

1 **Implications of advanced wastewater treatment: electrocoagulation**
2 **and electroflocculation of effluent discharged from a wastewater**
3 **treatment plant**

4
5 Farooq Sher^{a,*}, Kashif Hanif^a, Sania Zafar Iqbal^b, Muhammad Imran^c

6 *a. School of Mechanical, Aerospace and Automotive Engineering, Faculty of Engineering,*
7 *Environmental and Computing, Coventry University, Coventry CV1 2JH, UK*

8 *b. Department of Biochemistry, University of Agriculture, Faisalabad, 38000, Pakistan*

9 *c. School of Engineering and Applied Science, Aston University, Aston Triangle, Birmingham B4*
10 *7ET, UK*

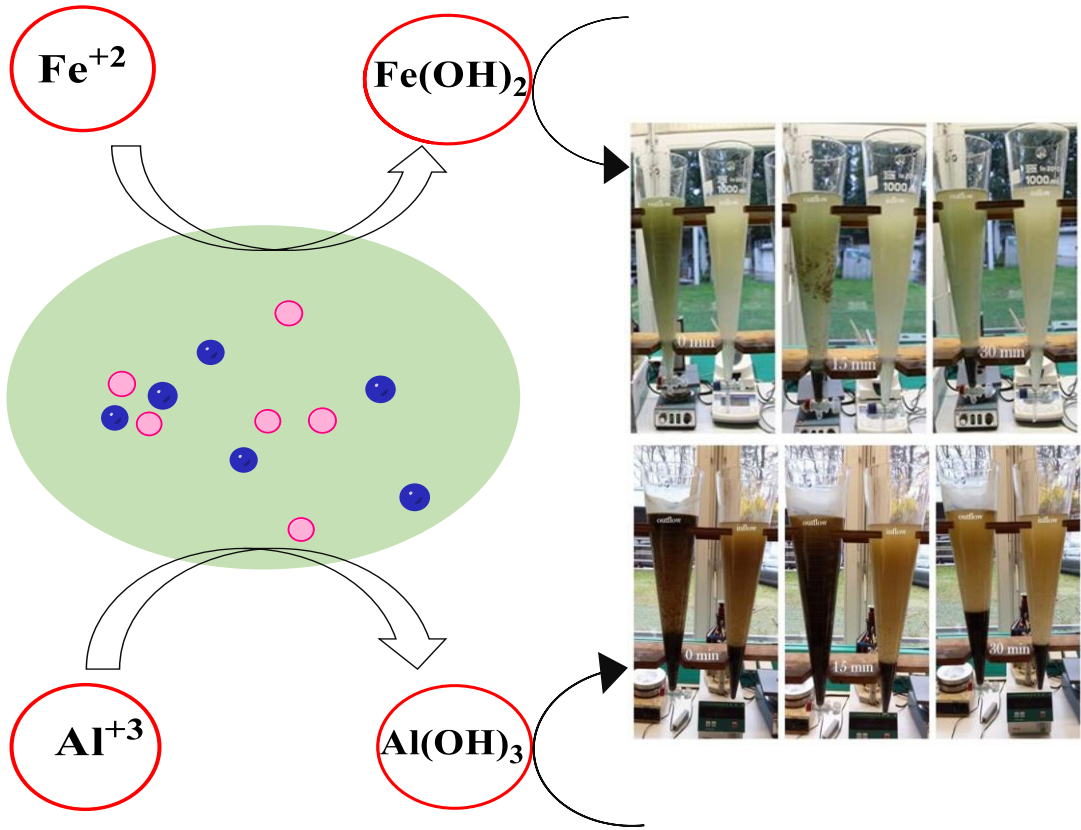
11
12 **Highlights**

- 13 • Pilot plant electrocoagulation (EC) reactor used for advanced wastewater treatment.
- 14 • Al and Fe electrodes were used for primary and secondary tank effluent treatment.
- 15 • Applied current values from 1–4 A in the case of COD removal by Fe and Al.
- 16 • Calculated amounts of removed micro-pollutants and the coagulants in agglomerates.
- 17 • The Al electrodes have produced more amounts of flocs and bubbles.

18

* Corresponding author:
E-mail address: Farooq.Sher@coventry.ac.uk (F.Sher)
Tel.: +44 (0) 24 7765 7754

19 **Graphical Abstract**



20

21

22 **Abstract**

23 In this research, wastewater treatment was inspected on a pilot-scale wastewater treatment plant
24 by electrochemical techniques, electrocoagulation (EC), electroflotation (EF) and electrophoretic
25 deposition (EPD). The wastewater samples have been characterised by applying different
26 parameters to determine the optimum working conditions of the electrocoagulation reactor. Two
27 electrodes have been tested separately with an outflow coming from primary and secondary
28 sedimentation tank. The outflows from these tanks are introduced in EC reactor then EC reactor
29 efficacy is determined for the removal of chemical oxygen demand (COD), suspended solids,
30 micropollutants and amount of coagulants in agglomerates at different current densities. The
31 amounts of suspended solids (SS) in influent and effluent streams were determined by the
32 membrane filtration technique. The operational applied current values range from 1–4 A in the
33 case of COD removal by Fe and Al. While for SS aggregation the applied current ranges from 0.5–
34 3 A and inflow rate was tested from 250 to 500 L/h. The pH of outflows increased by increasing
35 applied current and both of these parameters were found a positive increase in the amount of SS
36 aggregations after EC treatment. The COD removal efficiency was found to be 56–57% and 12–
37 18% in case Fe and Al electrode respectively after EC treatment. Furthermore, the results showed
38 that applied current is the most effective parameter, whereas the aluminium electrodes have
39 produced more amounts of flocs and bubbles in comparison to iron electrodes at a similar amount
40 of current density.

41

42 **Keywords:** Advanced wastewater treatment; Electrocoagulation; Micropollutants; Suspended
43 solids; Pollution and Cost analysis.

