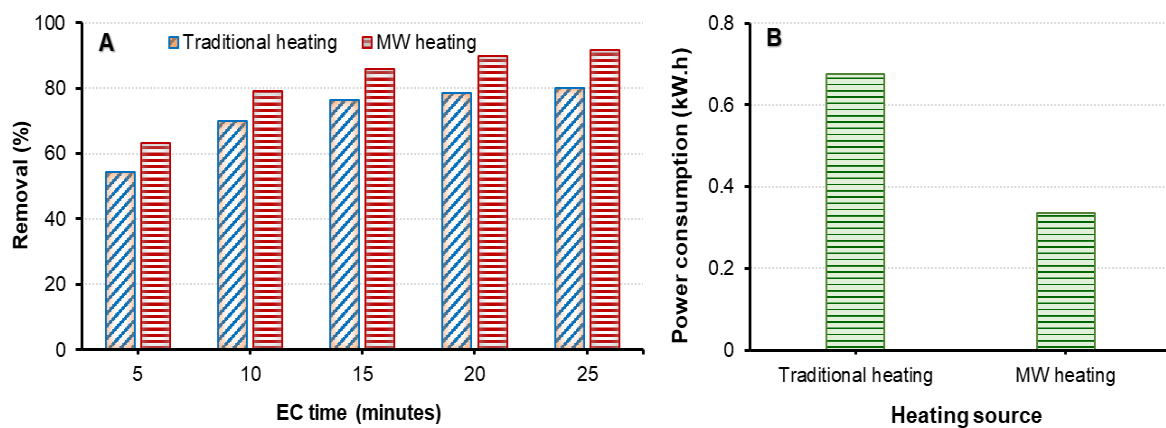


Figure 9: Comparison between MW and traditional heating in terms of: (A) Iron removal, (B) power consumption.

370 **4. Conclusions**

371 The results obtained from the present study proved that the presence of OM in aqueous media significantly
372 minimises the ability of EC units to remove heavy metals due to the ability of OM to chelate heavy metals forming

373 a stable complex and because EDTA accelerates the growth of passive anodic films. The results also confirmed
374 that the application of an MW field before the EC process is beneficial due to the ability of the MW field to
375 superheat the solution leading to thermal decomposition of OM into simple products, releasing chelated metal
376 ions, the latter easily removed by the EC unit. In terms of operating conditions, the following key facts can be
377 concluded:

- 378 1. The higher the current density, the more rapid the removal of pollutants, this due to the more rapid
379 liberation of aluminium coagulants from the sacrificial anode.
- 380 2. Increasing the distance between electrodes maximises the resistance for the flow of current in the EC,
381 this minimising the removal of targeted pollutants.
- 382 3. EDTA-iron complex removal is proportional to the MW power applied as higher MW power leads to the
383 transfer of more energy to the solution being microwaved.
- 384 4. Pollutant removal could be enhanced by increasing irradiation time, as this allows the generation of more
385 heat within the targeted solution.
- 386 5. Temperature levels during the MW process, play a central role in the removal of pollutants due to an
387 increase in the coupling of the MW energy with the molecules of the substance being heated.

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