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# Electrosorption of phenolic compounds from olive mill wastewater: Mass transport consideration under a transient regime through an alginate-activated carbon fixed-bed electrode

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1           **Electrosorption of phenolic compounds from olive mill**  
2           **wastewater: Mass transport consideration under a transient**  
3           **regime through an alginate-activated carbon fixed-bed electrode**

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18 **Abstract**

19 Olive mill wastewater (OMWW) is an environmentally critical effluent, specifically due to its  
20 high content of phenolic compounds (PCs), which are hazardous due to their antimicrobial  
21 activities in water. However, their properties have good health effects at suitable doses. For  
22 the first time, the electrosorption of PCs from actual OMWW has been proposed for their  
23 possible recovery as value-added compounds, while decontaminating OMWW. A bio-sourced  
24 alginate-activated carbon (AC) fixed-bed electrode was prepared based on the reuse of olive  
25 pomace solid waste as powdered AC. At the optimal AC content (1% w/v), the internal ohmic  
26 drop voltage was lower (2.26 V) and the mass transport coefficient was higher ( $9.7 \cdot 10^{-5} \text{ m s}^{-1}$ )  
27 along with the diffusivity ( $7.3 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$ ), which led to enhanced electrosorption rates.  
28 Afterward, an optimal electrode potential was obtained (-1.1 V vs. Ag/AgCl), while higher  
29 voltages led to faradaic reactions. Moreover, the adsorption capacity was lower ( $123 \text{ mg g}^{-1}$ )  
30 than that of electrosorption ( $170 \text{ mg g}^{-1}$ ) and was even higher ( $307 \text{ mg g}^{-1}$ ) with actual  
31 effluents. This was probably due to the influence of electromigration, which was confirmed  
32 by new models that could predict the electrosorption kinetics well considering mass transport  
33 and acid dissociation constants.

34

35 **Keywords:** Phenolic compounds; Activated carbon beads; Capacitive current; Fixed-bed  
36 column reactor; Modeling

## 37           **1. Introduction**

38   The release of olive mill wastewater (OMWW) into the aquatic environment is a major issue  
39   (Azzam and Hazaimah, 2021; Esteves et al., 2021; Haddad et al., 2021; Lissaneddine et al.,  
40   2021a). These effluents are toxic; they have a high inorganic salt content (electrical  
41   conductivity  $\sim 5\text{--}23 \text{ mS cm}^{-1}$ ) (Aharonov-Nadborny et al., 2016; Lissaneddine et al., 2021a)  
42   and a high organic load, i.e., high chemical oxygen demand (COD) ( $\sim 110 \text{ g-O}_2 \text{ L}^{-1}$ ) (Taylor  
43   et al., 2015). They are especially saturated with aromatic molecules, particularly phenolic  
44   compounds (PCs). They pose a serious environmental problem due to their phytotoxicity,  
45   which inhibits the activity of plants and microorganisms (Elayadi et al., 2021, 2019). PCs also  
46   affect surface water and groundwater, threatening aquatic life and altering soil quality  
47   (Elayadi et al., 2021). Moreover, hazardous OMWW is concentrated in a short period of time  
48   each year since it is a seasonal activity. However, PCs also represent added-value molecules  
49   at reliable doses that can be recovered for subsequent valorization in the pharmaceutical,  
50   cosmetic and food industries (Annab et al., 2019). Several physical, chemical, biological or  
51   combined technologies have been proposed to remove PCs, including separation methods  
52   (e.g., adsorption, membrane separation and ion exchange) and degradation techniques such as  
53   advanced oxidation processes (e.g., electro-Fenton, heterogeneous photocatalysis)(Brillas and  
54   Garcia-Segura, 2020; Genethliou et al., 2020; He et al., 2020; Lissaneddine et al., 2021a;  
55   Mousset, 2020; Mousset et al., 2021, 2018b, 2017, 2016a; Tundis et al., 2020; Ververi and  
56   Goula, 2019; Yue et al., 2019a). Separation technologies offer the possibility to recover these  
57   added-value compounds under specific applied conditions instead of degrading them.  
58   Electrosorption is gaining particular attention (Macías-garcía et al., 2019; Sahin et al., 2020)  
59   due to its advantages of selectivity, desorption control and low energy requirements (Sahin et  
60   al., 2020; Yang et al., 2021). Electrode materials are of primary importance in such an  
61   electrochemical system because they impact the electrosorption rates and yields. These

62 characteristics depend on the specific surface area, pore size distribution, electric  
63 conductivity, surface function and wettability (Lissaneddine et al., 2021b; Sun et al., 2016;  
64 Yue et al., 2019b). Three-dimensional (3D) particle electrodes provide a large electroactive  
65 surface area and can be easily implemented in fixed-bed reactors to favor mass transfer  
66 (Andrés García et al., 2018; Lissaneddine et al., 2021b; Macías-garcía et al., 2019; Yue et al.,  
67 2019b). Recently, 3D particle electrodes were shown to exhibit advantages in providing  
68 additional active sites and enhanced mass transfer compared to two-dimensional (2D)  
69 electrodes, which is conducive to improving the removal efficiency (Wang et al., 2021).  
70 Various 3D and bio-sourced materials have been employed for adsorption technologies. For  
71 instance, alginate offers advantages, as it acts as an embedding medium to assist in the  
72 preparation of easily separating and recycling adsorbents (Xu et al., 2021). It allows the  
73 implementation of powdered materials that have a larger specific surface area than granular  
74 adsorbents without the need for a postfiltration step to remove the fine adsorbent particles.  
75 Many researchers have employed beads with alginate coatings on several kinds of adsorbent  
76 materials, such as activated carbon (AC) (Girijan et al., 2021), or functionalized them to be  
77 more efficient (Wadhawan et al., 2020). These composite beads have been tested to remove  
78 organic pollutants in wastewater (Khan et al., 2021; Xu et al., 2021). In our recent article, bio-  
79 sourced beads based on a mixture of sodium alginate (SA) and powdered AC (SA-AC) made  
80 from olive pomace waste were used for the elimination of PCs from OMWW by adsorption  
81 (Lissaneddine et al., 2021a). However, pollutant transport within the beads is limited with  
82 such technology. This phenomenon should be less limited when electrosorption is used, as  
83 electromigration is favored, for example. To the best of the authors' knowledge, the  
84 interesting properties of alginate composite beads have not previously been tested for  
85 electrosorption.

86 Moreover, different studies have already demonstrated the performance of 3D electrodes for  
87 the electrosorption of organic compounds (e.g., acid red 88, orange II, methylene blue and  
88 metribuzin) from wastewater (Lissaneddine et al., 2021b). Matrix effects have rarely been  
89 considered, while a supporting electrolyte has always been added in electrosorption  
90 experiments (Lissaneddine et al., 2021b). However, matrix effects are known to interfere with  
91 the electrochemical process efficiency, and the addition of salts constitutes a supplementary  
92 source of pollution (Lissaneddine et al., 2021b; Mousset, 2020; Mousset et al., 2020, 2019,  
93 2018a). Moreover, the few existing models of organic electrosorption do not include the  
94 physico-chemical properties of the compounds or mass transport phenomena, although they  
95 are of primary importance in the process efficiency (Lissaneddine et al., 2021b).

96 It is within this context that a detailed investigation has been carried out for the first time to  
97 understand and predict the electrosorption of PCs from simulated and actual OMWW. The  
98 prepared SA-AC beads were newly implemented as a 3D electrode for electrosorption  
99 experiments. The effect of the main parameters, i.e., the electrode preparation (crosslinking  
100 agent and AC content) and experimental conditions (applied electrode potential), on the  
101 electrosorption kinetics and capacity are stated in detail. In addition, novel theoretical models  
102 are proposed to predict the evolution of the electrosorption of organic compounds in  
103 simulated and real effluents, considering the mass transport and difference in acid dissociation  
104 constants ( $pK_a$ ) of the PCs.

105

## 106 **2. Experimental**

### 107 **2.1. Effluent preparation and characterization**

108 Two kinds of effluents were studied: one was a simulated OMWW that was used to optimize  
109 the operating parameters, and the second was an actual OMWW. The latter was employed to  
110 study the matrix effect by comparison with the simulated effluent.

111 Simulated OMWW was prepared using potassium chloride (KCl) ( $6 \text{ g L}^{-1}$ ) and phenol ( $4 \text{ g L}^{-1}$ )  
112 and adjusted to pH 4 using hydrochloric acid (HCl) (Sigma Aldrich, Saint-Quentin-  
113 Fallavier, France) to be representative of the average characteristics of actual OMWW  
114 (Lissaneddine et al., 2021a). The phenol content was determined by UV-visible  
115 spectrophotometry (Shimadzu UV-2600 spectrophotometer) at the maximum absorption  
116 wavelength of 269 nm (Adebayo and Areo, 2021).

117 Real OMWW was collected from modern oil mills located in the region of Marrakech in  
118 Morocco during the olive oil extraction season from December to March in the 2020/2021  
119 campaign when the main products were PCs (Lissaneddine et al., 2021a). The main  
120 characteristics were pH = 4.8,  $[\text{Cl}^-] = 8.5 \text{ g L}^{-1}$ , conductivity =  $16 \text{ mS cm}^{-1}$ , and total PCs =  
121  $10.6 \text{ mg L}^{-1}$ ; details of the characteristics are described elsewhere (Lissaneddine et al., 2021a).  
122 The total PC content was measured by the Folin-Ciocalteu method using meta-phosphoric  
123 acid, petroleum ether, ethyl acetate and methanol as reagents (Sigma Aldrich, Saint-Quentin-  
124 Fallavier, France) (Singleton, V. L., & Rossi, 1965). All commercially available reagents  
125 were used without further purification, while ultrapure water ( $18.2 \text{ M}\Omega \text{ cm}$ ) (PureLab ELGA  
126 Classic, Veolia Water, Antony, France) was used in all experiments and analyses.