44 **1 Introduction**

45 The removal of anthropogenic micropollutants emitting from industrial, agricultural, domestic and
46 urban sources is one of today's major challenges. The number of such micropollutants is
47 significantly large and many of these are found in excessive quantities. Furthermore, pathogenic,
48 non-pathogenic organisms, pharmaceutical and drug residues viruses and vaccines present in large
49 amount in wastewaters. These micropollutants can have direct and indirect effects on the living
50 organisms by bio magnification along the food chain. All mentioned sources of wastewater
51 produce a huge amount of pollutants; organic carbon, biochemical oxygen demand (BOD) and
52 chemical oxygen demand (COD), suspended solids (SS), total nitrogen and phosphorus contents
53 [1, 2]. The main treatments till now available for the removal of these anthropogenic pollutants
54 and micropollutants involve aerobic biodegradation, filtration, flocculation, adsorption, froth
55 floatation and electrocoagulation (EC) [3]. However, some other methods have been applied in
56 combination to overcome the shortcomings of an individual process including; photo-electro-
57 Fenton, electro-Fenton and electro-oxidation [4]. EC is a highly studied process in this field these
58 days. It is a multistep process, which involves the agglomeration of contaminants via electrodes
59 (coagulant) through redox reaction carried out by applying an electric current. Conventional
60 electrocoagulation process was carried out with the help of inorganic chemical species as
61 coagulants including $(\text{NH}_4)_2\text{SO}_4$ and FeCl_3 .

62
63 Various other electrochemical and photo-assisted electrochemical processes have been examined
64 for the purification of wastewater [5], however, preference is given to the EC method. The EC
65 process proceeds by electric current using metallic electrodes. Therefore, it is an electrochemical
66 process being used for the removal of contaminants from wastewater [6, 7]. It involves the

67 conversion of hazardous organic pollutants of wastewater via redox reactions to non-hazardous
68 materials. Furthermore, EC process is famous as a green technology because of its simple
69 assembly, safety, short reaction time, selective capacity, negligible usage of chemicals, easy to
70 handle/ operate and good water purification efficiency [7, 8]. The electric current (I) supplies the
71 necessary force (electromotive) to drive redox reactions, resultantly the particulate/ contaminant
72 reaches to a stable state (solid). That is relatively less emulsifiable, less soluble and less colloidal
73 in comparison to the equilibrium values. Afterwards, the stable solids convert into hydrophobic
74 compounds/ precipitates that can be easily separated out by different separation techniques. The
75 EC method uses a negligible amount of chemicals, therefore no need for neutralization reaction as
76 there is no secondary contamination [9].

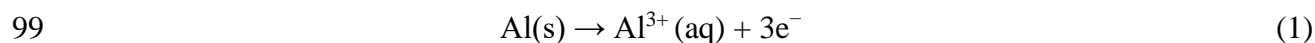
77

78 Three stages of EC technology implicate in the purification of wastewater are as: (1) electrolytic
79 oxidation of sample water to generate coagulants; (2) pollutant destabilization, emulsion,
80 deterioration and particle suspension; (3) agglomeration of resultant particles to generate flocs:
81 comprised of colloids entrapped sludge blanket formed from coagulation reaction [10]. These
82 flocs are similar to chemical flocs and are larger, acid-resistant, bounded less water and stable,
83 that's why can be easily separated out by rapid sand filtration [11, 12]. The EC process is
84 economical as it produces relatively less amount of sludge in comparison with conventional
85 processes [13]. The electrochemical reactions take place at anode and cathode as discussed in
86 equations (1–6). During EC, H_2 is evolved at cathode in the form of bubbles, that discards particles
87 by flotation known as electroflotation [14]. Furthermore, numerous hydroxide of metallic
88 electrodes (anode: Fe/ Al) and coagulant compounds are generated from these chemical reactions
89 to deteriorate coagulate and adsorb pollutants [11]. Aluminium and iron electrodes are widely used

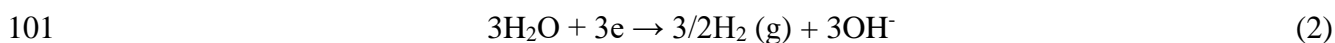
90 in the literature for wastewater treatment because of their increased ions production potentials [15,
91 16]. The cations of iron (Fe^{+2}) and aluminium (Al^{+3}) dissolve from the relevant anode in aqueous
92 are shown in reaction 1 and 4. Moreover, Al^{+3} and Fe^{+2} cationic species may form a number of
93 monomeric and polymeric hydroxide entities during the course of electrochemical reactions
94 relevant to each electrode. The transformation of metallo cationic species, into $\text{Al}(\text{OH})_3$ (reaction
95 3) and $\text{Fe}(\text{OH})_2$ (reaction 6) hydroxides is pH-dependent. These final species act as micropollutant
96 adsorbents.

97 Aluminum electrode reactions during EC:

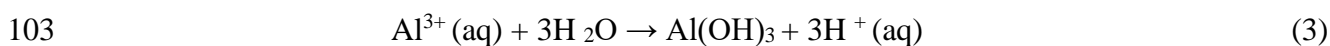
98 Anode:



100 Cathode:

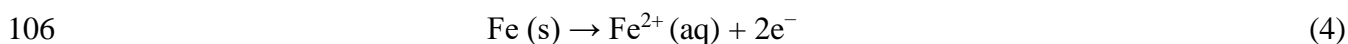


102 Overall:

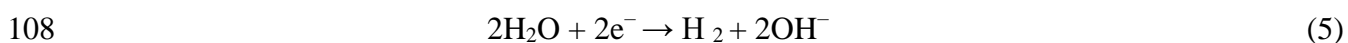


104 Iron electrode reactions during EC:

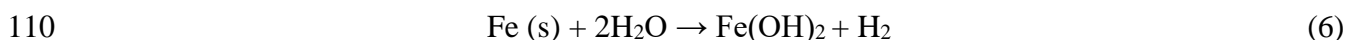
105 Anode:



