Electrochemical activation of peroxides for treatment of contaminated

water with landfill leachate: Efficacy, toxicity and biodegradability

evaluation

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Abstract

Contaminated water with landfill leachate (CWLL) with high salinity and high organic content (total organic carbon (TOC) = 649 mg/L and Chemical Oxygen Demand (COD) = 1175 mg/L) is a toxic and non-biodegradable effluent. The present research aimed to assess the treatment effectiveness of CWLL by electrocoagulation (EC)/oxidant process. The ferrous ions generated during the process were employed as coagulant and catalyst for the activation of different oxidants such as peroxymonosulfate (PMS), peroxydisulfate (PDS), hydrogen peroxide (HP), and percarbonate (PC) to decrease TOC in CWLL. Removal of ammonia, color, phosphorous, and chemical oxygen demand (COD) from CWLL effluent was explored at various processes. EC/HP had the best performance (~73%) in mineralization of organic pollutants compared to others under the condition of pH 6.8, applied current of 200 mA, oxidant dosage of 6 mM, and time of 80 min. The oxidation priority was to follow this order: EC/HP > EC/PMS > EC/PDS > EC/PC. These processes enhanced the biodegradability of CWLL based on the average oxidation state and biochemical oxygen demand (BOD)/COD indices. $SUVA_{254}$ and E_2/E_3 indices were also investigated on obtained effluents. The phytotoxicity evaluation was carried out based on the germination index, indicating that the electro-activated oxidant was an effective system to reduce the toxicity of polluted solutions. EC/HP showed supremacy compared to others in terms of efficiency, cost, and detoxification. Therefore, the electro-activated oxidant system is a good means for removing organic pollutants from real wastewater.

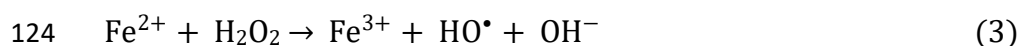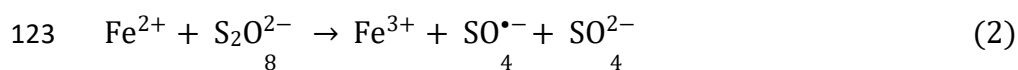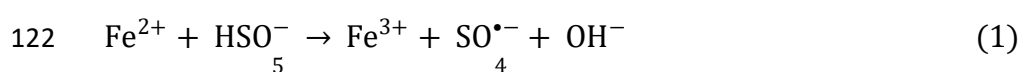**Keywords:** Landfill leachate; Electrochemical oxidation; Persulfate; Percarbonate; Hydrogen peroxide; Detoxification.

1. Introduction

Consumption produces solid and liquid wastes, and when made at massive scales —like in large cities— solid waste management constitutes a very problematic issue in terms of its disposal and recycling (Scott et al., 2019). Landfilling is the most common method for solid waste management in developing countries. In such countries, large volumes of solid wastes are sometimes brought to dump with no separation or adopting any other processing measures. There is no standard landfilling, and this can be the primary source of water and soil pollution (Ghanbari et al., 2020a). Conversely, many developed countries nowadays are using sanitary landfills as the standard yet lower-cost disposal mechanism for getting rid of the great bulk of rubbish (Antony et al., 2020). Penetration of rainfall or other water sources into the landfill wastes makes the pollutants extract, mixed with water, and form a highly contaminating effluent called leachate. Here a significant threat to the environment begins; as the leachate moves from the dumping site to the adjacent soils and water resources, when not treated (Antony et al., 2020). The leachate so developed, due to its highly complicated composition —numerous organic and inorganic compounds—constitutes a difficult challenge in terms of management of leachate treatment and containing the threats it poses for the water-soil environment (Brennan et al., 2016; Ye et al., 2019). Landfill leachate can contaminate groundwater and surface water. Contaminated water with landfill leachate (CWLL) is a severe problem in developing countries where landfill leachate management is ignored. CWLL should be treated since it poses the same characterizations as landfill leachate. As was said before, the selection of the leachate treatment process requires scrutiny due to different criteria such as the goal of treatment, composition, cost, environmental impact, effluent reuse, and so on. Traditionally, such high concentrated leachate, comprised of various organic materials including refractory compounds and numerous organic and inorganic toxic substances, is treated using sequential steps of biological, chemical, and

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80   physical processes (Renou et al., 2008). Slight biodegradability and toxic nature make the

81   researchers follow chemical procedures for the treatment of leachate. Accordingly, several

82   chemical processes are in use worldwide for the treatment of landfill leachate through

83   eliminating organic compounds and more excellent biodegradability (Li et al., 2019b;

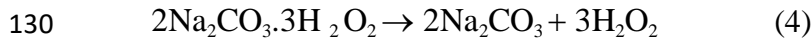84   Ghanbari et al., 2020a).

85   But the chemical approach of the treatment itself includes different processes, two

86   prominent of them being separation and oxidative processes. These two processes include

87   coagulation, flocculation, adsorption, filtration (Khataee et al., 2013), and degradation of

88   organic contaminants into tiny molecules or mineral compounds (Yekan Motlagh et al., 2019;

89   Ghanbari et al., 2020b). The physicochemical process in leachate treatment has its

90   drawback—that is, it leads to contaminant phase transfer rather than leading to mineralization

91   (Cortez et al., 2010). Further, the traditional physicochemical or biological methods of

92   leachate treatment are unable to provide the purification level as covered by the high

93   discharge standards that are obtained by many developed countries. Hence, the necessity of

94   thinking of novel replacement methods is obvious (Antony et al., 2020). One of the new

95   approaches to wastewater treatment is electrochemical processes (EPs), capable of attaining

96   higher efficiencies. The EPs are currently implemented based on separation and oxidation

97   mechanisms—aimed at industrial effluent treatment (Garcia-Segura et al., 2018). As one sub-

98   EP technology, the electrocoagulation (EC) method performs flexibly and effectively for

99   industrial wastewater treatment; here, the iron and aluminum electrodes lead to the

100   coagulation reactions, and coagulant agents are produced in the electrochemical reactor

101   (Mroczek et al., 2019; Ghanbari et al., 2020a). Advanced oxidation processes (AOPs) among

102   the destructive processes rely on producing highly reactive oxygen species (ROS) like

103   hydroxyl ($^{\bullet}OH$) and sulfate ($SO_4^{\bullet-}$) radicals that are non-selective and very reactive,

104   destructing the vast part of organic molecules (Burgos-Castillo et al., 2018; Eghbali et al.,

4

105 2019; Khataee et al., 2019). To produce ROS, different chemical oxidants like hydrogen

106 peroxide (HP), peroxymonosulfate (PMS), peroxydisulfate (PDS) anions, and percarbonate

107 (PC) can be used as precursors (Li et al., 2018; Eslami et al., 2020; Hassani et al., 2020).

108 According to the literature, the PMS, PDS, PC, and HP are chemical oxidants potentially

109 promising for the treatment industry through their activity, induced by transition metals,

110 ultraviolet (UV), heat, carbon-catalysts, microwave radiation, etc. (Ma et al., 2018; Hassani et

111 al., 2020). To combine AOPs and EC, chemical oxidants could be added to the EC reactor

112 when the iron electrode is applied. Accordingly, ferrous ions released from the iron electrode

113 play both coagulant and activator roles. Hence, EC/oxidant benefits both coagulation and

114 oxidation mechanisms. As previously mentioned, PMS, PDS and HP can react with ferrous

115 ions, increasing the kinetic of a generation of free radicals ($SO_4^{\bullet-}$ and $^\bullet OH$) (Liu et al., 2019;

116 Solís et al., 2020). Sulfate radicals are products of the activation of some oxidants like PMS (

117 HSO_5^-) and PDS ($S_2O_8^{2-}$) in the presence of ferrous ions as represented by the **Eqs. (1-2)**

118 while hydroxyl radicals are generated according to the Fenton reaction **(Eq. 3)**. However, in

119 PMS and PDS-based processes, hydroxyl radicals can be produced from the redox reaction

120 between OH^-/H_2O and sulfate radicals. At pH < 9, sulfate radicals are the dominant radicals

121 while at pH > 9, hydroxyl radicals are dominated (Giannakis et al., 2021).

122 $$Fe^{2+} + HSO_5^- \rightarrow Fe^{3+} + SO_4^{\bullet-} + OH^- \qquad (1)$$

123 $$Fe^{2+} + S_2O_8^{2-} \rightarrow Fe^{3+} + SO_4^{\bullet-} + SO_4^{2-} \qquad (2)$$

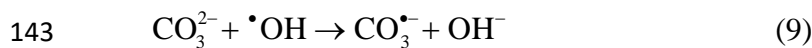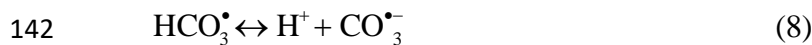124 $$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO^\bullet + OH^- \qquad (3)$$

125 Sodium percarbonate ($2Na_2CO_3 \cdot 3H_2O_2$) is an alternative oxidant to promote the

126 generation of reactive oxygen species. PC, which is a solid substance, acts like hydrogen

127 peroxide **(Eq. 4)**, having identical characteristics of this, but with additional advantages, i.e.,

128 safer handling as its solid-state and more environmentally friendly processes (Rivas et al.,

129 2010).