127

## 128 **2.2. Preparation and characterization of the 3D porous electrode**

129 The porous material (SA-AC beads) was prepared based on activated carbon of olive pomace  
130 according to a previous paper (Lissaneddine et al., 2021a). First, a mixture of a hydrogel of

131 sodium alginate (BioChemica) (1%) was fabricated at 60 °C. Second, AC powder was added  
132 to the alginate hydrogel with constant stirring for 24 h to obtain a homogeneous dispersion.  
133 Next, the dispersed solution was poured dropwise into a crosslinking solution for bead  
134 solidification. The presence of a crosslinking agent leads to competition between the gelling  
135 reaction and polyelectrolyte complex, which also results in the formation of a more porous gel  
136 (Mi et al., 2002). Two crosslinking agents (iron(III) (Fe) and calcium(II) (Ca)) were used in  
137 bead preparation, both at 100% weight per volume (% w/v). The proportions in % w/v of AC  
138 (0%, 0.5%, 0.75%, 1%, 2%, and 3%) were varied in the SA-AC bead preparation process. The  
139 surface area (Brunauer-Emmett-Teller (BET) analysis), surface morphology (scanning  
140 electron microscopy (SEM)), elemental composition (energy-dispersive X-ray spectroscopy  
141 (EDS)) and surface function (Fourier transform infrared spectroscopy (FTIR)) of the initial  
142 beads have been described previously (Lissaneddine et al., 2021a).

143 The electrochemical properties of the prepared SA-AC beads electrode were evaluated using a  
144 potentiostat (AMETEK, Massy, France) in a three-electrode system whose design is described  
145 in Section 2.3. The SA-AC beads, a graphite rod and a Ag/AgCl electrode were employed as  
146 the working electrode, counter electrode and reference electrode, respectively. The  
147 electroactive surface of the porous material was determined by cyclic voltammetry (CV) with  
148 -0.2 to 0.8 V vs. Ag/AgCl as the voltage range and 0.01 V s<sup>-1</sup> as the scan rate. The  
149 electroactive surface area ( $A$  in cm<sup>2</sup>) was calculated according to the Randles-Sevcik equation  
150 (Eq. 1) from the peak currents determined with the polarization curve (Mousset et al., 2016b).

$$151 \quad I_p = 2.69 \times 10^5 \times AD^{1/2}n^{3/2}\gamma^{1/2}C \quad (1)$$

152 where  $I_p$  is the peak current (in A),  $n$  is the number of electrons participating in the redox  
153 reaction ( $n=1$ ),  $D$  is the diffusion coefficient of [Fe(CN)<sub>6</sub>]<sup>3-</sup> ( $7.60 \times 10^{-6}$  cm<sup>2</sup> s<sup>-1</sup>),  $C$  is the  
154 concentration of [Fe(CN)<sub>6</sub>]<sup>3-</sup> ( $1 \times 10^{-5}$  mol cm<sup>-3</sup>) and  $\gamma$  is the scan rate.



155 Tafel slopes were obtained from linear sweep voltammetry (LSV) with -2 to 2 V vs. Ag/AgCl  
156 as the voltage range and  $0.01 \text{ V s}^{-1}$  as the scan rate and were used to calculate the exchange  
157 current density ( $I_0$ ) of the 3D porous electrode (Adnan et al., 2021b; Fang et al., 2017;  
158 Mousset et al., 2016b). Electrochemical impedance spectroscopy (EIS) using the frequency  
159 range from 50,000 to 0.1 Hz and an amplitude of 10 mV was performed to characterize the  
160 working electrode/electrolyte interface using the equivalent electrical circuit (EEC) method  
161 (Mousset et al., 2016b). The EIS data were fitted using ZSimpWin data analysis software  
162 (AMETEK, Massy, France).

163

### 164 **2.3. Electrochemical setup and procedure**

165 The electrosorption performance achieved with the SA-AC beads working electrode was  
166 tested on a fixed-bed column reactor (2.6 cm internal diameter) (Fig. 1) using a graphite rod  
167 (210 mm length, 12 mm diameter) (Final Advanced Material, Didenheim, France) as a current  
168 collector to connect SA-AC beads (35 g) with the potentiostat in monopolar mode. The  
169 average particle size of the SA-AC beads was 2 mm, and the height of the bed electrode was 9  
170 cm. The counter electrode was a graphite rod (100 length, 8 mm diameter) (Final Advanced  
171 Material, Didenheim, France) and was considered the anode. A nonconductive  
172 polytetrafluoroethylene (PTFE) mesh (with a pore diameter of 1 mm) was placed between the  
173 cathode and the anode to avoid contact in between without interfering with the electrosorption  
174 mechanism. In all experiments, 100 mL of effluent was introduced with ascending flow using  
175 a peristaltic pump (Fisher Scientific, Illkirch-Graffenstaden, France) at a constant flow rate of  
176  $10 \text{ mL min}^{-1}$ . The cathode potential was varied from -0.8 to -1.3 V vs. Ag/AgCl, and the open  
177 circuit (OC) was tested for the blank experiment in the absence of an applied voltage. The  
178 electrosorption capacity ( $q$  in  $\text{mg g}^{-1}$ ) was calculated according to Eq. 2 (Li et al., 2016):

179 
$$q = \frac{(C_0 - C_e)V}{m} \quad (2)$$

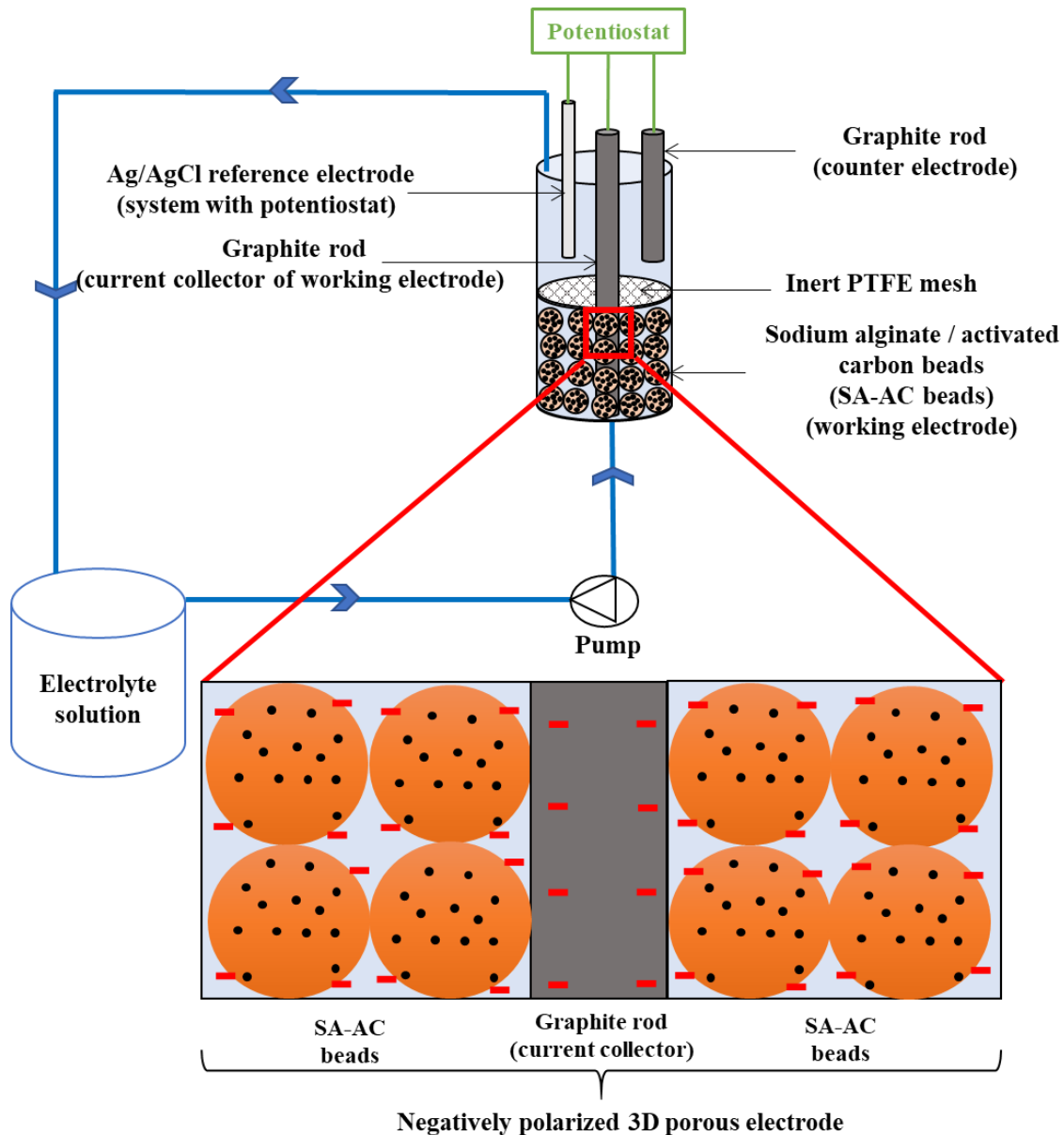
180 where  $C_0$  and  $C_e$  are the initial and equilibrium concentrations of the pollutant, respectively  
181 ( $\text{mg L}^{-1}$ ),  $V$  is the volume of the circulated solution (L), and  $m$  is the adsorbent mass of the  
182 SA-AC electrode (g).