107 Cathode:



109 Overall:



111

112

113 Mostly efficiency of different electrodes including steel [17], Al [7], Mg [14] and Fe [18] has been
114 evaluated for the removal of COD, colour, total phosphorus and ammonia at different pH and
115 different values of current densities. However, there is a lack of information regarding a complete
116 study on a suitable EC reactor for the optimisation of different operating parameters, coagulant's

117 nature effect on flocs generation and calculation of the amount of suspended solids and
118 micropollutants using advanced scale wastewater treatment plant. Therefore, this study is of much
119 worth, as it is about the testing of a pilot-scale plant EC reactor for the removal of suspended solids
120 and micropollutants from an effluent discharged from a wastewater treatment plant using green
121 technology. Two types of effluents from primary and secondary sedimentation tank have been
122 used. Membrane filtration technique has been applied for the calculation of SS in effluents.
123 Moreover, different flow rates and current densities have been used to determine the optimum
124 conditions and limitations of the pilot scale EC reactor. Furthermore, operational cost analysis of
125 the EC reactor is performed to verify the feasibility of the designed test EC reactor plant for
126 micropollutants removal from the wastewater.

127 **2 Experimental**

128 **2.1 Wastewater sampling and characteristics**

129 Wastewater samples were collected from a local wastewater treatment plant. Mainly two types of
130 samples depending upon their quality are used in the tests; one type was taken from the outflow of
131 primary sedimentation tank and the other was taken from the secondary sedimentation tank of the
132 wastewater treatment plant. Primary tank effluent has a higher amount of pollutant in comparison
133 to secondary tank effluent. The wastewater characteristics of these samples are discussed in Table
134 1. Inflow and outflow points are situated on the EC reactor from where samples were collected
135 after constant conditions of operation at EC setup. Samples were not collected at once but with
136 constant short intervals of time to ascertain the homogenous composition of the samples.

137 **2.2 Experimental assembly of EC reactor**

138 The experimental setup is schematically shown in Fig. 1. A 150 L open tank is used as a feed tank.
139 Some of the pollutants like suspended solids (SS) might settle during an experimental run. Hence,
140 the feeding of 150 L tank is equipped with a stirrer for continuous stirring and maintaining a
141 consistent influent to the treatment system. The feed tank is fed with primary or secondary
142 sedimentation tank effluent one by one according to the experiment by changing the feed lines.
143 The effluent from feed tank is fed to the EC reactor using a pump (range 250–1500 L/h) and a flow
144 meter connected in line to ensure a constant inflow (500 L/h) to the EC reactor. The pilot plant
145 setup is mainly divided into two main parts. The first part consists of an EC reactor and second
146 part consists of EF setup and EPD discs. The EC reactor is a fibre box having two chambers in
147 series provided with slits to insert metal plates inside that act as electrodes. Each cell has 17 plates
148 and as a whole, there are 34 plates of one material (either Fe or Al) used at a time in the EC reactor.
149 The slits inside the EC cells are fitted with plastic cylindrical objects of about 3 cm in length and
150 3 mm in diameter to raise the height of the plates to ensure better operation. The plates act as
151 bipolar electrodes in a parallel connection and are connected to a DC power supply through a
152 polarity changer circuit. The polarity changer circuit has various time settings to change the
153 polarity on the connecting electrodes that ensure proper functionality of electrodes. In bipolar
154 systems, the side of electrode facing anode is negatively charged and vice versa on the other side
155 of cathode. In EC reactor, there is an inlet and outlet, the inlet is just a circular hole where the
156 connecting pipe fits in to provide inflow to the reactor.

157

158 The outlet of reactor is provided with an overflow rectangular opening and a circular hole at the
159 bottom for outflow. The EC reactor is provided with an auxiliary plastic glass cover to ensure a

160 better view of EC process inside the reactor and controlling parameters by visually viewing the
161 process. The plates used inside the EC reactor are 300×155×3 mm of dimensions with an effective
162 surface area of about 1.488 m² [3]. In experiments, Al and Fe electrodes are used separately by
163 applying different values of current densities. The current density applied for the Fe and Al
164 electrodes ranges from 0.67 to 2.69 A/m² and 0.34 to 2.02 A/m² respectively. The amount of
165 suspended solids in the case of Al electrodes, agglomeration was carried out nicely with lower
166 current densities. However, for Fe electrodes higher current densities were required for noticeable
167 treatment. In the presence of these observations and due to economic factors, lower current
168 densities were applied for Al electrodes treatment. The inflow to electrocoagulation reactor was
169 maintained at about 500 L/h and for some reactions at 250 L/h. The pH of inflows was maintained
170 by using NaOH and H₂SO₄ where required. The pH of effluents was examined with the help of a
171 pH meter (Hanna Ins. 301). A typical pH meter consists of a special measuring probe (a glass
172 electrode) connected to an electronic meter that measures and displays pH reading. There is a bulb
173 at the bottom of the probe; the bulb is a sensitive part of the probe that contains a sensor. To
174 measure the pH of a solution, the probe is dipped into the solution. The probe is fitted in an arm
175 known as probe arm.

176 **2.3 Characterisation of wastewater samples**

177 Wastewater from primary sedimentation tank was treated with Fe and Al bipolar electrodes to
178 examine the COD reduction efficacy of the test pilot plant at above mentioned variable current
179 densities [6]. The COD of samples were calculated with the help of spectrophotometer (Hach Dr
180 5000, USA). While samples collected from the secondary sedimentation tank were examined for
181 their amount of SS substances before (inflow) and after (outflow) EC treatment. The outflow of
182 secondary tank has not been tested for COD removal, because of the lower level of COD. However,

183 the effluent from primary sedimentation tank is treated at higher current densities to see the effect
184 of electrocoagulation on COD removal efficiency. Similar to COD measurements, the amount of
185 SS substances in samples were inspected by using separate Fe and Al coagulant, at different current
186 densities and resultant pH values to identify the optimum conditions at which EC set-up could
187 work efficiently. The measurement of SS substances in the samples was done by using a membrane
188 separation technique, according to DIN 19643-2 that is called after filtration solids (AFS) [19].