130 $$2Na_2CO_3.3H_2O_2 \rightarrow 2Na_2CO_3 + 3H_2O_2 \qquad (4)$$

131 $$CO_3^{2-} + H_2O \rightarrow HCO_3^- + OH^- \qquad (5)$$

132 $$HCO_3^- \leftrightarrow CO_3^{2-} + H^+ \qquad (6)$$

133

134   The recent decade has witnessed the development of PC as an oxidant to degrade the

135 organic contaminants in the presence of transition metal like Fenton oxidation (Sablas et al.,

136 2020; Yue et al., 2020). Although it has been reported that (bi)carbonate radicals may

137 contribute to the degradation of organic compounds in PC activation (**Eqs. 7-9**), its reactivity

138 is less than $HO^\bullet$. Moreover, it is known that bicarbonate anions can involve in reaction with

139 HP to produce peroxymonocarbonate ions (HCO_4^-) as shown by **Eq. (10)** (Regino and

140 Richardson, 2007).

141 $$HCO_3^- + {}^\bullet OH \rightarrow HCO_3^\bullet + OH^- \qquad (7)$$

142 $$HCO_3^\bullet \leftrightarrow H^+ + CO_3^{\bullet-} \qquad (8)$$

143 $$CO_3^{2-} + {}^\bullet OH \rightarrow CO_3^{\bullet-} + OH^- \qquad (9)$$

144 $$HCO_3^- + H_2O_2 \rightarrow HCO_4^- + H_2O \qquad (10)$$

145   Although the EC technique has been often used for landfill leachate treatment, to the best

146 of our knowledge, no comparative study has been carried out on the function of ferrous ions

147 in the electro-activated oxidants for contaminated water with landfill leachate (CWLL) so far.

148   Herein, the objectives of this study were as follows: (1) to evaluate the EC/oxidant

149 performance for Total Organic Carbon (TOC) reduction from CWLL with a focus on

150 parameters of the reaction like the oxidant dosage, applied current, and the reaction time; (2)

to compare the function of various EC/oxidant processes in terms of ammonia, color, phosphorous and Chemical Oxygen Demand (COD) elimination from CWLL; (3) to assess biodegradability improvement through the application of biochemical oxygen demand (BOD)/COD ratio and Average Oxidation State (AOS); (4) to determine aromaticity and humification of CWLL with different EC/oxidant systems; (5) to evaluate the phytotoxicity with germination index (GI) for the final wastewater; (6) to evaluate the cost of all electrochemical processes.

2. Material and methods

2.1. Reagent and chemicals

Sodium persulfate ($Na_2S_2O_8$ >99%), sodium percarbonate ($Na_2CO_3.1.5H_2O_2$), and Oxone salt ($KHSO_5 \cdot 0.5KHSO_4 \cdot 0.5K_2SO_4$) as PMS source were provided from Aldrich Inc. Silver sulfate (Ag_2SO_4 >99%) and mercury sulfate ($HgSO_4$ >99%) were supplied from Samchun Company. Potassium dichromate ($K_2Cr_2O_7$, >99%), sulfuric acid (H_2SO_4, >96%), ferrous ammonium sulfate ($Fe(NH_4)_2(SO_4)_2.6H_2O$, >99%) and sodium hydroxide (NaOH, >99%) were supplied from Alfa Aesar Company. **Table 1** represents some of the characteristics of the CWLL used in this study.

Table 1. Physico-chemical characterization of CWLL sample.

Parameter	Units	Value
TOC	mg/L	649
COD	mg/L	1175
TDS	mg/L	1590
BOD	mg/L	140
pH	-	6.8
Color	ADMI	3480
Ammonia	mg/L	256
Phosphorus	mg/L	4.86

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2.2. Experimental setup

CWLL was collected from Abadan municipal landfill site (Abadan, Iran, latitude: 30°26'2.83″ and longitude 48°21'41.73″) in which surface water was severely contaminated by landfill leachate. After transferring the sample to the laboratory, it was kept in closed polyethylene containers at a temperature of 4 °C. Electrochemical oxidation and electrocoagulation processes were conducted in the same reactor (**Fig. S1**). The reactor was a Plexiglas cylinder with a 3.5 cm radius and 18 cm height. 600 mL of the sample was introduced into the reactor (electrochemical cell). One pair of iron electrodes ($3\times15\times0.1$ cm) was used as anode and cathode. Electrodes were washed with ethanol and water before each experiment. The distance of the anode and cathode was 3 cm, while the effective surface area of the anode was ~ 54 cm^2. The solution was mixed by a magnetic stirrer while the solution temperature was in the range of 25-28 °C. Electrochemical reactions were started by turning on a DC power (0-2 A, 0-30 V) to supply a known electrical current. EC and EC/oxidant processes were operated at the natural pH of CWLL (~6.8). EC/oxidant process was conducted by adding a known concentration of the oxidant (PMS, PDS, HP, and PC (based on H_2O_2)) before applying an electrical current. EC and EC/oxidant were started by turning on DC power. 2 mL of the sample was undertaken and quenched by sodium nitrite to prepare for TOC analysis. The impact of applied current was evaluated in the range of 100-300 mA, while oxidant dosage was studied between 1 to 10 mM. The weighting of the iron electrode determined the amount of iron consumption before and after electrolysis time.

2.3. Analytical methods

The concentrations of COD and BOD were determined according to the standard methods (APHA, 2017). COD values were determined using the colorimetric method by a spectrophotometer (Hach, DR 5000). ADMI (American Dye Manufacturers Institute) method was employed to determine the color value in the initial and final samples. Ammonia was

194 measured based on the Nessler method using Hach reagents. The TOC analyzer (Shimadzu,

195 Japan) was used to determine the TOC of the samples. Ferrous ions were measured by the

196 1,10-phenanthroline method using a spectrophotometer (Perkin Elmer, LB25). Total

197 phosphorous was determined by the colorimetric method (Stannous Chloride Method)

198 (APHA, 2017).

199 **2.4. Phytotoxicity test**

200   The germination index is used to evaluate the phytotoxicity of untreated and treated

201 samples in different ratios. Thirty seeds of *Raphanus sativus* (Radish) and *Solanum*

202 *Lycopersicum* (Tomato) were regularly put in a Petri dish which was contained a paper filter.

203 10 mL of each effluent of the remediation process was introduced into the Petri dish. All Petri

204 dishes were incubated for 72 h at 25 °C. Distilled water was used for control tests. The GI

205 was determined based on **Eq. (11)**.

206
$$GI(\%) = \frac{G_s \times L_s}{G_c \times L_c} \times 100 \qquad (11)$$

207 where, G_s and G_c are the number of germinated seeds in effluent and control conditions,

208 respectively. L_s is the length of radicle in effluent, and L_c is the length of radicle in the control

209 condition.

210 **3. Results and discussion**

211 **3.1. Comparison of EC/Oxidant processes for TOC reduction**

212   The process efficiency of the EC and electro-activated oxidants, including EC/PMS,

213 EC/PDS, EC/HP, and EC/PC, were tested and compared for the TOC reduction efficiency in

214 CWLL samples. To do that, CWLL samples were subjected to 5 mM of oxidant, 200 mA of

215 applied current, and working at natural pH (6.8). Accordingly, it was first investigated the

216 sole application of the EC system. It was observed from **Fig. 1**; the individual EC process

217 supplied moderate TOC removal (40.4%) due to the insufficient generation of reactive

218 species. As can be seen, when the EC system was combined with oxidants, the efficiency of

219 CWLL removal was enhanced. The highest TOC removal efficiency was obtained using

220 EC/HP process with a yield of 69.6%. A synergistic impact was achieved in integrating

221 electro-activated oxidants and EC (Ghanbari et al., 2020b). TOC removal efficiency after 80

222 min reaction time follows the order EC/HP > EC/PMS > EC/PDS > EC/PC > EC process.