183 The mass transfer coefficients ( $k_m$ ) within the column were determined by LSV using the  
184 limiting-current technique with solutions containing 0.05 M  $\text{K}_3\text{Fe}(\text{CN})_6$ , 0.10 M  $\text{K}_4\text{Fe}(\text{CN})_6$   
185 and 0.50 M  $\text{Na}_2\text{CO}_3$  (Sigma Aldrich, Saint-Quentin-Fallavier, France) (Adnan et al., 2021b;  
186 Cañizares et al., 2006; Selman and Tobias, 1978). The internal ohmic resistances were  
187 determined according to Eq. 3 (Adnan et al., 2022; Miller et al., 2019):

188 
$$\sum RI = E_{cell} - |E_C| - |E_A| \quad (3)$$

189 where  $\sum RI$  is the ohmic drop,  $E_{cell}$  is the cell voltage, and  $|E_A|$  and  $|E_C|$  are the absolute  
190 values of the anode and cathode potentials, respectively.

191 The electroactivity of phenol ( $4 \text{ g L}^{-1}$ ) was assessed by LSV performed from -2 to 2 V vs.  
192 Ag/AgCl at  $0.01 \text{ V s}^{-1}$  scan rate using KCl ( $6 \text{ g L}^{-1}$ ) as the supporting electrolyte. This was  
193 performed to determine whether the direct electroreduction and/or electrooxidation of phenol  
194 could occur.



195

196

**Fig. 1.** Fixed-bed electro sorption schematic setup.

197

198

## 2.4. Modeling

199

A new model was proposed to predict the rate of phenol electro sorption onto beads by

200

incorporating the transport phenomena defined by the Nernst–Planck equation (Jasielec,

201

2021) that include diffusion, electromigration and convection. Electromigration was neglected

202

with the simulated effluent since the phenol molecule ( $pK_a = 9.95$ ) is uncharged at the applied

203

pH of 4. Although the setup is under forced convection within the column, the low flow rate

204 would imply that the diffusion of phenol toward the 3D porous material does not occur in the  
 205 stationary regime. Supposing that the beads are all spherical and have the same diameter ( $\sim$   
 206 3.5 mm), the phenol rate toward the 3D porous material ( $r_c$ ) can obey Fick's law under the  
 207 transient regime according to Eq. 4 (Miomandre et al., 2005):

$$208 \quad r_c = -\frac{1}{S} \frac{dC}{dt} = \frac{1}{V} (C_t - C_t^{el}) \sqrt{\frac{D}{\pi t}} \quad (4)$$

209 where  $D$  is the diffusion coefficient of phenol,  $S$  is the specific surface area of the beads,  $t$  is  
 210 time, and  $C_t$  and  $C_t^{el}$  are the concentrations of phenol in the solution and at the electrode  
 211 surface at time  $t$ , respectively.

212  $C_t^{el}$  can be expressed as a function of  $C_0$ , as follows:

$$213 \quad C_t^{el} = C_0 - C_{t-1} \quad (5)$$

214 where  $C_{t-1}$  and  $C_0$  are the concentrations of phenol at time  $t-1$  and  $t = 0$  (in min),  
 215 respectively, in the discrete model.

216 Then,  $C_t - C_t^{el}$  can be expressed according to Eq. 6:

$$217 \quad C_t - C_t^{el} = C_t + C_{t-1} - C_0 \quad (6)$$

218 After incorporating Eq. 5 into Eq. 4 and integrating Eq. 4 from  $C_t = C_0$  to  $C_t$ , Eq. 7 can be  
 219 obtained:

$$220 \quad \ln(C_t + C_{t-1} - C_0) = \frac{2S\sqrt{D}}{\pi V} t^{\frac{1}{2}} + \ln(C_0) \quad (7)$$

221 When dealing with real effluents (OMWW), a mixture of PCs with different pKa values (3.0–  
 222 4.0 to 10.0–11.0) should be considered (Lissaneddine et al., 2021a). Therefore, the influence  
 223 of the electromigration of ionic PCs must be added to Eq. 4:

224  $r_c = -\frac{1}{s} \frac{dC}{dt} = \frac{1}{V} (C_t - C_t^{el}) \sqrt{\frac{D}{\pi t}} + j_k$  (8)

225 with

226  $j_k = \frac{F}{RT} Z_k D_k C_k E = \frac{F}{RT} Z_k D_k C_k \rho_s j_{app}$  (9)

227 where  $j_k$ ,  $D_k$ ,  $Z_k$  and  $C_k$  are the flux, diffusion coefficient, valence and concentration of the  
 228 charged PCs ( $k$ ), respectively;  $R$  is the gas constant;  $T$  is the absolute temperature;  $F$  is the  
 229 Faraday constant;  $E$  is the electric field;  $\rho_s$  is the solution resistivity; and  $j_{app}$  is the applied  
 230 current density.

231 After integrating Eq. 8 from  $C_t = C_0$  to  $C_t$ , Eq. 10 can be obtained:

232  $t^{\frac{1}{2}} = \frac{X + \sqrt{X^2 - 4YZ}}{2Y}$  (10)

233 with

234  $X = \frac{2s\sqrt{D}}{\pi V}$  (11)

235  $Y = \frac{FZ_k D_k \rho_s j_{app}}{RT}$  (12)

236  $Z = \ln \left( \frac{C_t + C_{t-1} - C_0}{C_0(C_t - C_0)} \right)$  (13)

237 The fittings of the models with experimental results were evaluated with the root mean square  
 238 error (RMSE) as written in Eq. 14 (Adnan et al., 2021a):

239  $RMSE = \sqrt{\frac{\sum_{i=1}^K (y_i - y'_i)^2}{K}}$  (14)

240 where  $y_i'$  is the experimental value,  $y_i$  is the model value and  $K$  is the number of iterated  
241 values.

242 The lower the RMSE value is, the more suitable the model.

### 243 **3. Results and discussion**

#### 244 **3.1. Electrochemical characterization of the 3D porous electrodes**

245 The electrochemical properties of the initial SA-AC beads electrodes were determined to  
246 evaluate their electrochemical performance as an active electrode material. The electroactive  
247 surface area ( $I_0$ ), charge transfer resistance ( $R_{CT}$ ) and electrical double-layer capacitance ( $C_{DL}$ )  
248 values were not impacted by the content of AC in the beads. Therefore, only their average  
249 values were reported.

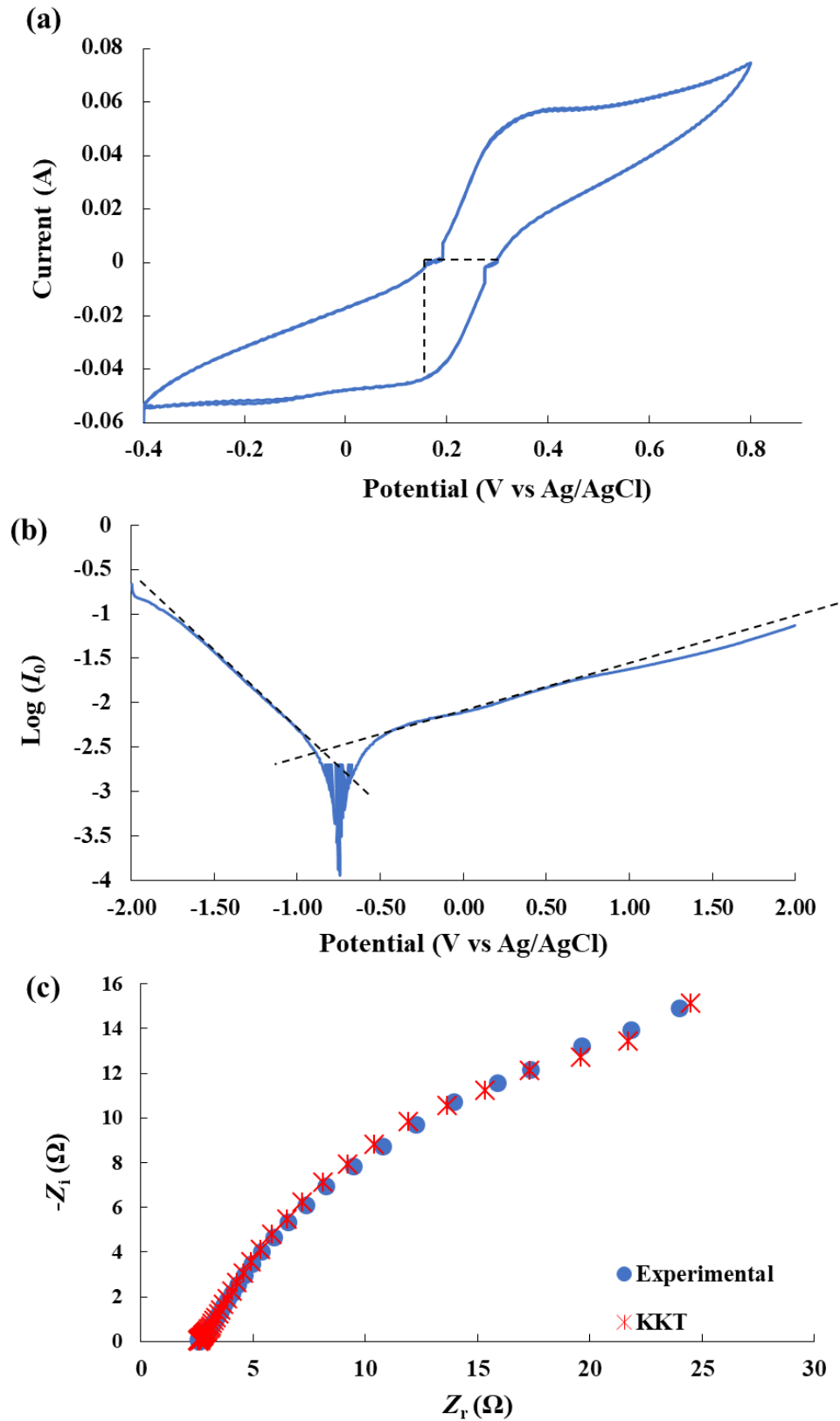
250 The electroactive surface area of porous electrodes determines the number of polarized active  
251 sites, which in turn determines the rate and yield of electrosorption. In addition, the  
252 electroactive surface area is a key parameter for the electrosorption efficiency (Zhu and Zhao,  
253 2019). CV showed that the beads possessed a large electroactive surface area of  $23 \times 10^3 \pm 5.5$   
254  $\times 10^3 \text{ cm}^2$  (Fig. 2a). Comparatively, previous studies estimated surfaces with areas of  $0.14 \text{ cm}^2$   
255 with an oxidized activated carbon electrode (Deryło-Marczewska et al., 2019),  $0.93 \text{ cm}^2$  with  
256 an AC nanotube-supported palladium electrode (Wang et al., 2010) and  $531 \text{ cm}^2$  with an  
257 activated carbon fiber electrode (Ergan and Gengec, 2020). The value obtained with SA-AC  
258 emphasized that the encapsulation of AC in the beads allowed a large electroactive surface  
259 area to be maintained compared to porous electrodes proposed in the literature. This is of  
260 prime interest to achieve high electrosorption capacity. This is in agreement with the  
261 adsorption ability of SA-AC beads determined in a previous study, in which the BET specific  
262 surface area of SA-AC beads was  $523 \text{ m}^2 \text{ g}^{-1}$  and the porous nature of the adsorbent was  
263 shown (total pore volume of  $0.308 \text{ cm}^3 \text{ g}^{-1}$ ), as confirmed by FTIR, SEM and EDS