189

190 The filtration was done by a pre-weighed filter for a specific volume of each sample at a specific
191 nitrogen pressure of 5 bars. During filtration, the filter paper allows only particles, which have a
192 diameter, less than 0.45 μm . After filtration, the wet filter paper is again dried in an oven for 1 h
193 at 105 °C temperature. It evaporates all the water content and collects only the solids with it, which
194 needs to reside in the desiccator to cool it down for 30 min. Finally, by measuring the weight of
195 filter paper, total SS were calculated in the treated wastewater. Each sample was subjected to AFS
196 setup three times and an average value was taken for experimental calculations. The AFS of inflow
197 and outflow samples were made to compare the quantitative analysis on the basis of flocs
198 formation and coagulant dissolution.

199

200 After AFS calculations, the amount of coagulants dissolved during the generation of flocs was
201 calculated experimentally and theoretically to observe the reliability of the process. The
202 experimental calculations of the amount of Fe and Al coagulation dose in outflows in the case of
203 present pilot plant set up was a major task of this study. For the total amount of Fe in the outflow,
204 the spectrophotometric analysis was performed while for Al in outflow, the gravimetric analysis

205 was done [20]. Moreover, the theoretical amount of Fe and Al coagulation dose was calculated
206 according to Eq. (1) [21].

207

$$208 \quad C_x = \frac{M_{\text{total},x}}{V_{\text{total,liquid}}} = \frac{I \times t \times m_x \times N}{Q \times t} = \frac{I \times m_x \times N}{Q} \quad (1)$$

209

210 where C_x = dosage of released metal (Fe or Al) from the outlet of electrolysis cell (mg/L), $M_{\text{total},x}$
211 = total mass of released metal (mg) by Faradays law within the retention time (t, s), $V_{\text{total,liquid}}$ =
212 total volume of the effluent (L) within retention time, I = applied current (amperes), N = number
213 of channels in the electrolysis cell, Q = flow rate (L/s) and m_x = electrochemical equivalent of the
214 metals. The electrochemical equivalent of aluminium metal is 0.093 mg/C and of iron metal is
215 0.193 mg/C.

216

217 The particle-size distribution (PSD) of a powder or granular material or particles dispersed in a
218 fluid is a list of values or a mathematical function that defines a relative amount, typically by mass
219 of particles present according to size. The samples from the effluent streams after the EC process
220 with Fe and Al electrodes are subjected to PSD characterisation. During the course of experiments,
221 every time when samples were subjected to PSD, 1 L sample was taken in order to examine the
222 resulting flocs in detail. The samples were taken separately for Fe and Al electrodes at their
223 respective amount of applied current densities. Then PSD is performed with the help of a laser
224 granulometer (Mastersizer 3000 from Malvern) [22]. With this device, particles can be measured
225 with a size range of 0.01–3500 microns. For the measurements, two light sources, one red at 632.8
226 nm and blue at 470 nm were used. At last, the operational cost analysis for optimum operating
227 current densities of Fe and Al electrode was also performed to verify the feasibility of designed

228 test EC reactor plant for micropollutants and suspended solids removal from the effluents by using
229 Eq. (2). While the energy consumption for this process was determined by using Eq. (3) [8].

$$230 \quad \text{Operating cost} = \text{energy cost} + \text{material cost} \quad (2)$$

$$231 \quad \text{Energy consumption} = \frac{V \times I}{Q} \quad (3)$$

232 Where V is the voltage applied on EC reactor, I is the current applied in Amperes on the EC reactor
233 and Q is the flow of the influent through the EC reactor in m³/h. The nergy consumption is
234 expressed in Wh/m³.

235

236 **3. Results and discussion**

237 **3.1 Treatment from primary sedimentation tank**

238 **3.1.1 COD removal efficiency of Fe and Al electrodes**

239 Applied current or current density is the most important operational parameter, in electrochemical
240 EC process. To test the role of current in COD removal, different current densities have been used
241 in the case of Fe and Al electrode at same initial pH of 7.3 and a flow rate of 500 L/h. In the case
242 of Fe electrode, two types of settings were used. In the first run, current was applied at 3 A (current
243 density= 2.02 A/m²) and in the second run current applied was 4 A (current density= 2.69 A/m²).
244 It was observed with the samples that flocs were formed within 5 min after the samples were placed
245 in Imhoff cones (Fig. 2). The COD was also measured afterwards. The settling of the flocs formed
246 at 3 A are shown in Fig. 2, as a function of time for settling. Furthermore, it can be clearly seen
247 from Fig. 2 that after 30 min of settling, the amount of solids in outflow was noticeably settled to
248 an amount of 32 mL/L. On the other part, the suspension in the inflow was still very stable that
249 was nowhere near settling during the 30 min of reaction. However, the greenish colour of outflow

250 after EC process indicated that the flocs formed contain purely $\text{Fe}(\text{OH})_2$, which shows the
251 dissolution of Fe electrodes in the form of Fe (II), also showed by other researchers [23-25].

252
253 The outflow samples were tested for the amount of COD removal. Fig. 3 shows that with 3 A
254 current, COD removal was from 233 to 100 mg/L and with 4 A current, COD removal was from
255 250 to 109 mg/L. A significant amount of COD removal from the wastewater of primary
256 sedimentation tank up to 56–57% with Fe electrocoagulation was achieved as shown in Fig. 6.
257 However, the increase in current did not make a significant difference in the COD removal, as it
258 is also confirmed by Roopashree and Lokesh [26], that more COD removal with Fe electrodes was
259 achieved at lower currents. Furthermore, there have been studies where high removal of organic
260 matter was obtained with iron electrodes, such as Katal and Pahlavanzadeh [27] and Perng et al.
261 [28]. However, the electric charge per volume has been really high [28] measured 70.5% removal
262 of COD (from 254 to 75 mg/L) from paper mill effluent by using 17,280 C /L.