223 This result showed that in electro-activated systems, iron consumption is responsible for high

224 catalytic activity and activation of oxidants to generate $^\bullet OH$ and $SO_4^{\bullet-}$ radicals (Li et al.,

225 2019a). The removal of TOC during the treatment time indicates an approximate steadiness

226 when the reaction time reaches 80 min. Thus, 80 min of reaction time was selected for

227 subsequent experiments.
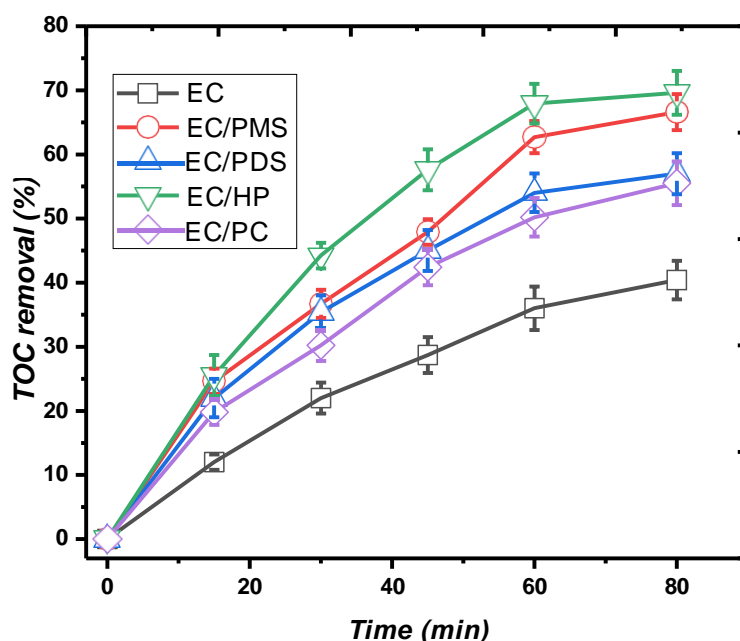
228

229 **Fig. 1**. TOC removal from CWLL sample by different processes. (oxidant = 5 mM, pH = 6.8,
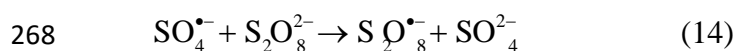230 and applied current = 200 mA).

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232 **3.2. Effect of applied current and oxidant dosage on TOC removal of EC/oxidant**

233 **systems (EC/PDS, EC/PMS, EC/HP, and EC/PC)**

234   The applied current as a significant parameter may affect the electrochemical processes'

235 performance (Brillas, 2020). The sacrificial anodes' corrosion rate and the hydrogen gas

236 generation adjacent to the cathode are related directly to the applied current. In this study, the

237 effects of applied current and PDS dosage on the TOC removal are examined to evaluate the

238 electro-activated PDS efficiency. For this study, the range 100 to 300 mA under the

239 conditions of pH = 6.8, PDS = 5.0 mM, and 80 min electrolysis time was applied to evaluate

240 the applied current effect on the TOC elimination performance via EC/PDS process, with the

241 results shown in the **Fig. (2a)**. With the applied current of the electrochemical cell set at the

242 lowest value of 100 mA, a 45% TOC elimination efficiency was achieved. By the increase of

243 applied current from 100 mA to 250 mA, this efficiency heightened from 48.9% to 65.4%.

244 Such higher efficiencies can be attributed to the increased iron electrode corrosion rate based

245 on Faraday's law, resulting in the increased sulfate radical production through the activation

246 of PDS by ferrous ions. With the further increase of the applied current to 300 mA, however,

247 the opposite trend of elimination efficiency (60%) was recorded. This can be associated with

248 the $SO_4^{\bullet-}$ radicals scavenging by the excess Fe^{2+} **(Eq. 12)** gathered in the reactor, as was

249 mentioned before (Wang and Chu, 2011). **Fig. S2** shows Fe^{2+} concentration in the solution

250 under different applied currents. As shown in this figure, with an increase of applied current,

251 ferrous ions increased. Hence, higher Fe^{2+} suppressed EC/PDS performance. This

252 phenomenon has been reported in different EC/oxidant processes. In this way, it should be

253 noted that although the coagulation mechanism needs a higher coagulant dosage (higher

254 applied current), excess ferrous ions may scavenge sulfate radicals.

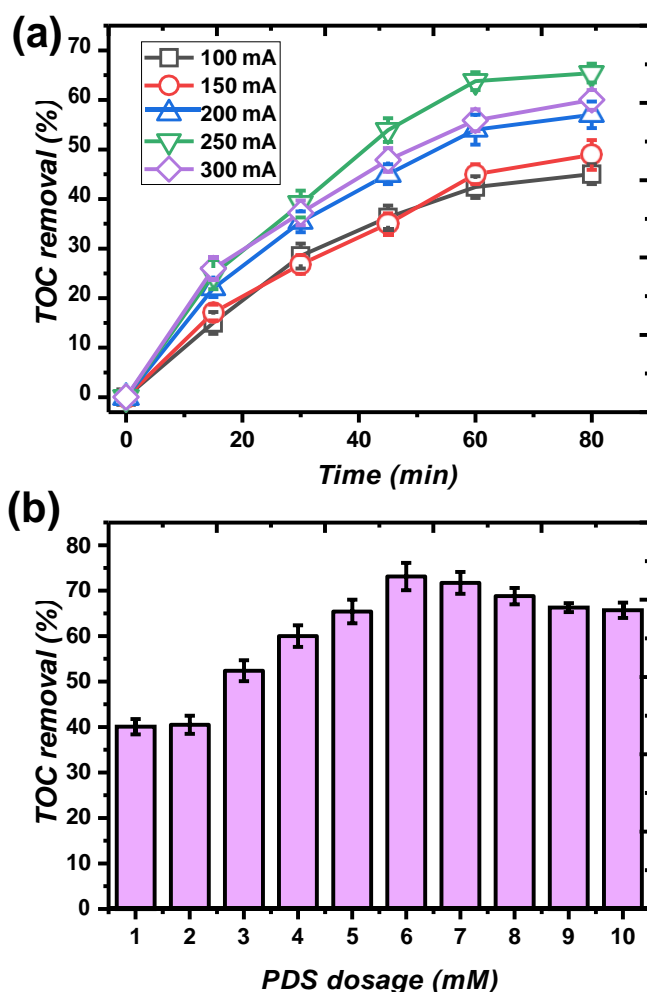255 $Fe^{2+} + SO_4^{\bullet-} \rightarrow Fe^{3+} + SO_4^{2-}$ (12)
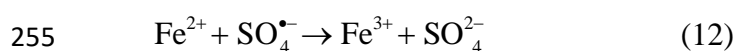
256 The added oxidant amount determines the number of free oxidizing radicals produced

257 throughout the treatment process. Regulating the process at the optimum dosage of PDS will

258 prevent the excess utilization of chemicals, resulting in reduced operating costs. Hence the

259 PDS dosage effect on the overall process is examined **(Fig. 2b)**. The mentioned figure shows

260 that the TOC elimination performance of CWLL has been increased when the PDS dosage is

261 increased from 1 mM to 6 mM. On the contrary, the excess PDS dosage over >7mM will

262 decrease the TOC elimination performance. This presumably could be attributed to the $SO_4^{\bullet-}$

263 radicals' reaction with each other in replacement of reacting with the organic contaminants

264 (**Eq. 13**). Whereas the PDS overdose possesses a scavenging capacity for the $SO_4^{\bullet-}$ radical

265 (**Eq. 14**), it possibly negatively affects the TOC elimination performance. Consequently, the

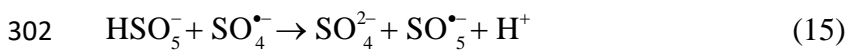266 PDS amount of 6 mM was the optimum condition with 73.1% TOC removal efficiency.

267 $$SO_4^{\bullet-} + SO_4^{\bullet-} \rightarrow S_2O_8^{2-} \qquad\qquad (13)$$

268 $$SO_4^{\bullet-} + S_2O_8^{2-} \rightarrow S_2O_8^{\bullet-} + SO_4^{2-} \qquad\qquad (14)$$