264 (Lissaneddine et al., 2021a).  $I_0$  was determined to evaluate the catalytic performance of the  
265 electrode and electron transfer across the electrode (Sharma et al., 2013; Situmorang et al.,  
266 2020). The  $I_0$  calculated from the Tafel slopes was 3.24 mA (Fig. 2b). This value was high  
267 compared to the values of electrodes based on other materials described in the literature, such  
268 as activated carbon derived from rice husk (0.06 to 0.17 mA) (Jiao et al., 2020) and  
269 ammonium bicarbonate and activated carbon (0.084 to 1.5 mA) (Li et al., 2014). Knowing  
270 that a higher  $I_0$  leads to a quicker response to a change in potential (Sharma et al., 2013; Sun  
271 and Chen, 2016), better electrode performance is expected.

272 To further characterize the porous electrode/electrolyte interface, EIS data were obtained after  
273 validation of impedance spectra with the linear Kramers-Kronig transform (KKT) technique  
274 (Fasmin and Srinivasan, 2015) (Fig. 2c). The chi-squared residues ( $\chi^2$ ) calculated by using  
275 the KKT technique to fit the experimental spectra and the Randle-type EEC model for fitting  
276 along with KKT were determined to be  $1.5 \times 10^{-5}$  and  $9.3 \times 10^{-7}$ , respectively. Among all the EEC  
277 models tested, the best agreement with EIS spectra was obtained with the Randle-type circuit  
278 model shown in Fig. 3. This model further demonstrated that exchanges with the electrolyte  
279 occur directly within the SA-AC beads, not at the bead and current collector surfaces. This  
280 can also explain why the parameters related to the electrode/electrolyte interface  
281 (electroactive surface area,  $I_0$ ,  $R_{CT}$  and  $C_{DL}$ ) were relatively constant, regardless of the AC  
282 load within the beads. The associated  $R_{CT}$  and  $C_{DL}$  values of the EEC model were  $1.7 \pm 0.7 \Omega$   
283 and  $0.15 \pm 0.084 \mu\text{F cm}^{-2}$ , respectively.  $R_{CT}$  was low compared with  $R_{CT}$  values of other  
284 reported porous electrodes, such as almond shell-based activated nanoporous carbon ( $5.8 \Omega$ )  
285 (Jain and Tripathi, 2014), activated carbon ( $4.2 \Omega$ ) (Tian et al., 2020) and  $\text{Cu}_2\text{O}$ -doped  
286 activated carbon ( $9.39 \Omega$ ) (Zhang et al., 2015). This trend of  $R_{CT}$  compared with other porous  
287 materials corroborated the higher  $I_0$  value in the present case. This would therefore promote  
288 electrosorption on the SA-AC beads electrodes. In addition, a higher  $C_{DL}$  value was found

289 compared to that of activated carbon ( $0.0058 \mu\text{F cm}^{-2}$ ) (Tian et al., 2020), which should  
290 increase the electric double layer and, therefore, the electrosorption efficiency (Lissaneddine  
291 et al., 2021b). This is discussed in the next subsection.



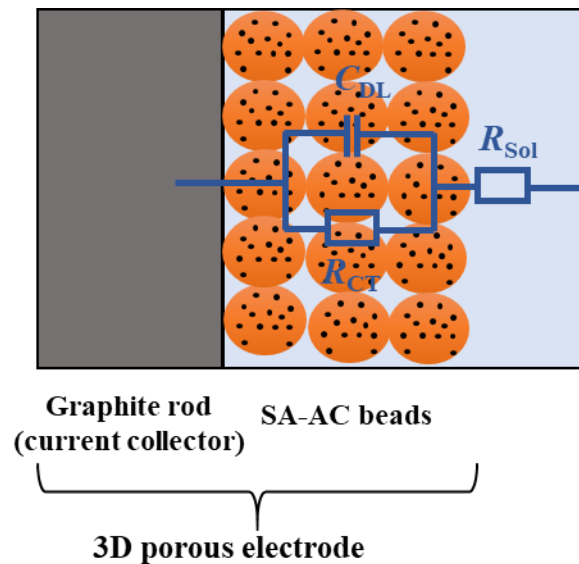


292

293 **Fig. 2.** Electrochemical properties of the prepared SA-AC beads electrode using CV (a), LSV

294

(b) and EIS (c).



295

296 **Fig. 3.** Scheme of the EEC model integrated into the 3D porous electrodes.

297

### 298 **3.2. Electrosorption efficiency**

299

#### 3.2.1. Effect of the iron and calcium contents in the beads

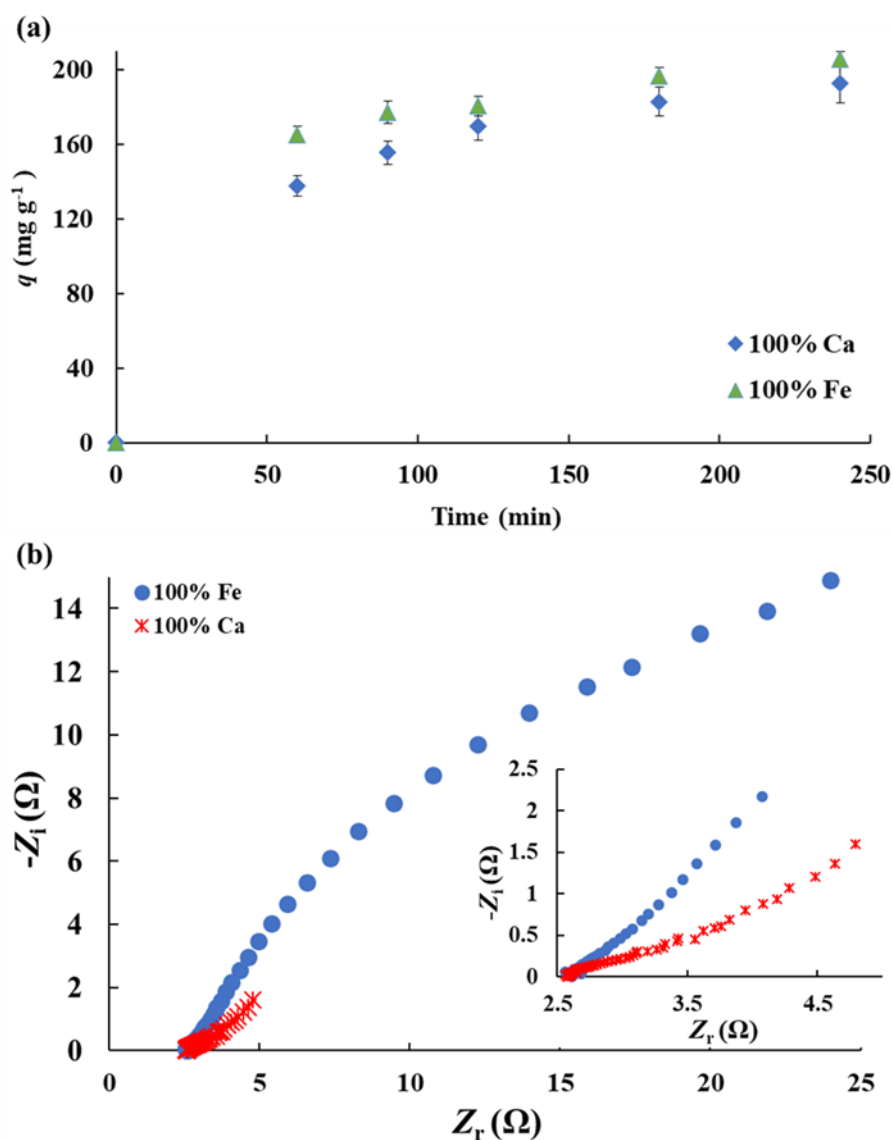
300 To optimize the stability of the bio-sourced adsorbent during electrosorption, iron or calcium  
 301 was added to the beads during preparation (Fig. 4). The results show that the electrosorption  
 302 capacities were similar, on the order of  $204 \pm 3 \text{ mg g}^{-1}$  and  $197 \pm 10 \text{ mg g}^{-1}$  after 240 min for  
 303 Fe and Ca, respectively.