263
264 While in the case of Al electrode, wastewater from primary sedimentation tank was introduced in
265 the EC reactor. The current applied was in the range of 1–2 A and corresponding current density
266 values were 0.67–1.34 A/m². The COD removal is shown in Fig. 4, with 1 A applied current COD
267 was decreased from 121 to 107 mg/L and with 2 A current COD was decreased from 105 to 86
268 mg/L. Furthermore, the sample treated with 1A applied current was put in Imhoff apparatus to see
269 the settling effects of solids and watched for a 30 min interval. It can be seen in Fig. 5 that there
270 was no significant colour imparted to the outflow and the settling with a half-hour interval was
271 quite good. That indicates the amount of flocs produced are almost doubled before and after EC
272 process. Moreover, the results confirmed the COD removal in the range of 12–18%, which is

273 almost 3 folds less than the COD removal achieved in the case of Fe electrodes as shown in Fig.
274 6. However, it is stated in the literature [24, 26] that Fe electrodes are more efficient than Al
275 electrodes in colour and COD removal.
276

277 **3.2 Treatment from secondary sedimentation tank**

278 **3.2.1 Effect of current density on AFS in Fe coagulation**

279 When Fe electrodes were used and wastewater from secondary sedimentation tank was introduced
280 in the EC reactor, the current was applied in the range of 1–3 A and current density was changed
281 from 0.67 to 2.02 A/m². Different operating conditions were opted for secondary sedimentation
282 tank effluent analyses because of their changed characteristics. Higher current densities were
283 applied for primary sedimentation tank analysis in comparison with lower values of current
284 densities for secondary sedimentation tank analysis. This is because of higher amount of
285 contaminants were detected in the primary sedimentation tank, while very little values were
286 observed in secondary tank effluents (Table 1). According to AFS amounts found in the inflow
287 and outflows after the EC reactor. It can be seen in Fig. 7 that the amount of AFS produced is
288 directly proportional to the amount of current applied. Furthermore, higher amounts of outflows
289 in comparison to inflows indicated that a large amount of electrode dissolution happened during
290 the course of the EC process. In addition, it showed that dissolution of Fe electrodes is the primary
291 reaction at anodes during typical current densities when pH approaches neutral values [23, 29].
292 Each experiment was run thrice to check the reproducibility of the results and named as WW-Fe-
293 01, WW-Fe-02 and so on. Similar was the case with Al coagulation.

294

295 In Fig. 8 coagulation dose of Fe, theoretical amount of Fe and AFS in outflows are compared
296 against different current densities. The measured amounts of Fe in outflow is based on reaction
297 (6), where one mole of Fe (s) reacts with two moles of water to produce one mole of Fe(OH)₂. It
298 was assumed that first, all iron electrodes produce Fe(OH)₂ precipitates which were dirty green in
299 colour and their amounts were then tested by Hach Lange test cuvettes, to prove the fact that the
300 whole amount of Fe that produced as coagulation dose by Fe electrodes is in the form of Fe(OH)₂.
301

302 According to the results, current density has a direct effect on the dissolution rate in the studied
303 range (0.67–2.02 A/m² or 7.19–21.58 C/L) (Fig. 9). Iron dissolved at the rate of 2.93 mg Fe/C,
304 while the theoretical value for dissolution of Fe (II) is 3.09 mg Fe/C. Therefore, it can be concluded
305 that iron dissolved in Fe (II) form and dissolution followed Faraday's law. This was consistent with
306 the results of other researchers [23-25]. These results are very important to establish the fact that
307 Fe (II) is a poor coagulant and should be oxidized to Fe (III) form before it is employed to remove
308 organic matter [1].
309

310 In order to test the volumetric inflow parameters of the EC reactor, a set of experiments was
311 performed with a low flow rate of about 250 L/h. Furthermore, the results are compared with the
312 inflow rate of 500 L/h. The amount of AFS obtained during different flow rates is shown in Fig.
313 10. The results clearly show that when inflow rate is decreased from 500 to 250 L/h, the amount
314 of AFS is increased to almost doubled, provided with the current applied remains constant.
315 Moreover, these results also support the arguments, that if charge per litre volume of inflow is
316 increased the amount of AFS and coagulation dose of Fe is also increased linearly. In other words,
317 if inflow rate to EC reactor has an indirect effect on the AFS produced provided the current density

318 is kept constant. However, this rapid agglomeration could not be helpful, as it needs more
319 coagulation dose and might not be cost-effective. Therefore, this parameter was not further
320 optimised.

321 **3.2.2 Effect of current density on AFS in Al coagulation**

322 Al electrodes were used and the wastewater from secondary sedimentation tank was introduced in
323 the EC reactor. The current was applied in the range of 0.5–1.0 A and current density was changed
324 from 0.34–0.67 A/m². The amount of AFS found in the inflow and outflow, before and after the
325 EC process are shown in Fig. 11 that indicates the current applied has a direct effect on the amount
326 of AFS produced. Furthermore, the electrodes dissolution is the primary reaction at anodes and
327 same is the case with aluminium electrodes.

328

329 In Fig. 12 coagulation dose of Al, theoretical amount of Al and AFS amounts in outflows are
330 presented corresponding to the range of current densities. The measured amount of Al based on
331 the reactions (1–3), where one mole of Al_(s) reacts with three moles of water to produce Al(OH)₃
332 and afterwards polymerised to other hydroxide species [6]. At the first instance, it is assumed that
333 aluminium electrodes produce on the whole Al(OH)₃, which is then experimentally proved by the
334 gravimetric test. The test was performed at 1 A current and 0.67 A/m² current density applied to
335 the EC reactor. The amount of Al based on the gravimetric measurement, AFS by the EC process
336 and calculated theoretically by following Eq. (1).