269

270

Fig. 2. (a) The effect of applied current on TOC removal (PDS = 5 mM) and (b) the effect of PDS dosage on TOC removal in EC/PDS process (applied current = 250 mA, pH = 6.8, and reaction time = 80 min).
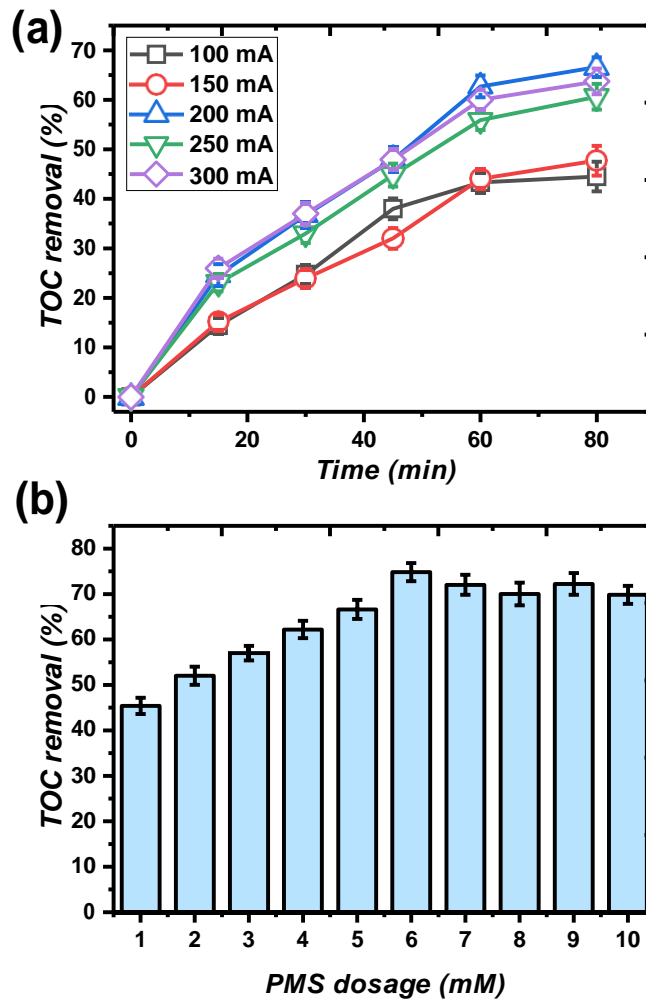
The impact of applied current on the TOC removal of CWLL was investigated in the range of 100-300 mA under the conditions of pH = 6.8, PMS = 5.0 mM, and 80 min electrolysis time. The obtained results are given in **Fig. 3a**, where an increase in removal rate was achieved when the applied current boosted from 100 to 200 mA. Nonetheless, further increasing the applied current of over 200 mA, led to diminishing the TOC removal rate.

13

281   At lower applied currents, the initial rate enhancement is likely related to the increased

282   amount of ferrous ion (Fe^{2+}) concentration nearby the anode as the applied current increases.

283   Faster production of Fe^{2+} ion leads to the more rapid generation of $SO_4^{\bullet-}$ radicals, thereby the

284   faster TOC removal of CWLL (Wang and Chu, 2011). However, according to **Eq. (12)**, the

285   excess Fe^{2+} ions may act as $SO_4^{\bullet-}$ radical scavengers when the Fe^{2+} ions accumulated up to a

286   certain amount in the solution (Rastogi et al., 2009). According to **Fig. S2**, Fe^{2+} concentration

287   increased with an increase of applied current in which Fe^{2+} concentration enhanced from 40.2

288   to 44.3 mg/L when applied current increased from 250 to 300 mA. Thus, the TOC removal of

289   CWLL is postponed because it competes with the radicals' reaction with excessive Fe^{2+} ions

290   at a higher applied current.

291   To verify the performance of electro-activated PMS, the impact of PMS dosage was

292   assessed on TOC removal. **Fig. 3b** depicts PMS dosage's effect on TOC removal during

293   80 min electrolysis time, pH of 6.8, and 200 mA applied current. As can be seen, TOC

294   removal was enhanced with increasing PMS dosage from 1mM (45.4%) to 6 mM (74.8%).

295   An enhancement in PMS concentration as the main factor of generating $SO_4^{\bullet-}$ radicals could

296   increase the removal of the organic compounds in CWLL (Wang and Chu, 2012). If the

297   excessive PMS concentrations (> 6 mM) were adopted, the TOC removal process would be

298   shortened, obviously. This decrease has corresponded to PMS overdosing in which PMS

299   played the scavenging role for $SO_4^{\bullet-}$ and $^\bullet OH$ radicals. To be specific, excess PMS reacted

300   with $SO_4^{\bullet-}$ and $^\bullet OH$ radicals and generated sulfurpentaoxide radical ($SO_5^{\bullet-}$) (**Eqs. 15 and 16**),

301   which is a weaker oxidant comparing to $SO_4^{\bullet-}$ radical (Ling et al., 2010).

302    $$HSO_5^- + SO_4^{\bullet-} \rightarrow SO_4^{2-} + SO_5^{\bullet-} + H^+ \qquad (15)$$

303    $$HSO_5^- + {}^\bullet OH \rightarrow SO_5^{\bullet-} + H_2O \qquad (16)$$

14

In comparison with PDS, PMS exhibits a slight superiority in TOC removal from CWLL. PMS has an asymmetric structure which is relatively easier to be activated with transition metals. Besides, the reaction rate constant of PMS+Fe^{2+} (30000 M^{-1} s^{-1}) is significantly higher than that of PDS+Fe^{2+} (12 M^{-1} s^{-1}) for the generation of sulfate radicals (Zhou et al., 2021).


Fig. 3. (a) The effect of applied current on TOC removal (PMS = 5 mM) and (b) the effect of PMS dosage on TOC removal in EC/PMS process (applied current = 200 mA, pH = 6.8, and reaction time = 80 min).

Several experiments were conducted at applied current ranging from 100 mA to 300 mA to determine the appropriately applied current to achieve optimum TOC elimination in
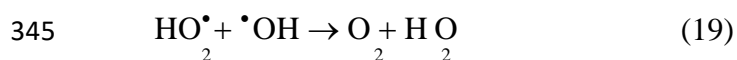
the electro-activated HP (EC/HP) system. The applied current effect on the TOC elimination performance in the CWLL effluent is represented in **Fig. (4a)**. The TOC elimination performance obtained was 48.3%, 60%, 69.6%, 67.8%, and 58% for the applied currents of 100, 150, 200, 250, and 300 mA, respectively, among which the highest performance was obtained at 200 mA. As per the applied current lower than 200 mA, the TOC removal performance was improved by the gradual increase of the applied current due to the enlargement of the ferrous ions production rate, which consequently increased of $^\bullet OH$ radicals' concentration (Özcan et al., 2008; Wang et al., 2010).

Of the reasons possibly explaining the reduction of TOC elimination efficiency could be the generation of excess Fe^{2+} ion at higher applied current that in turn could scavenge $^\bullet OH$ radical **(Eq. (17)**. In conclusion, the applied current is a determinant for generating the optimal amount of Fe^{2+} ion required for HP activation.
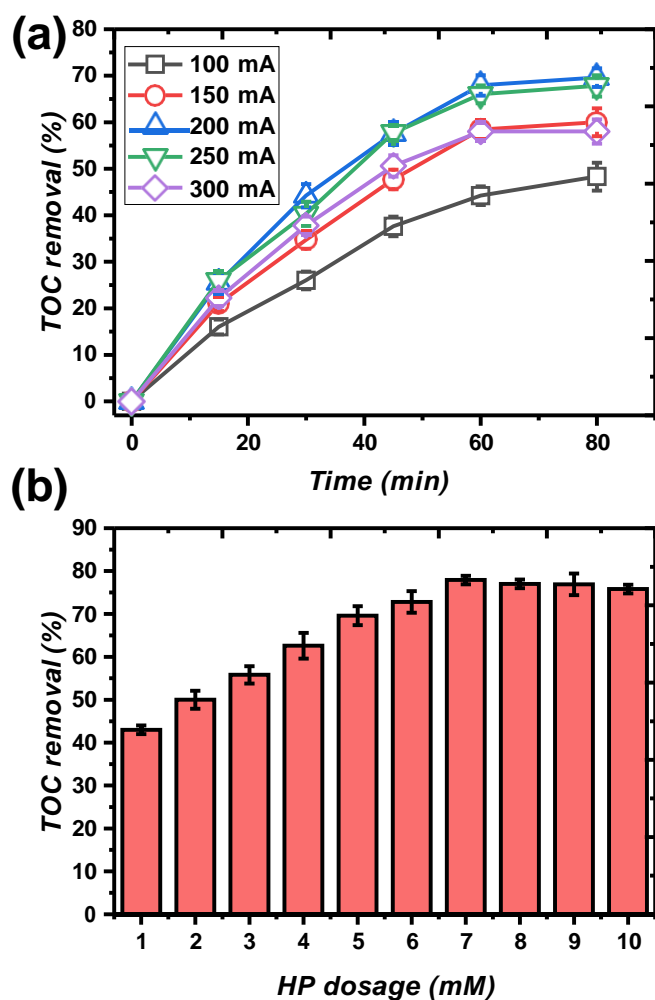
$$Fe^{2+} + {}^\bullet OH \rightarrow Fe^{3+} + OH^- \qquad (17)$$
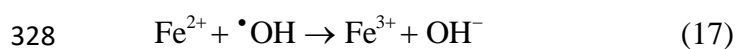
It is known that HP as an oxidant is the primary agent for $^\bullet OH$ radical generation in electrochemical systems. The impact of HP concentration in the range of 1-10 mM was evaluated on TOC removal of CWLL under the condition of pH = 6.8, 200 mA applied current, and 80 min reaction time (**Fig. 4b**). As depicted, an increase in HP concentration causes more $^\bullet OH$ radicals production and further removes organic pollutants in CWLL. This trend was achieved when HP concentration was boosted from 1 mM to 7 mM that led to TOC removal efficiency enhancement, so that TOC removal efficiencies at the end of reaction time were 43% and 77.9% for 1 mM, and 7 mM, respectively. In HP concentration higher than 7 mM, TOC removal would retard and during 80 min reaction time reached 75.8% for 10 mM HP dosage **(Fig. 4b)**. This trend happens when excessive HP reacts with $^\bullet OH$ radical, producing hydroperoxyl radical ($^\bullet OOH$), that its oxidation ability is weaker than that of $^\bullet OH$

340   **(Eq. 18)**. Furthermore, hydroperoxyl radicals ($^{\bullet}$OOH) could react with $^{\bullet}$OH radicals **(Eq. 19)**

341   and decrease the oxidation ability of the electrochemical system (Ling et al., 2010; Hassani et

342   al., 2018). For other reasons, it might be due to the recombination of $^{\bullet}$OH radicals caused by

343   excess HP (Wang and Wang, 2017).