304 To select the optimal element, the electrical conductivities of the two porous electrodes (Fe  
 305 and Ca) were compared by EIS measurements. Fig. 4b illustrates the Nyquist representations.  
 306 The  $R_{CT}$  values were ranked as follows: Fe ( $1.7 \Omega$ ) < Ca ( $2.6 \Omega$ ). The  $R_{CT}$  value of Fe is 1.5-  
 307 fold less than that of Ca, highlighting a slight increase in electrode conductivity achieved with  
 308 Fe. This trend was assumed to be due to differences in electron hopping, polarons and ionic  
 309 radii. The transition energy between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  is 0.2 eV (Iqbal and Farooq, 2007), which  
 310 indicates pronounced electron hopping and, hence, a rapid increase in conductivity with iron.  
 311 Similar results have been observed in a previous study involving doping magnesium

312 aluminate with divalent and trivalent metal ions. The magnesium aluminate doped by  $\text{Fe}^{3+}$   
313 had lower resistivity, i.e., higher conductivity, than that doped with  $\text{Ca}^{2+}$  (Iqbal and Farooq,  
314 2007). This is because the ionic radius of  $\text{Ca}^{2+}$  (0.99 Å) is larger than that of  $\text{Fe}^{3+}$  (0.64 Å) and  
315 the small polaron and electron hopping achieved with iron (Iqbal and Farooq, 2007).

316 This can further explain why in this study, beads prepared with Fe displayed slightly higher  
317 electrosorption capacities. Since the increase in material conductivity has previously been  
318 demonstrated to increase the electrosorption efficiency (Lissaneddine et al., 2021b), the  
319 selected optimal element of crosslinking agent used for beads preparation was Fe.

320



321

322 **Fig. 4.** Effect of the presence of iron or calcium in beads preparation on the electrosorption  
 323 efficiency of phenol (a) and on the electrode conductivity estimated from the Nyquist plot (b).

324 The operating conditions were as follows: anode: graphite rod; cathode: graphite rod and 35 g  
 325 SA-1% w/v AC beads; cathode potential: -1.2 V vs. Ag/AgCl; electrolyte: 6 g L<sup>-1</sup> KCl and 4 g  
 326 L<sup>-1</sup> phenol; flow rate: 10 mL min<sup>-1</sup>; and  $m/V = 0.35$  g mL<sup>-1</sup>.

327

### 328 3.2.2. Influence of AC percentage

329 The influence of AC percentage, i.e., the adsorbent dose, on phenol electrosorption kinetics in  
 330 simulated OMWW is illustrated in Fig. 5a. First, the electrosorption capacity increased

331 rapidly over time as the active sites became accessible. With increasing time, the active sites  
332 tended to be saturated so that the electrosorption capacity slowly increased until adsorption  
333 equilibrium was finally reached. Second, the electrosorption capacity increased for all  
334 percentages of AC tested with increasing time. In addition, there was an increase in  
335 electrosorption capacity with increasing percentages of AC up to 1% w/v (183 mg g<sup>-1</sup>). This  
336 trend was in agreement with the literature, in which the phenol adsorption capacity increased  
337 with increasing powdered AC content in alginate gels (Jodra and Mijangos, 2003). Moreover,  
338 a further increase in the percentage of AC (3% w/v) led to a decline in the electrosorption  
339 capacity of phenol to 178 mg g<sup>-1</sup>.

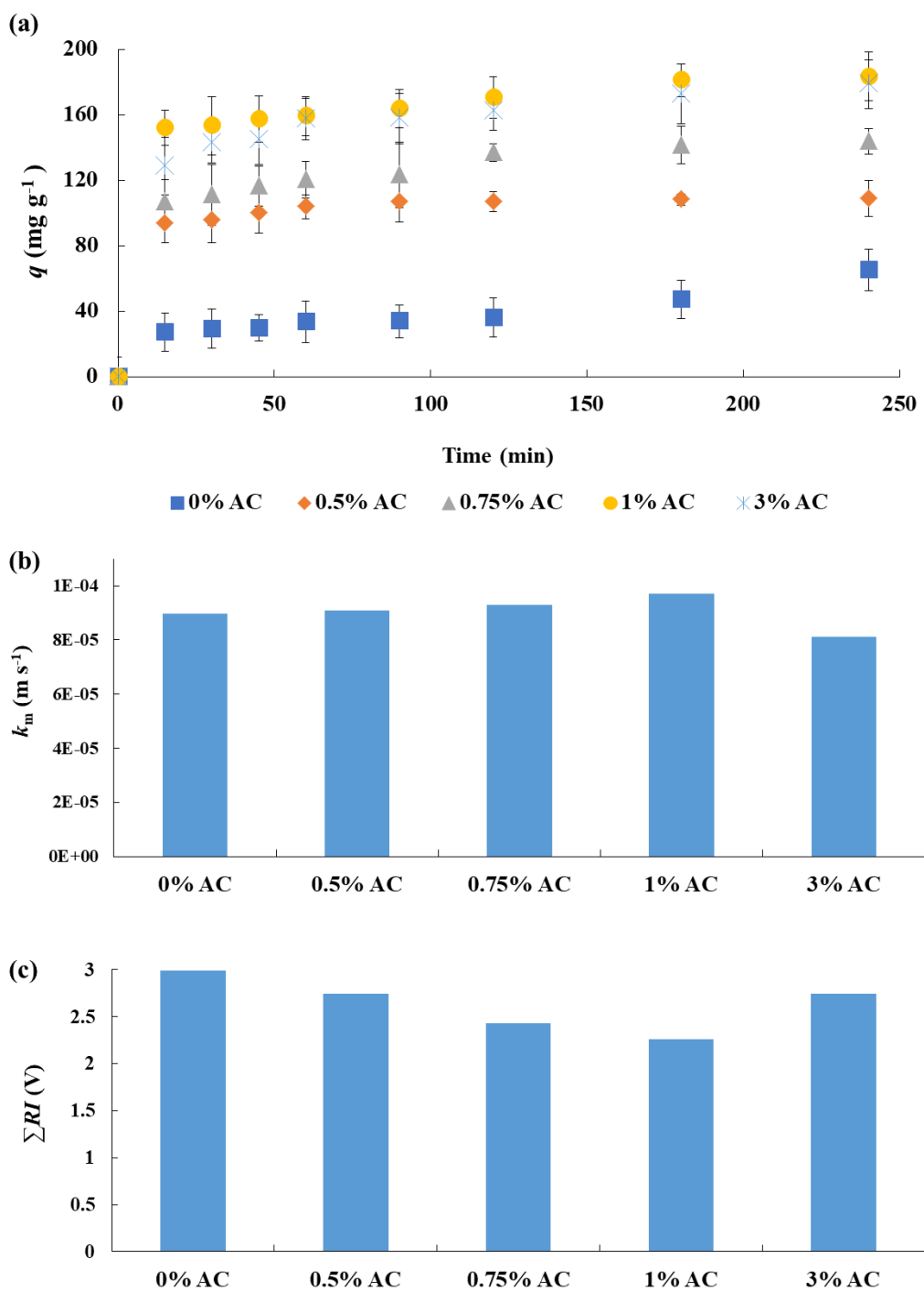
340 Interestingly, the trend of  $k_m$  evolution with AC percentages (Fig. 5b) was similar to the  
341 behavior of phenol electrosorption (Fig. 5a). The  $k_m$  values increased from  $8.9 \cdot 10^{-5} \text{ m s}^{-1}$  for  
342 0% w/v AC to  $9.7 \cdot 10^{-5} \text{ m s}^{-1}$  for 1% w/v AC and decreased to  $8.1 \cdot 10^{-5} \text{ m s}^{-1}$  for 3% w/v AC.  
343 This further confirmed that electrosorption occurred mainly within the beads, as suggested by  
344 the EEC model (Section 3.1), not at the graphite rod and bead interfaces. Moreover, this  
345 suggests that the electrosorption capacity strongly depended on the diffusivity of the phenol  
346 within the beads. It has been previously shown that when AC particles are trapped in SA gels,  
347 the PCs diffuse into the micropores of the SA gels and are adsorbed by the AC enveloped in  
348 the SA matrix (Chung et al., 2003). The increase in the amount of AC could then increase the  
349 diffusivity and, therefore, the mass transport. At an AC content that is too high (3% w/v), AC  
350 may agglomerate, which could lead to a reduction in the elasticity of the hydrogel network  
351 and the formation of a denser network (Thakur, 2018; Tiwari et al., 2012). In a previous  
352 adsorption work, AC agglomerated in the beads, which led to an increase in the diffusional  
353 path length and a decrease in the adsorption capacity (Araga et al., 2017; Iqbal et al., 2016). In  
354 addition, the mass transfer coefficient has been shown to decrease with increasing adsorbent  
355 particle size (Krishna, M. S., Raju, G. J., & Rao, 1966). These statements agree with the

356 decreased electrosorption capacity of phenol at 3% w/v AC. To better visualize the  
357 mechanism of electrosorption with an increase in AC content within the beads, a scheme is  
358 proposed in Fig. 6. This drawing highlights the constant external diffusion layer regardless of  
359 AC content, while the internal diffusion within the beads is affected. Mass transport is then  
360 influenced along with the electrosorption efficiency.

361 Furthermore, increasing the percentage of AC possibly decreased the internal ohmic  
362 resistance of the SA-AC bead electrode from 2.9 V (0% w/v AC) to 2.2 V (1% w/v AC) (Fig.  
363 5c), which subsequently resulted in a higher electrosorption capacity. A further increase in  
364 AC content (3% w/v AC) led to an increase in  $\sum RI$  to 2.7 V. The reasons for this latter trend  
365 could be explained as follows. The mass transport coefficient decreased for 3% w/v AC (Fig.  
366 5b), and according to the results of Fang et al. (2005, 2006), the delayed transport of the ions  
367 into electrode (modified activated carbon aerogel) pores resulted in an increase in the internal  
368 ohmic resistance (Fang et al., 2005; Fang and Binder, 2006). Consequently, the AC  
369 percentage influenced ion transport and the internal ohmic resistance. Bian et al. (2016)  
370 demonstrated that the electrosorption performance significantly improved with a decrease in  
371 the electrode's internal ohmic resistance (14 to 3.6  $\Omega$ ) for inorganic compounds on a  
372 membrane capacitive deionization reactor (Bian et al., 2016). This was further evidenced in  
373 this study for organic compound electrosorption on a fixed-bed column reactor with beads  
374 electrode. To conclude,  $\sum RI$  seemed to be correlated with mass transport as well as the AC  
375 content in the beads and the related electrosorption efficiency.