337

338 According to results, the current passing through the EC reactor was directly proportional to the
339 dissolution rate [10] and followed Faraday's law. In the studied range (0.34–0.67 A/m² or 2.45–
340 4.82 C/l), Fig. 13 shows dissolving rate of Al measured was 1.27–2.39 mgAl/C, whereas the

341 theoretical rate according to Faraday's law was 2.22 mgAl/C. Therefore, it can be concluded that
342 the amount of dissolution of Al at lower values of current densities was lower than the theoretical
343 values, however, approaches equal to theoretical values at higher current densities. When the
344 experiments run for Al electrodes at 250 L/h, the same results were obtained as shown in Fig. 10.

345 **3.2.3 Outflows pH**

346 In addition to the applied current, another factor that influences the reactions inside the EC reactor
347 is pH. Literature confirms that pH of the outflows changes in comparison to inflows as reaction
348 taking place at cathode (H_2 evolution) in EC reactor. Moreover, it also depends on inflow's pH
349 and type of selected anode [1, 5, 6]. In the previous section, it is noted that with an increase in
350 current density the amount of outflow AFS is increased. The pH factor was also observed before
351 and after EC process under the applied operational current densities and resultant AFS for both
352 electrodes. The pH of outflows is observed a correspondent increase with current density for all
353 experiments [6]. In all the experiments performed in both series, with Al and Fe electrodes, the pH
354 was slightly increased in the outflows. The inflow's pH for all run at variable applied current, in
355 case of Fe is 7.07, and while in case of Al is 7.24. In addition, the increase of pH was more in the
356 case of Fe electrodes (Fig. 14) than to Al electrodes (Fig. 15), in accordance with applied current
357 values.

358 **3.2.4 Flocc production analyses**

359 In EC process, hydrogen bubbles are produced on the cathodes (reaction 2 and 5) [30]. The
360 production of visible hydrogen bubbles on the electrodes indicates that EC process is going well.
361 Electrodes material and current density both have an effect on the bubble size and amount.
362 Furthermore, it is observed that very small hydrogen bubbles are produced in both cases when

363 aluminium or iron electrodes are used [5, 10]. According to these, smallest hydrogen bubbles are
364 produced during EC process at neutral or acidic pH. Current density has a direct effect on the
365 amount of bubbles, more was the current density, more bubbles were produced on the electrodes.
366 In additions, the gas bubbles produced can carry some of the flocs to the top surface of the
367 electrodes and then the layer can be removed in the form of foam.

368
369 The flocs layers produced due to the hydrogen bubbles can be seen in Fig. 16. It shows the layers
370 formed during the EC operation with Fe and Al electrodes simultaneously. Moreover, the sludge
371 formed during EC process was settlable and during an initial test with wastewater from the
372 secondary sedimentation tank, it was figured out that sludge sometimes settles down in the EC
373 chambers. Therefore, it was decided to empty the chambers and refill it with water at the end of
374 each day operation. Each time water is replaced from within EC cell and sludge was moved out of
375 the EC reactor. At the first instance, a little amount of sludge was deposited on the chambers of
376 EC cell.

377
378 The samples from the effluent streams after EC process with Fe and Al electrodes are subjected
379 for PSD analysis in a laser granulometer. Fig. 17 shows particle size distribution (PSD) for samples
380 in which Fe and Al electrodes are used for coagulation at 0.5–2 A applied current values. This
381 PSD analysis shows the cumulative volume of particles against the particle diameter in μm . From
382 Fig. 17, it is confirmed that a higher amount of current produced bigger amount of flocs with both
383 electrodes that later on could be separated by secondary separation technique. In the case of Fe
384 electrodes while increasing applied current more amount of flocs are observed however no such

385 increment has been observed in the case of Al electrode. The comparison of PSDs of Al with Fe
386 confirmed that at a same value of applied current, Al generates more flocs than Fe.

387 **3.3 Cost analysis of optimum current densities**

388 The operating cost for the current EC process is calculated with Eq. (2). The energy consumption
389 is then expressed in Wh/m³. Furthermore, the energy consumption costs for Fe and Al electrodes
390 at 2 A and 1 A operation are found to be 0.03 €/m³ and 0.02 €/m³ respectively. In addition, it is
391 observed that energy consumption is a direct relation with applied current, greater the applied
392 current the higher will be the consumption of energy [5]. Based on the measured coagulant doses
393 of Fe and Al electrodes as described in the previous section, the amounts of Fe and Al electrodes
394 consumed per day were found 0.011 Kg/m³ and 0.036 Kg/m³ at 2 A for Fe and 1 A for Al. Based
395 on these optimum conditions, the used metal plates in the EC reactor can last for 86 and 99 days
396 for Fe and Al electrodes with its full capacity of operation at 2A and 1A respectively. Moreover,
397 the material cost of Fe, Al operating plant at 2 A, 1 A was calculated to be 0.01 € /m³ and 0.02 €
398 /m³ respectively. According to reaction (5), the operating cost of EC operation for Fe and Al
399 electrodes at mentioned current values of 2 A and 1 A is calculated to be 0.04 € /m³ and 0.03 € /m³
400 respectively.