344 $$H_2O_2 + {}^{\bullet}OH \rightarrow HO_2^{\bullet} + H_2O \qquad (18)$$

345 $$HO_2^{\bullet} + {}^{\bullet}OH \rightarrow O_2 + H_2O \qquad (19)$$
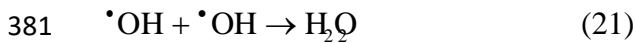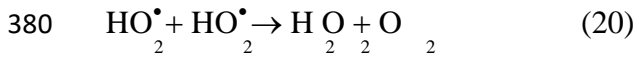
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348

349   **Fig. 4.** (a) The effect of applied current on TOC removal (HP = 5 mM) and (b) the effect of

350   HP dosage on TOC removal in EC/HP process (applied current = 200 mA, pH = 6.8 and

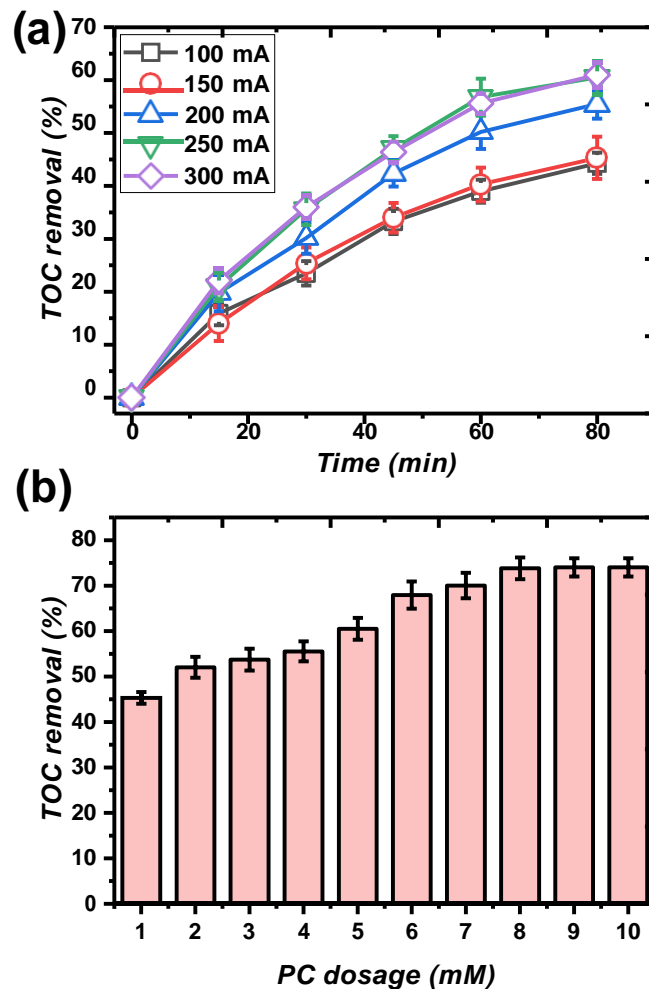351   reaction time = 80 min).

352

353   Similarly, the impact of applied current on the TOC removal of CWLL in the EC/PC

354   system was evaluated in the range of 100-300 mA. According to **Fig. 5a,** the TOC removal

355   rate seemed to be substantially boosting with enhancing applied current. Followed by 80 min

356   reaction period, TOC removals were 44.3, 45.3, 55.5, 60.5, and 61% for the applied current

357   of 100, 150, 200, 250, and 300 mA, respectively. Higher applied current permits more

358   dissolution of electrode material with a higher generation rate of iron hydroxides resulting in

359   higher destruction of organic pollutants in CWLL (Chen et al., 2018; Ghanbari et al., 2020b).

360   Therefore, the applied current of 250 mA was ample to achieve more than 60% TOC removal

361   efficiency.

362   The impact of the PC dosage on the EC/PC process is investigated, and the results are

363   presented in **Fig. 5b**. For this, PC dosage was varied in the range of 1-10 mM. It was

364   observed that an increase in PC amount improved the TOC removal efficiency when PC

365   amount was enhanced from 1 mM to 8 mM. Therefore, the higher the PC amount, the higher

366   the process efficiency due to the increased generation rate of $^\bullet$OH radicals. However, the

367   removal rate leveled off (at around 8 mM) as the PC dosage further increased over 8 mM.

368   This trend could be attributed to the scavenging of $^\bullet$OH radicals through higher amounts of

369   oxidants which has been extensively described in the literature (Nazari et al., 2019; Hassani

370   et al., 2020). Indeed, a higher dosage of oxidant (HP or PC) may scavenge $^\bullet$OH radicals to

371   generate hydroperoxyl radicals ($HO_2^\bullet$) with slight reactivity for the elimination of recalcitrant

372   pollutants in CWLL (**see Eq. 20**) (Eslami et al., 2020). Besides, free radicals' recombination

373   could also be a postulate for decreased removal performance based on **Eq. (21)** (Bremner et

374   al., 2009; Eslami et al., 2020). Compared to EC/HP, EC/PC exhibited lower efficiency while

375 PC is solid HP. The presence of carbonate ions in the structure of PC may be the main reason

376 for this reduction in which carbonate ions have high reactivity for the reaction with hydroxyl

377 radicals (k = 3.2-4.2 $\times 10^8$ M^{-1} s^{-1}) (**Eq. 9**)(Giannakis et al., 2021). Besides, carbonate radical

378 ($CO_3^{\bullet-}$, E° = 1.59 V) has a low reactivity for the oxidation of organic pollutants compared to

379 hydroxyl radical.

380 $HO_2^{\bullet} + HO_2^{\bullet} \rightarrow H_2O_2 + O_2$ (20)
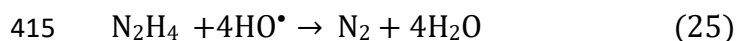
381 $^{\bullet}OH + {}^{\bullet}OH \rightarrow H_2O_2$ (21)
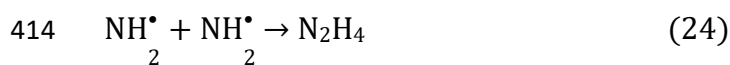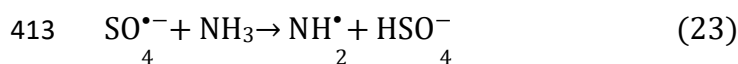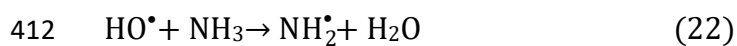
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Fig. 5. (a) The effect of applied current on TOC removal (PC = 5 mM) and (b) the effect of
PC dosage on TOC removal in EC/PC process (applied current = 200 mA, pH = 6.8, and
reaction time = 80 min).