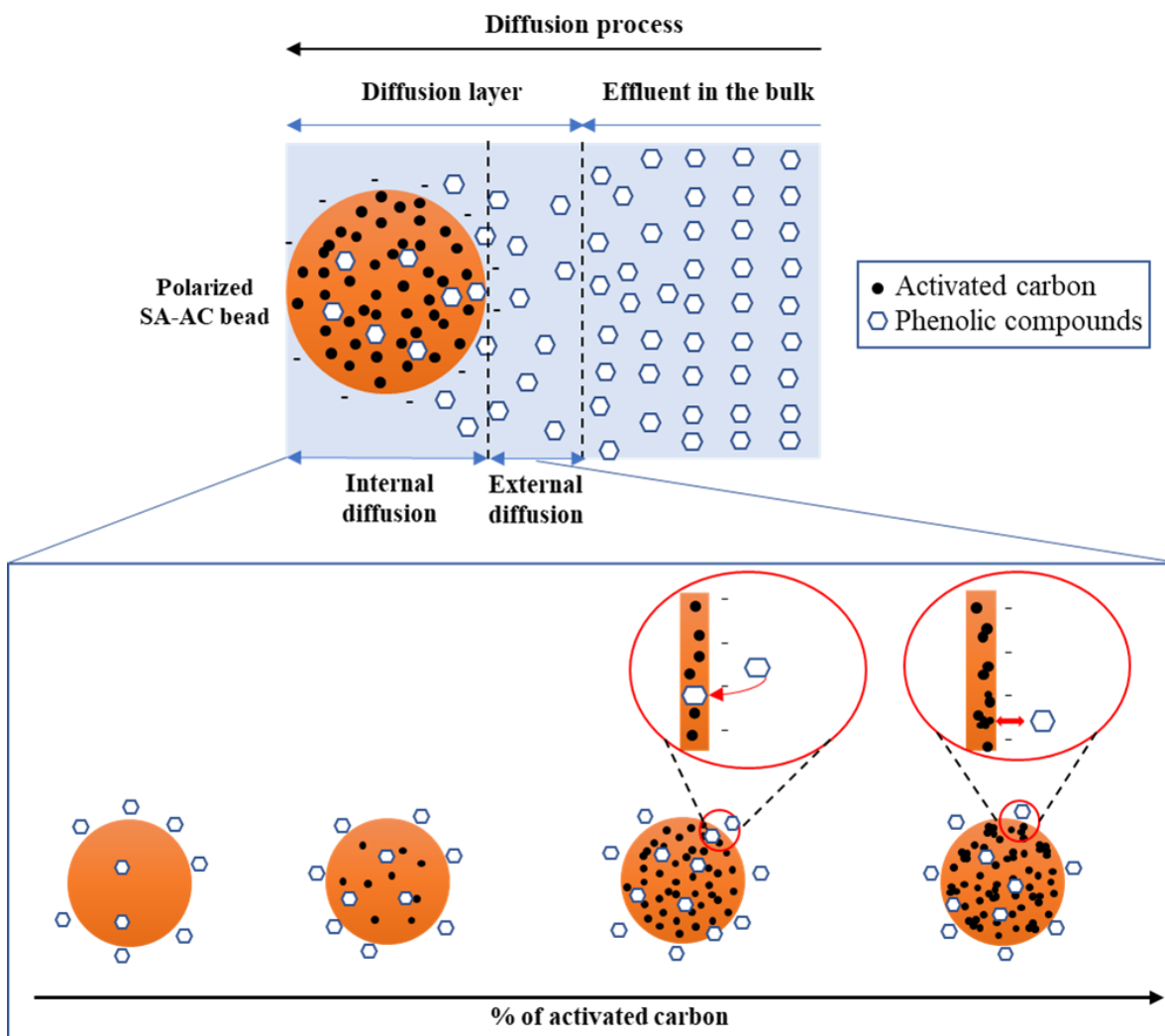
376 Based on these results, the optimum percentage of AC was taken as 1% w/v for the next  
377 electrosorption experiments.

378



379

380 **Fig. 5.** Effect of AC percentage (% w/v) on the evolution of the electroadsorption capacity of  
 381 phenol (a), mass transport (b) and internal ohmic resistance (c). The operating conditions were  
 382 as follows: anode: graphite rod; cathode: graphite rod and 35 g SA (100% Fe)-AC beads;  
 383 cathode potential: -1.2 V vs Ag/AgCl; electrolyte: 6 g L<sup>-1</sup> KCl and 4 g L<sup>-1</sup> phenol; flow rate:  
 384 10 mL min<sup>-1</sup>; and  $m/V = 0.35$  g mL<sup>-1</sup>.



385

386 **Fig. 6.** Mechanism of phenol electrosorption in SA-AC beads with increasing AC content.

387

388

### 3.2.3. Effect of the cathode potential

389 The electrode potential is the driving force of the electrosorption efficiency. Preliminary

390 experiments were performed by applying an anode potential and using the beads as the

391 working electrode. However, the beads could not support the high voltage involved under

392 oxidation conditions, meaning that they were not stable (data not shown). Subsequent

393 experiments were therefore performed by applying a cathode potential. The influence of

394 varying the cathode potential from -0.8 to -1.3 V vs. Ag/AgCl on the phenol electrosorption

395 efficiency compared to the OC condition is shown in Fig. 7. The electrosorption capacity



396 increased with decreasing applied potential from -0.8 to -1.2 V vs. Ag/AgCl, reaching a  
397 maximum electrosorption capacity of 170 mg g<sup>-1</sup> at equilibrium (Fig. 7a). It has been  
398 previously demonstrated that the potential can impact the electrosorption capacity of  
399 uncharged molecules (Gileadi, 1966), which was the case for phenol (pKa = 9.95) considering  
400 a solution pH of 4. However, the electrosorption capacity decreased to 73 mg g<sup>-1</sup> when the  
401 applied potential equaled -1.3 V vs. Ag/AgCl. At this range of cathode potentials, water  
402 (H<sub>2</sub>O) reduction starts to occur according to Eq. (15), in which hydrogen gas (H<sub>2</sub>) is produced  
403 on the surface of the 3D electrode (Ziati and Hazourli, 2019).