401

402 **4. Conclusions**

403 This study deals with wastewater treatment on a pilot-scale plant set up by electrochemical
404 technology. The electrocoagulation and electroflocculation techniques were applied with a revived
405 design to meet the needs of the process to remove suspended solids and micropollutants out of the
406 wastewater stream. By using various settings of applied currents, the process was tested for the

407 better quality of effluent using water from the outflow of primary sedimentation tank and treated
408 water from the outflow of secondary sedimentation tank. Furthermore, the results are optimised
409 for a better downstream separation process. Wastewater from primary sedimentation tank was
410 treated with both Fe and Al electrodes. In the case of Fe electrodes, more than 50% of COD
411 removal was observed which was about 12–18% with Al electrodes. The coagulation dose of Fe
412 and Al electrodes were measured, on the basis of AFS produced and according to Faraday's Law.
413 With both Fe and Al electrodes, the coagulation dose went on increasing with increasing current
414 density. The best setting for Fe and Al electrodes in terms of current and current densities for
415 effluent from secondary sedimentation tank were 3 A (2.02 A/m^2) and 1 A (0.67 A/m^2) respectively.
416 In addition, the higher the coagulation dose, the bigger were the flocs in size and hence can easily
417 settle down in downstream sedimentation tank. The operating cost of operation for Fe and Al
418 electrodes at the most feasible mentioned current values of 2 A and 1 A were calculated as 0.04 €
419 / m^3 and 0.03 € / m^3 respectively. The work provided promising results in comparison with the past
420 work done by other researchers. Furthermore, these findings provided a new space and
421 enhancements, in the existing literature for continuing research in this regard. In future, a
422 combination of electrode materials and natural coagulants could be a good option for efficiency
423 improvement of this type of pilot scale.

424

425 **References**

- 426 1. Eryuruk, K., U.T. Un, and U.B. Ogutveren, Electrochemical treatment of wastewaters from
427 poultry slaughtering and processing by using iron electrodes. *Journal of cleaner production*,
428 2018. 172: p. 1089-1095.
- 429 2. Sher, F., A. Malik, and H. Liu, Industrial polymer effluent treatment by chemical
430 coagulation and flocculation. *Journal of Environmental Chemical Engineering*, 2013. 1(4):
431 p. 684-689.
- 432 3. Natarajan, R., F. Al Fazari, and A. Al Saadi, Municipal waste water treatment by natural
433 coagulant assisted electrochemical technique—Parametric effects. *Environmental*
434 *technology & innovation*, 2018. 10: p. 71-77.
- 435 4. GilPavas, E., I. Dobrosz-Gómez, and M.Á. Gómez-García, Optimization of sequential
436 chemical coagulation-electro-oxidation process for the treatment of an industrial textile
437 wastewater. *Journal of water process engineering*, 2018. 22: p. 73-79.
- 438 5. Yavuz, Y. and Ü. Ögütveren, Treatment of industrial estate wastewater by the application
439 of electrocoagulation process using iron electrodes. *Journal of environmental management*,
440 2018. 207: p. 151-158.
- 441 6. Izadi, A., et al., Treatment of paper-recycling wastewater by electrocoagulation using
442 aluminum and iron electrodes. *Journal of Environmental Health Science and Engineering*,
443 2018. 16(2): p. 257-264.
- 444 7. Hashim, K.S., et al., Electrocoagulation as a green technology for phosphate removal from
445 River water. *Separation and Purification Technology*, 2019. 210: p. 135-144.
- 446 8. Papadopoulos, K.P., et al., Treatment of printing ink wastewater using electrocoagulation.
447 *Journal of environmental management*, 2019. 237: p. 442-448.
- 448 9. Ensano, B.M.B., et al., Applicability of the electrocoagulation process in treating real
449 municipal wastewater containing pharmaceutical active compounds. *Journal of hazardous*
450 *materials*, 2019. 361: p. 367-373.
- 451 10. Elazzouzi, M., et al., A novel electrocoagulation process using insulated edges of Al
452 electrodes for enhancement of urban wastewater treatment: Techno-economic study.
453 *Process Safety and Environmental Protection*, 2018. 116: p. 506-515.
- 454 11. Tanyol, M., A. Ogedey, and E. Oguz, COD removal from leachate by electrocoagulation
455 process: treatment with monopolar electrodes in parallel connection. *Water Science and*
456 *Technology*, 2018. 77(1): p. 177-186.
- 457 12. Omwene, P.I., M. Koby, and O.T. Can, Phosphorus removal from domestic wastewater
458 in electrocoagulation reactor using aluminium and iron plate hybrid anodes. *Ecological*
459 *engineering*, 2018. 123: p. 65-73.
- 460 13. Yadav, A.K., et al., Removal of various pollutants from wastewater by electrocoagulation
461 using iron and aluminium electrode. *Desalination and water treatment*, 2012. 46(1-3): p.
462 352-358.
- 463 14. Devlin, T.R., et al., Electrocoagulation of wastewater using aluminum, iron, and
464 magnesium electrodes. *Journal of hazardous materials*, 2019. 368: p. 862-868.
- 465 15. Davarnejad, R., M. Mohammadi, and A.F. Ismail, Petrochemical wastewater treatment by
466 electro-Fenton process using aluminum and iron electrodes: Statistical comparison. *Journal*
467 *of Water Process Engineering*, 2014. 3: p. 18-25.