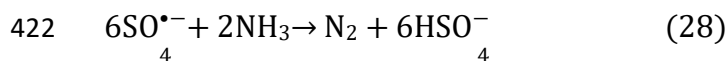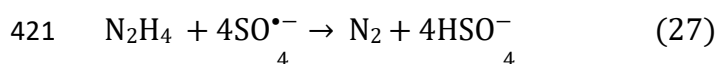### 3.3. Comparison of treatment systems on ammonia, color, phosphorous, and COD removal

The ammonia, color, phosphorous, and COD elimination from the CWLL were evaluated in various processes, including without oxidant and electro-activated oxidants at the electrolysis time of 80 min, pH 6.8, and oxidant dosage of 6 mM (**Fig. 6**). This research proved that the electro-activated process is rather superior to the sole EC in the destruction of organic pollutants from CWLL. They produced both $^\bullet OH$ and $SO_4^{\bullet -}$ radicals, and they reached the effluent disposal regulations. However, EC/HP system is more efficient compared with the other electro-activated systems. Followed by 80 min reaction time, 83.6%, 80.3%, 79.1%, 70.3%, and 43.9% of COD elimination was obtained by EC/HP, EC/PMS, EC/PDS, EC/PC, and EC systems, respectively. Similarly, EC/HP, EC/PMS, EC/PDS, EC/PC, and EC systems attained 79%, 75.9%, 73.4%, 69.3%, and 58.9% of color removal, respectively. For ammonia, removal efficiency manifested that EC/HP, EC/PC, EC/PMS, EC/PDS, and EC systems accomplished 64.3%, 60.3%, 59.8%, 58.8%, and 24.3% respectively. In a comparative study, the removal rate in terms of COD and color in various elimination processes offers the following order: EC/HP > EC/PMS > EC/PDS > EC/PC > EC. In contrast, for ammonia removal they show the following order: EC/HP > EC/PC > EC/PMS > EC/PDS > EC. This finding demonstrated that hydroxyl radical-based AOPs are more effective than sulfate radical-based AOPs in ammonia oxidation. The degradation mechanism of ammonia by hydroxyl and sulfate radicals has been rarely considered. In this way, hydroxyl and sulfate radicals can oxidize NH_3 to $NH_2^\bullet$ through hydrogen-abstraction reactions according to Eqs. (22 and 23). The recombination of $NH_2^\bullet$ results in the formation of hydrazine (N_2H_4) (Eq. 24). The reaction of hydrazine with hydroxyl radicals is kinetically favorable to generate N_2 and water (Eq. 25) (Yan and Liu, 2020).

412    $HO^\bullet + NH_3 \rightarrow NH_2^\bullet + H_2O$　　　　(22)

413    $SO_4^{\bullet-} + NH_3 \rightarrow NH_2^\bullet + HSO_4^-$　　　　(23)

414    $NH_2^\bullet + NH_2^\bullet \rightarrow N_2H_4$　　　　(24)

415 $N_2H_4 + 4HO^\bullet \rightarrow N_2 + 4H_2O$　　　　(25)

416   On the other hand, a high concentration of hydroxyl radicals may directly oxidize ammonia

417   into N_2 based on **Eq. (26)** (Amanollahi et al., 2021).

418    $6HO^\bullet + 2NH_3 \rightarrow N_2 + 3H_2O$　　　　(26)
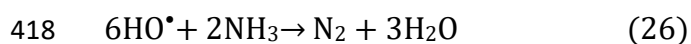
419   For degradation of ammonia by sulfate radicals, some reactions can be considered similar to

420   the hydroxyl radicals mechanism. Howbeit, Eqs. (27 and 28) should be further investigated.

421    $N_2H_4 + 4SO_4^{\bullet-} \rightarrow N_2 + 4HSO_4^-$　　　　(27)

422    $6SO_4^{\bullet-} + 2NH_3 \rightarrow N_2 + 6HSO_4^-$　　　　(28)

423   Phosphorous removal was also evaluated in five processes. As can be seen, around 82% of

424   phosphorous was eliminated from CWLL by all methods. Since EC performance was

425   competitive with others, it can be concluded that the mechanism of phosphorous removal is

426   the coagulation of phosphorous. Generally, phosphate ion has a strong affinity for iron. Iron

427   oxides in water have surface OH groups. Phosphate ion in the solution undergoes ligand

428   exchange with the hydroxyl ions. Adsorption of PO_4^{3-} on surfaces of iron oxide results in

429   inner surface complexation. This complex [Fe(OH)$_{3-x}$. (PO$_4$)$_x$] either adsorb onto positively

430   charged Fe(III) hydrolysis species or act as the core of precipitation for Fe(III) hydrolysis

431   products (Omwene et al., 2018). Moreover, direct precipitation of FePO$_4$ and Fe$_3$(PO$_4$)$_2$ may

432   occur in the EC process according to Eqs. (29 and 30). These precipitants are stable with low

433   solubility and are rapidly settled.

434 $$Fe^{2+} + PO_4^{3-} \rightarrow Fe_3(PO_4)_2 \qquad (29)$$

435 $$Fe^{3+} + PO_4^{3-} \rightarrow FePO_4 \qquad (30)$$
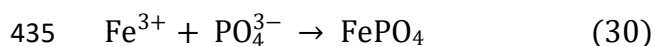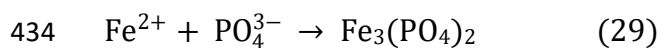
436   As a result, the enhanced TOC removal efficiency in the combined systems (EC/oxidant)

437 was related to the combination of coagulation and oxidation mechanisms in which separation

438 and oxidation processes simultaneously occurred. To compare EC/oxidant process, Specific

439 Oxidant Efficiency (SOE) was determined based on **Eq. (31)** (Li et al., 2020).

440 $$SOE = \frac{\Delta TOC}{\Delta oxidant} \qquad (31)$$

441   where ΔTOC and $\Delta oxidant$ are TOC decay and oxidant consumed during electrolysis time

442 (mM). This index indicates that the higher SOE represents higher efficiency of oxidant

443 consumption. The results of SOE are presented in **Fig. S3**. As can be seen, EC/HP shows a

444 high value (9.14) compared to other processes (~7.0) confirming the high performance of

445 EC/HP in terms of oxidant efficiency and TOC removal.

446 The discharge standard of the effluent into municipal sewage for COD index is 250 mg/L.

447 According to the results obtained, EC/PDS, EC/PMS, and EC/HP reduced COD values to

448 245, 230, and 190 mg/L, respectively to meet this standard. However, discharging of toxic

449 wastewater into municipal sewage should be conducted warily.

450
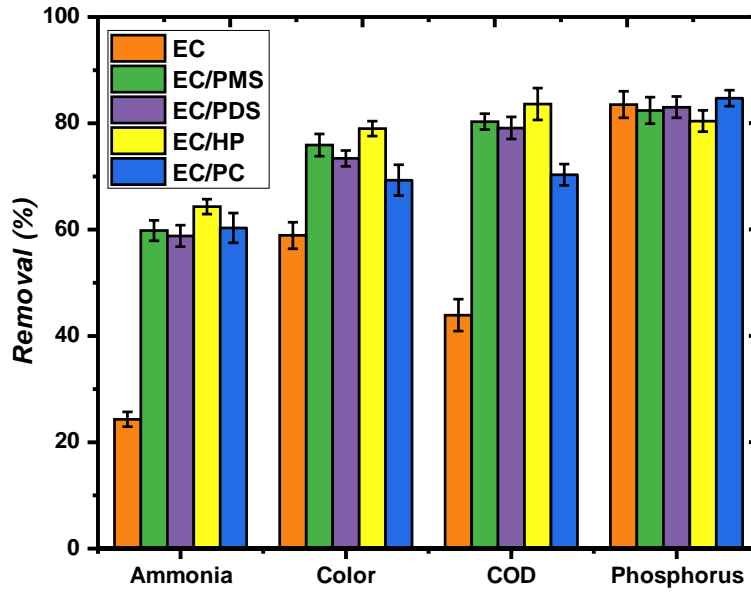
Fig. 6. Ammonia, color, COD and phosphorus removal from CWLL by different processes
(oxidant = 6 mM, applied current = 200 mA, and reaction time = 80 min).

3.4. Biodegradability assessment

The biodegradability of the water sample has been measured by various indices. In this research, the average oxidation state (AOS) and BOD_5/COD ratio, have been considered for biodegradability ascertainment of CWLL sample after treatment, and the results are given in **Fig. 7**. AOS has been evaluated using the following relation (**Eq. 32**) (Lu et al., 2011):

$$AOS = 4 - 1.5\frac{COD}{TOC} \qquad (32)$$

where TOC and COD refer to total organic carbon and the chemical oxygen demand of the solution after the treatment system, respectively. In **Fig. 7a**, AOS values increased from 1.28 to 1.44, 1.89, 2.01, 2.36, and 1.67 respectively, for EC, EC/PMS, EC/PDS, EC/HP, and EC/PC processes. These results indicated the formation of oxidized organic matter after treatment by EC and electro-activated oxidants systems. These oxidized organic matters display that both EC and electro-activated oxidants systems can improve the biodegradability