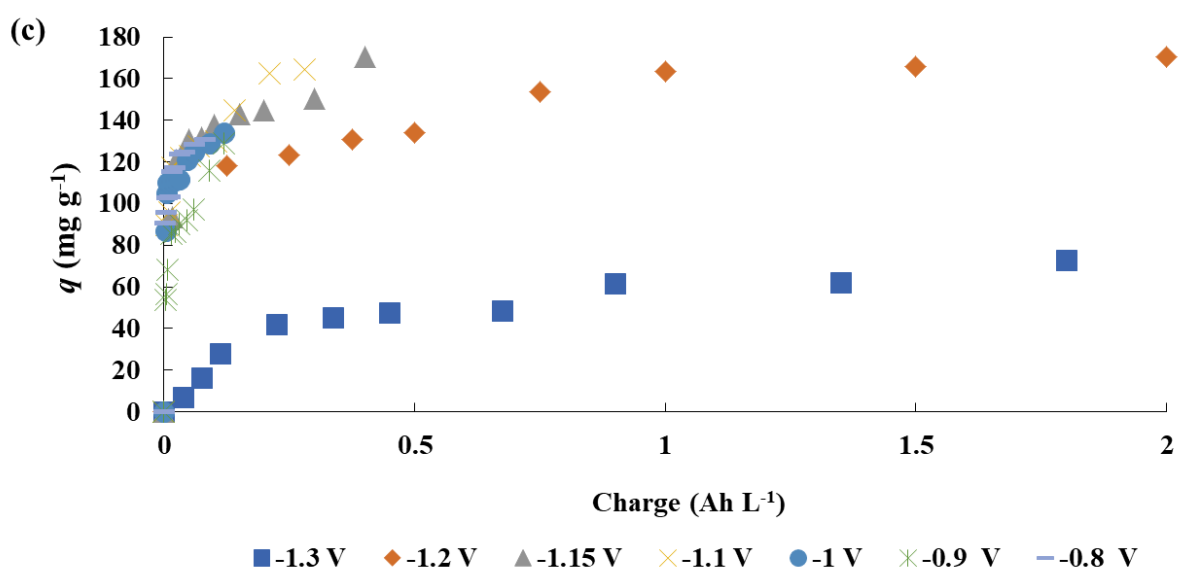
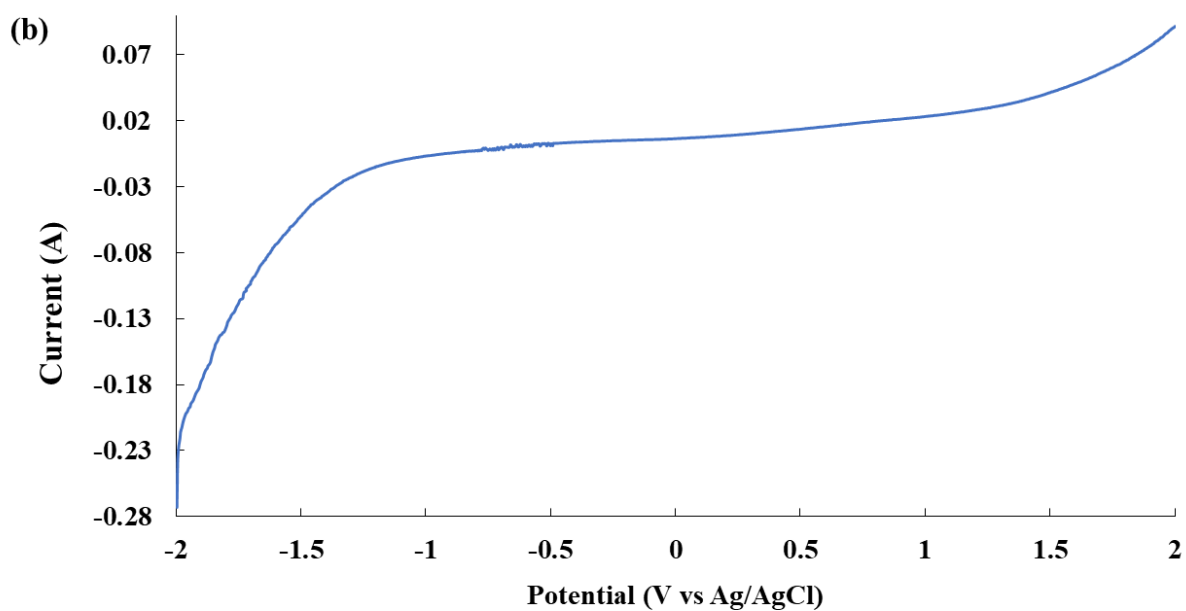
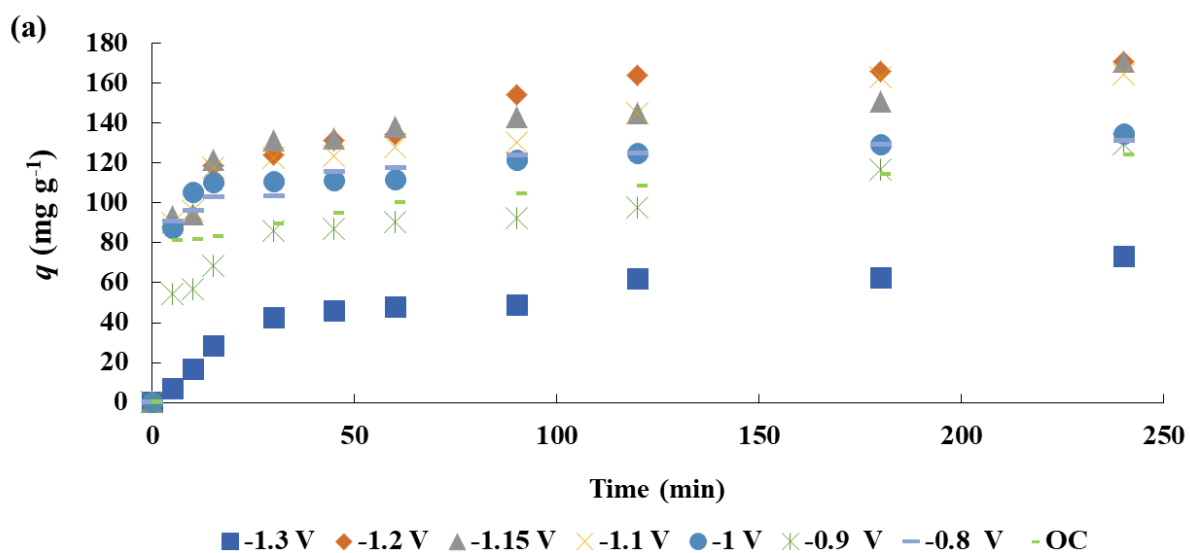
405 This H<sub>2</sub> electrogeneration reaction limits the availability of the active sites on the cathode  
406 surface (Adnan et al., 2021a; Hazourli et al., 1996). The involvement of this faradaic reaction  
407 therefore strongly reduced the electrosorption capacity to 73 mg g<sup>-1</sup>, which was even lower  
408 than that under OC conditions (123 mg g<sup>-1</sup>). In this latter case, the absence of electric current  
409 emphasized the gain (38%) obtained with electrosorption at optimal voltage compared with  
410 the adsorption mechanism alone. To further assess whether electrooxidation or  
411 electroreduction of phenol can occur, an LSV curve was plotted in the presence of phenol and  
412 KCl (Fig. 7b). Neither reduction nor oxidation peaks could be observed between -2 and 2 V  
413 vs. Ag/AgCl. This means that no electron transfer occurred between phenol and the  
414 electrodes. Thus, it was assumed that only capacitive phenomena were involved (i.e.,  
415 electrosorption including adsorption). Uncharged molecules such as phenol at pH 4 can  
416 electrosorb because electrode polarization induces the replacement of solvent molecules with  
417 the organic compound at the interface (Gileadi, 1966). This enhances electrosorption (Gileadi,  
418 1966).

419 The energy requirement is an important economic parameter to consider in electrosorption  
420 process optimization. Considering the electrosorption efficiency as a function of specific  
421 charge values (Fig. 7c), decreasing the applied potential increased the specific charge. The  
422 maximum electrosorption capacity (approximately  $170 \text{ mg g}^{-1}$ ) was only reached after 0.28  
423  $\text{Ah L}^{-1}$  at  $-1.1 \text{ V vs. Ag/AgCl}$ , compared to  $2 \text{ Ah L}^{-1}$  at  $-1.2 \text{ V vs. Ag/AgCl}$ . This means that  
424 the energy requirement would be 7 times lower at  $-1.1 \text{ V vs Ag/AgCl}$ .

425 Thus, the applied cathode potential was  $-1.1 \text{ V vs. Ag/AgCl}$  for the next electrosorption  
426 experiment with real OMWW.

427

428



430 **Fig. 7.** Effect of cathode potential (V vs. Ag/AgCl) on phenol electrosorption considering the  
431 contact time (a), LSV with SA-AC beads as the working electrode (b), and effect of cathode  
432 potential on specific charge (c). Operating conditions: anode: graphite rod; cathode: graphite  
433 rod and 35 g SA (100% Fe)-1% w/v AC beads; electrolyte: 6 g L<sup>-1</sup> KCl and 4 g L<sup>-1</sup> phenol;  
434 flow rate: 10 mL min<sup>-1</sup>; and  $m/V = 0.35$  g mL<sup>-1</sup>.

435

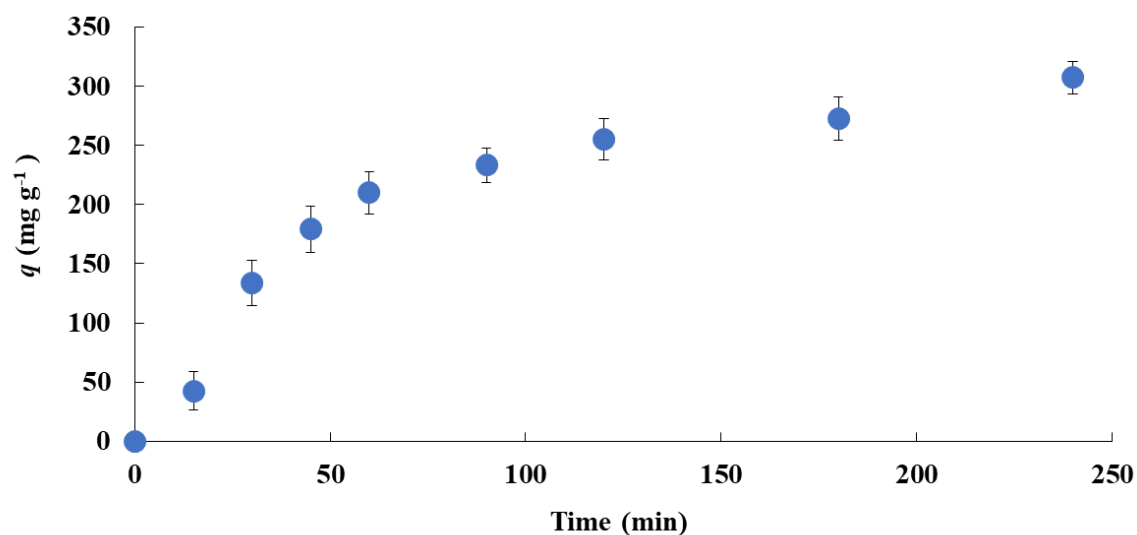
436

#### 3.2.4. Multicomponent effect with actual effluent

437 Electrosorption experiments with real OMWW were carried out under optimal conditions: 1%  
438 w/v AC, 100% w/v Fe and -1.1 V vs. Ag/AgCl cathode potential (Fig. 8). The results showed  
439 that the maximum electrosorption capacity of PCs was higher (307 mg g<sup>-1</sup>) than that with  
440 phenol solutions (164 mg g<sup>-1</sup>) in simulated OMWW. This difference could be explained by  
441 the presence of multiple PCs with different physico-chemical properties, such as diverse pKa  
442 values. The main PCs identified in actual OMWW have been listed in a previous paper  
443 (Lissaneddine et al., 2021a), and their pKa values ranged from 3.0-4.0 to 10.0-11.0. Since the  
444 pH of the solution was between 4 and 5, the compounds, whose pKa values are equal to or  
445 below this pH value, predominantly exist in their acidic form. In contrast, the PCs, whose pKa  
446 is higher than the solution pH, primarily exist in their basic form. In addition, it is usually  
447 assumed that the acidic forms of the PCs remain uncharged because the hydroxyl (OH/O<sup>-</sup>) and  
448 carboxyl (COOH/COO<sup>-</sup>) groups are protonated. In contrast, their basic forms are ionized since  
449 these groups are deprotonated (Baransi et al., 2012; Liu et al., 2020). The charged PCs were  
450 therefore subjected to electromigration in addition to diffusion near the electrode. Although  
451 the SA-AC beads surfaces was negatively charged (cathode), electrostatic repulsion between  
452 the negatively charged PCs and the negatively charged SA-AC beads surfaces could still  
453 promote contact with the beads (Parida and Pradhan, 2010). The electrostatic repulsion could  
454 also be explained by the molecular complexation of PCs in OMWW. The polarizability of

455 PCs depends on the combination of dispersion interactions and hydrophobic effects, which is  
456 the main driving force for the molecular complexation of PCs in OMWW (Bouknana et al.,  
457 2019). In addition, since the hydraulic flow of the negative fixed-bed electrode is less than  
458 that of the positive counter electrode, electromigration can also favor the contact of PCs  
459 within the fixed-bed electrode before reaching the anode. Therefore, the electrosorption rates  
460 and yields were maximized. The influence of electromigration was previously observed with  
461 other organic compounds (e.g., acid red 88, orange II, and methylene blue) in simulated  
462 effluents (Lissaneddine et al., 2021b; Sun et al., 2016; Yue et al., 2019b). The present results  
463 newly demonstrated the influence of electromigration with a multicomponent system  
464 containing organics in an actual effluent.

465 Based on this trend, the SA-AC beads electrode can be used to separate not only phenol but  
466 also other PCs (47%) from actual OMWW by electrosorption for subsequent valorization in  
467 future research studies.



468  
469 **Fig. 8.** Effect of contact time on the electrosorption of PCs from real OMWW under optimal  
470 operating conditions. Operating conditions: anode: graphite rod; cathode: graphite rod and 35

471 g SA (100% Fe)-1% w/v AC beads; cathode potential: -1.1 V vs. Ag/AgCl; flow rate: 10 mL  
472 min<sup>-1</sup>; and  $m/V$  ratio = 0.35 g mL<sup>-1</sup>.

### 473 **3.3. Theoretical evolution of organic compound electrosorption in** 474 **simulated and actual effluents**

475 The models developed in Section 2.4 were used to predict the electrosorption evolution of  
476 phenol in simulated OMWW as a function of AC content (Fig. 9a) and PCs in real OMWW  
477 (Fig. 9b). All the RMSE values were lower than  $2.95 \times 10^{-2}$ , which confirmed the good  
478 agreement of the model with the experimental data. Thus, diffusion in simulated effluent and  
479 diffusion along with electromigration in real effluent are the main phenomena involved in the  
480 kinetics of organic compound electrosorption.

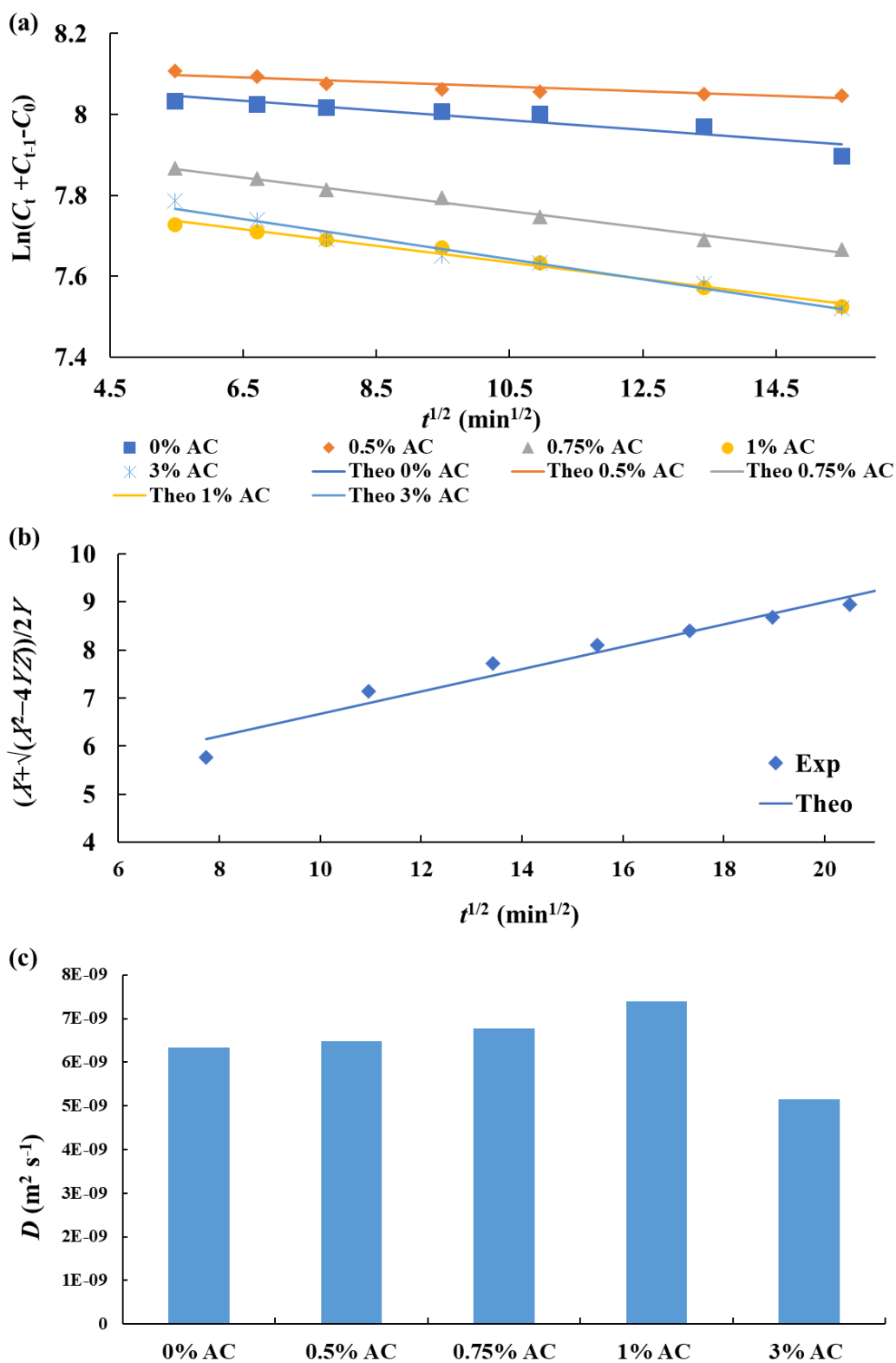
481 The good fitting with real effluent data further supported the estimated average valence of  
482 ionic PCs, which was 3.8. This value was obtained by weighting the valence of each PC  
483 charged in solution at a pH of 4.8 based on the ratio of their basic form to their average  
484 concentration in real effluent. The main ionic PCs identified in actual OMWW were gallic  
485 acid (valence of 5), ferulic acid (valence of 5), caffeic acid (valence of 4), cinnamic acid  
486 (valence of 2), and coumaric acid (valence of 3) according to a previous study (Lissaneddine  
487 et al., 2021a).

488 Furthermore, based on the slope values  $(\frac{2S\sqrt{D}}{\pi V})$  of  $\ln(C_t + C_{t-1} - C_0) = f(t^{1/2})$  (Eq. 7), the  
489 diffusivity constant for phenol could be estimated as a function of AC content ( $D_{X\%}$ ) as  
490 follows (Eq. 16):

$$491 \quad D_{X\%} = \frac{\pi V^2}{4S^2} \quad (16)$$

492 The evolution of  $D_{X\%}$  as a function of AC content is plotted in Fig. 9c. The trend followed  
493 that of the electrosorption capacity of phenol at different AC percentages (Fig. 5a), i.e.,

494 optimal diffusion ( $7.3 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$ ) at 1% w/v AC. This was also consistent with the  $k_m$  value  
 495 evolution (Fig. 5b). This further corroborated the fact that the electrosorption capacity  
 496 strongly depended on the diffusivities of the PCs within the beads.



498 **Fig. 9.** Experimental (symbol) versus theoretical correlations (continuous line) as a function  
499 of  $t^{1/2}$  in simulated OMWW as a function of AC content (a) and PCs in real OMWW and (b)  
500 effect of the percentage of AC on the diffusion coefficient (c).

## 501 **Conclusions**

502 This study proposed a novel approach to remove PCs from simulated and actual OMWW by  
503 implementing electrosorption onto SA-AC beads as porous bio-sourced electrodes. The  
504 impacts of key parameters (crosslinking agent (iron and calcium), AC content and cathode  
505 potential) on the removal efficiency of PCs were explored. The main new insights are listed as  
506 follows:

- 507 • The higher conductivity obtained with the Fe crosslinking agent led to a slightly  
508 higher electrosorption efficiency than that obtained with Ca.
- 509 • The AC content within the beads impacted the mass transfer and diffusion coefficients  
510 as well as the internal ohmic resistance. An optimal AC amount was observed (1%  
511 w/v) because an excessively high concentration implied agglomeration.
- 512 • A cathode potential of -1.1 V vs. Ag/AgCl was found to be optimal considering the  
513 electrosorption capacity and energy requirements. Too high voltage leads to faradaic  
514 reactions that hamper the electrosorption mechanism. Electrosorption was better than  
515 adsorption efficiency, while uncharged phenol electrosorption could still occur.
- 516 • The electrosorption efficiency was higher with real wastewater than with simulated  
517 effluent. This could be due to the presence of multiple organic components that have  
518 different pKa values.
- 519 • New discrete models were proposed to understand and predict the electrosorption  
520 efficiency. The kinetics were mainly limited by diffusion in the transient regime in the



521 synthetic effluent, while the electromigration of ionic PCs also impacted  
522 electrosorption with real OMWW.

523 • The results of the EEC model along with the kinetic models confirmed that  
524 electrosorption mainly occurred within the beads, not at the 3D electrode/electrolyte  
525 interface, unlike what is often assessed.

526 The good electrosorption efficiency obtained for PCs removal from OMWW is promising for  
527 PCs recovery for subsequent valorization. The reusability of porous electrodes is also an  
528 important indicator when evaluating the electrosorption performance of materials. Since the  
529 organic compounds were trapped in the beads, electrosorption may be more irreversible and,  
530 therefore, desorption may be less easy. This is the subject of incoming research that will  
531 provide a more detailed evaluation of the particle cathode stability and reusability. A further  
532 possibility is to combine electrosorption for recovery with a degradation process for the  
533 elimination of unwanted compounds, which is also the subject of ongoing research.

534

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542

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