of CWLL (Garg and Mishra, 2010). For untreated leachate, the BOD_5/COD of 0.12 was obtained, which indicates low biodegradability. For electrocoagulation, this ratio was almost the same as raw leachate. However, this ratio was increased to 0.26, 0.24, 0.36, and 0.17 for EC/PMS, EC/PDS, EC/HP, and EC/PC **(Fig. 7b)**, respectively; that indicates an integration of EC and oxidants could improve the biodegradability of treatment. EC/HP showed a high value for BOD/COD ratio (0.36), indicating its effluent is relatively biodegradable. In conclusion, the increase in BOD_5/COD, and AOS values illustrates that the integration of EC with oxidants could be the best pre-treatment for biological systems.
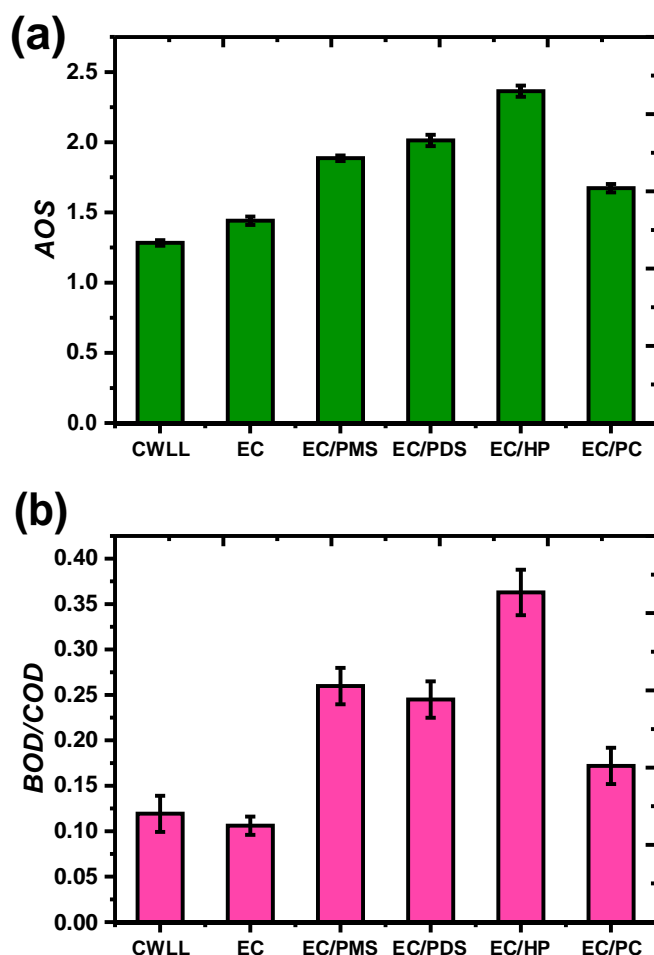

Fig. 7. (a) AOS value, and (b) BOD/COD value for CWLL before and after treatment by different processes.

24

3.5. Evaluation of E_2/E_3 and $SUVA_{254}$ indices after electrochemical treatments

Aromaticity and molecular mass of the effluents after each electrochemical treatment were evaluated by specific ultraviolet absorbance 254 ($SUVA_{254}$) and E_2/E_3 indices. $SUVA_{254}$ is obtained by UV_{254} absorbance divided by TOC concentration ($SUVA_{254}=A_{254}/TOC\times100$) while E_2/E_3 is calculated by A_{250} divided by A_{365}, indicating humification and molecule mass (Xu-jing et al., 2011). **Fig. 8** shows $SUVA_{254}$ and E_2/E_3 indices in CWLL and after electrochemical treatment. As can be seen, $SUVA_{254}$ and E_2/E_3 did not change after the EC process, indicating that the EC process is not destructive. Electrochemical oxidation processes significantly reduced both indices. $SUVA_{254}$ was reduced from 0.43 to 0.115, 0.12, 0.088 and 0.128 after the treatment by EC/PMS, EC/PDS, EC/HP and EC/PC respectively. These demonstrated that refractory organics with high aromaticity were considerably degraded by EC/oxidant processes. Moreover, the E_2/E_3 index was increased by all electrochemical processes. The E_2/E_3 index raised from 4.66 to 9.6, 10.17, 11.06, and 9.05 after applying EC/PMS, EC/PDS, EC/HP, and EC/PC, respectively. This increase in the E_2/E_3 is attributed to the degradation of large molecules (such as fulvic and humic acids) and the production of intermediates with low molecule weight (Jiang et al., 2019). According to these results, EC/HP had slight supremacy than other systems with lower $SUVA_{254}$ and higher E_2/E_3.
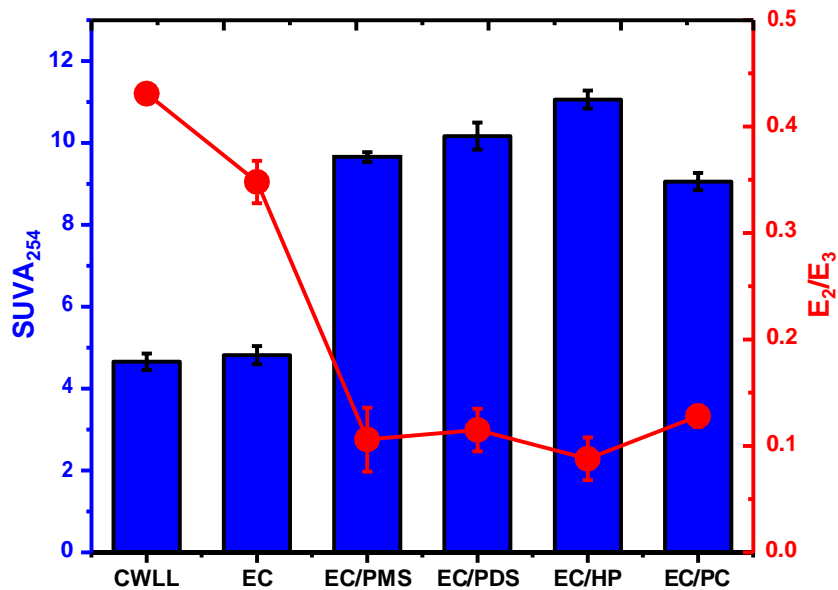
Fig. 8. SUVA$_{254}$ and E$_2$/E$_3$ indices after electrochemical treatment.

3.6. Phytotoxicity evaluation

Table 2 contains the germination index (GI (%)) results, simultaneously comprising the solution effect on growth and germination. McLauchlan et al. (2004) believed that the above 70% germination index was non-phytotoxic and lower than that figure could be deemed phytotoxic. The obtained GI percentage figures in raw CWLL were of no significance, denoting that the CWLL itself was considerably phytotoxic. After the EC treatment, the phytotoxicity decreased, which is evidence of the increased GI values. As for the EC/PMS and EC/HP systems, making dilutions to 25% and 50% caused the removal of toxicity effect, whereas, for the EC/PC and EC/PDS systems, only a dilution of 25% removed the toxicity (>70%). Generally, EC/HP with a dilution of 25% has the highest GI value (93.5%). These results showed that after treatment by EC/HP, diluted effluent could be used for water reuse in the agriculture sector (McLauchlan et al., 2004). The detoxification of the effluent for irrigation is not just a criterion for this section; several issues should be considered before application. In summary, it can be concluded that the electro-activated oxidants can remove

512 or at least weaken the CWLL toxicity. Besides, the tomato has greater sensitivity to toxic-

513 containing effluents than radish in terms of germination and growth.

514 **Table 2**. Germination Index for differently treated effluents with their dilutions.

Effluent	GI (%) for Radish	GI (%) for Tomato
CWLL	<4.5±2	0
CWLL (50%)	12.8±3.5	6.3±3.3
CWLL (25%)	18.9±6.2	18.9±8.2
EC	7.6±2.7	8.5±3.4
EC (50%)	20.2±4.5	12.6±4.4
EC (25%)	56.5±8.3	29.3±7.6
EC/PMS	31.1±9.5	22.2±6.7
EC/PMS (50%)	70.7±7.5	52.1±6.4
EC/PMS (25%)	90.4±6.4	75.6±8.7
EC/PDS	28.8±8.2	23±4.9
EC/PDS (50%)	62.2±8.8	40.3±5.1
EC/PDS (25%)	84.3±9.6	68.8±8.1
EC/HP	40.8±7.9	33.9±9.2
EC/HP (50%)	74±6.9	50.2±9.8
EC/HP (25%)	93.5±5.1	79.9±8.7
EC/PC	25.3±8.5	24.7±7.9
EC/PC (50%)	40.3±7.2	42.3±6.0
EC/PC (25%)	73.4±11.5	69.8±12.1

515

516 **3.7. Electrical Energy Consumption and Cost evaluation**

517 The most important economic factor of electrochemical processes is electrical energy

518 consumption (EEC). EEC can be calculated based on both the treated volume and TOC

519 removed according to **Eqs**. (**33 and 34**) (Martínez-Huitle and Brillas, 2009).

520 $$EEC = \frac{VIt}{V_s} \qquad (33)$$

521 $$EEC_{TOC} = \frac{VIt}{V_s \times \Delta TOC} \qquad (34)$$

522 where V is an average of voltage (V), I is applied current (A), t is electrolysis time (h), V_s is

523 the volume of the CWLL (L), ΔTOC is the removal of TOC at t time (g/L), EEC (kWh/m^3) is

524 electrical energy consumption per cubic meter of CWLL treated and EEC_{TOC} (kWh/kg TOC)

525 is electrical energy consumption per kg TOC removed.

526 An economic evaluation was investigated by considering the electrode materials' costs,

527 chemical oxidants, and the consumed electricity. This evaluation was based on **Eq. (35)**

528 (Mena et al., 2019).

529 $$\text{Operation Cost} = a(C_e) + b(C_0) + c(EEC) \qquad (35)$$

530 where C_e is kg iron lost, and C_o is kg oxidant consumed. The parameters of a, b, c are the

531 costs of USD/kg iron, USD/kg oxidant, and USD/kWh, respectively.

532 **Table 3** shows the EEC, EEC_{TOC}, and operation costs of different processes. Regarding all

533 EC/oxidant processes with approximately the same efficacy, their EEC and EEC_{TOC} values

534 were close together. However, EC/PMS exhibited a lower EEC_{TOC} value compared to others.

535 In operation cost, except for the EC process, EC/PMS had the maximum operation cost after

536 EC/PDS, EC/PC, and EC/HP. These results are attributed to the high price of PMS compared

537 to other oxidants (Wacławek et al., 2017).

538 Regarding the results, EC/HP showed supremacy compared to other processes. Indeed, HP

539 had not only acceptable results for TOC, color, ammonia, and COD removals from CWLL,

540 but it also exhibited a cost-effective oxidant for the EC process. According to the literature,

541 EC/ozone for textile wastewater treatment (Bilińska et al., 2019) showed high electrical

542 energy consumption (115.13 kWh/m^3) and cost (11.86 USD/m^3). In another study, the total

543 cost of coagulation followed by the PDS/microwave process was calculated to be 46.2

544 USD/m^3 for landfill leachate treatment (Tripathy and Kumar, 2019). Application of EC

545 process for landfill leachate had a total cost of 1.40 USD/kg COD (Tanyol et al., 2017). In

546 comparison with other studies, EC/oxidant can be considered as an economical process for

547 the treatment of wastewater with low cost.

548 Although recent other alternative oxidants (PMS, PDS, and PC) have some advantages,

549 including safe storage, non-radical pathway, the longer half-life of sulfate radicals, and the

550 simultaneous use of sulfate radicals and hydroxyl radicals in media (in the case of PMS and

551 PDS), some limitations such as residual sulfate ions in the effluent, an increase of sodium or

552 potassium in effluent and higher cost cause that HP still keeps its position among water and

553 wastewater treatments (Giannakis et al., 2021). However, the selection of an oxidant for

554 AOPs needs a comprehensive investigation of efficiency, environmental impacts, availability,

555 cost, oxidant residual, safe storage, etc.

556

557 **Table 3.** EEC, EEC$_{TOC}$, and operation cost of different processes.

Processes	EEC kWh/m^3	EEC$_{TOC}$ kWh/kg TOC	Operation Cost (USD/m^3)
EC	5.32	20.30	0.57
EC/PDS	5.00	10.35	1.62
EC/PMS	4.96	10.70	8.69
EC/HP	5.43	11.52	0.88
EC/PC	5.08	12.0	1.03

558

3.8. Comparison with other AOPs

560 EC/HP as the best process among other EC processes was compared to AOPs in literature.

561 **Table 4** shows different studies for the treatment of leachate-based effluents. As can be seen,

all the AOPs involved eliminated organic compounds from the leachate, with COD removal

ranging from 57% to 88%. Fenton and EC processes have the problem of sludge production

similar to current work while they are usually efficient for COD removal (Mahtab et al.,

2020; Ding et al., 2021). The use of UV and ultrasound (US) is an effective method for

peroxide activation, but with highly electrical energy consumption (Moradian et al., 2020).

Persulfate anions (PMS and PDS) generate sulfate ions as the main by-products of oxidizing

organic pollutants. Hence, sulfate radical processes suffer from this imperfection (Abu Amr

et al., 2013; Zhang et al., 2014). Ozonation and catalytic ozonation are effective processes for

wastewater treatment, but the cost of ozone generation is one of the most important

disadvantages. Moreover, generating aldehydes as by-products are a critical issue in treated

effluents (Abu Amr et al., 2013; Abu Amr et al., 2017). EC/HP exhibited high efficiency and

low energy consumption compared to others, while sludge production is still the main

challenging for scale-up.

Table 4. Comparison of the performance of different AOPs for treatment of landfill leachate.

Process	Condition	Yield	Disadvantages	Advantages	Ref.
Electro/PDS/Fe^{2+}	pH = 3, PDS = 62.5 mM, Fe^{2+} = 15.6 mM, Current density = 13.89 mA/cm^2, and Time = 60 min	Initial COD = 1900 mg/L Yield = 62.2%	High dosage PDS and iron, Residual iron and sulfate ions	Low energy consumption	(Zhang et al., 2014)
Fenton Process	pH = 3.1, HP = 75 mM, Fe^{2+} = 40 mM, and Time = 36 min	Initial COD = 8900 mg/L Yield = 61%	Sludge production, need to acidic condition, high dosage of HP	Efficient and low reaction time	(Mahtab et al., 2020)
EC/electro-Fenton like (dual anodes)	pH = 7, current density = 30 mA/cm^2, and electrical charge = 9 Ah/L	Initial COD = 1300-1600 mg/L Yield = 57%	Sludge production, Low efficiency	In situ electrogeneration of oxidant, relatively low energy consumption	(Ding et al., 2021)
Ozone/$ZrCl_4$	pH = 6, O_3 27g/m^3, $ZrCl_4$/COD = 1:2, and Time = 90 min	Initial COD = 3750 mg/L Yield = 88%	Residual zirconium, cost of ozone production and $ZrCl_4$	Efficient, simple operation	(Abu Amr et al., 2017)
PMS/UV/US	pH = 3, PMS = 4	Initial COD =	High energy	Low oxidant dosage	(Moradia

	mM, and Time = 150 min	2450 mg/L Yield = 59%	consumption, reduction of biodegradability, residual sulfate ions		n et al., 2020)
Ozone/persulfate	pH = 10, O_3 = 80 g/m^3, COD/PDS = 1:7, and Time = 210 min	Initial COD = 3750 mg/L Yield = 72%	High consumption of PDS, long reaction time, residual sulfate ions	Simple operation	(Abu Amr et al., 2013)
$CuFe_2O_4$/PMS/UV	pH = 5.7, $CuFe_2O_4$ = 0.15 g/L, PMS = 15 mM, and Time = 120 min	Initial COD = 1700 mg/L Yield = 77.9%	High energy consumption, High oxidant dosage, Residual sulfate ions	Efficient, recoverable catalyst	(Ghanbar i et al., 2020a)
EC/HP	HP = 6 mM, applied current = 200 mA, and Time = 80 min	Initial COD = 1175 mg/L Yield = 83.6%	Sludge production	Efficient, low energy consumption	Current work

576

4. Conclusion

The combined EC with oxidants (i.e., electro-activated system) was performed to TOC removal of CWLL. The obtained results illustrated that the oxidants were successfully activated by the produced ferrous ions in the EC process. The individual EC process supplied moderate TOC removal (40.4%) due to the inadequate contributes to generating reactive species. However, the combined processes (EC/oxidant), with a synergistic impact, improved the TOC removal efficiency of CWLL. The concurrent utilization of EC and oxidant enhanced removal performance as the order of TOC removals was EC/HP > EC/PMS > EC/PDS > EC/PC > EC. The impacts of operating factors, including applied current, oxidant dosage, and electrolysis time, were assessed on various processes of EC/PMS, EC/PDS, EC/HP, and EC/PC in terms of the TOC removal. The highest ammonia, color, and COD removal efficiencies of 64.3%, 79%, and 83.6%, respectively, were obtained for the EC/HP system under optimum conditions of the applied current of 200 mA, HP dosage of 6 mM, pH 6.8, and reaction time of 80 min. The integration of EC and oxidants improved the

591 biodegradability of the treated sample. The germination index was enhanced after EC and

592 electro-activated oxidant processes. Economic valuation exhibited that EC/HP is the best

593 option for the treatment of CWLL. Based on the results of this research, it was reasonable to

594 infer that the electro-activated oxidant process could be a promising way to the treatment of

595 different kinds of pollutants due to its high mineralization efficiency. In summary, the

596 electro-activated oxidant system was proven to be a convenient operation, affordable, and

597 suitable system for the TOC elimination of CWLL, thus providing a novel strategy for

598 oxidants activation aimed at the removal of organic pollutants from water and wastewater.

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