- 468 16. Can, O.T., E. Gengec, and M. Kobya, TOC and COD removal from instant coffee and
469 coffee products production wastewater by chemical coagulation assisted electrooxidation.
470 *Journal of Water Process Engineering*, 2019. 28: p. 28-35.
- 471 17. Mahmad, M.K.N., et al., Electrocoagulation process by using aluminium and stainless steel
472 electrodes to treat total chromium, colour and turbidity. *Procedia Chemistry*, 2016. 19: p.
473 681-686.
- 474 18. Chou, W.-L., C.-T. Wang, and S.-Y. Chang, Study of COD and turbidity removal from real
475 oxide-CMP wastewater by iron electrocoagulation and the evaluation of specific energy
476 consumption. *Journal of hazardous materials*, 2009. 168(2-3): p. 1200-1207.
- 477 19. Dudziak, M., et al., Application of Ultrafiltration in a Swimming Pool Water Treatment
478 System. *Membranes*, 2019. 9(3): p. 44.
- 479 20. Baierle, F., et al., Biomass from microalgae separation by electroflotation with iron and
480 aluminum spiral electrodes. *Chemical Engineering Journal*, 2015. 267: p. 274-281.
- 481 21. Heidmann, I. and W. Calmano, Removal of Ni, Cu and Cr from a galvanic wastewater in
482 an electrocoagulation system with Fe-and Al-electrodes. *Separation and Purification
483 Technology*, 2010. 71(3): p. 308-314.
- 484 22. Lee, S.Y. and G.A. Gagnon, Comparing the growth and structure of flocs from
485 electrocoagulation and chemical coagulation. *Journal of Water Process Engineering*, 2016.
486 10: p. 20-29.
- 487 23. Sasson, M.B., W. Calmano, and A. Adin, Iron-oxidation processes in an electroflocculation
488 (electrocoagulation) cell. *Journal of Hazardous Materials*, 2009. 171(1-3): p. 704-709.
- 489 24. Linares-Hernández, I., et al., Influence of the anodic material on electrocoagulation
490 performance. *Chemical engineering journal*, 2009. 148(1): p. 97-105.
- 491 25. Bagga, A., S. Chellam, and D.A. Clifford, Evaluation of iron chemical coagulation and
492 electrocoagulation pretreatment for surface water microfiltration. *Journal of Membrane
493 Science*, 2008. 309(1-2): p. 82-93.
- 494 26. Roopashree, G. and K. Lokesh, Comparative study of electrode material (iron, aluminium
495 and stainless steel) for treatment of textile industry wastewater. *International journal of
496 environmental sciences*, 2014. 4(4): p. 519.
- 497 27. Katal, R. and H. Pahlavanzadeh, Influence of different combinations of aluminum and iron
498 electrode on electrocoagulation efficiency: Application to the treatment of paper mill
499 wastewater. *Desalination*, 2011. 265(1-3): p. 199-205.
- 500 28. Perng, Y.-S., et al., Application of a pilot-scale pulsed electrocoagulation system to OCC-
501 based paper mill effluent. *TAPPI JOURNAL* 8 (3):, 2009. 14(20).
- 502 29. Mouedhen, G., et al., Behavior of aluminum electrodes in electrocoagulation process.
503 *Journal of hazardous materials*, 2008. 150(1): p. 124-135.
- 504 30. El-Ashtoukhy, E.-S., T. Zewail, and N. Amin, Removal of heavy metal ions from aqueous
505 solution by electrocoagulation using a horizontal expanded Al anode. *Desalination and
506 Water Treatment*, 2010. 20(1-3): p. 72-79.
- 507
508

509
510
511
512
513
514

List of Tables

Table 1. Wastewater characteristics.

Variable	Wastewater from primary sedimentation tank	Wastewater from secondary sedimentation tank
pH	7.30	7.30
Conductivity ($\mu S/cm$)	1350	850–1100
Suspended solids (mg/L)	100–150	1.4–6.6
COD (mg/L)	233.2	20
Total Phosphorous (mg/L)	5	0.45
Total Nitrogen (mg/L)	40.9	11.5

515
516

List of Figures

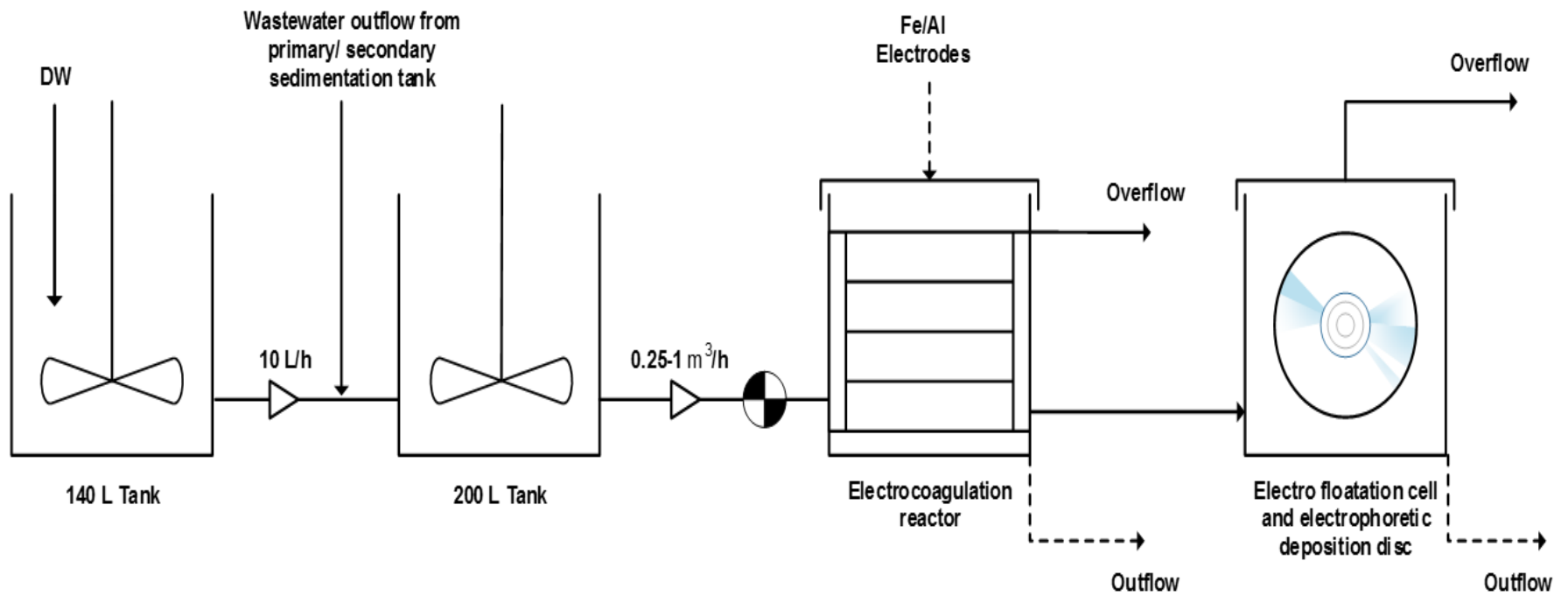


Fig. 1. Electrocoagulation pilot plant process flow diagram.

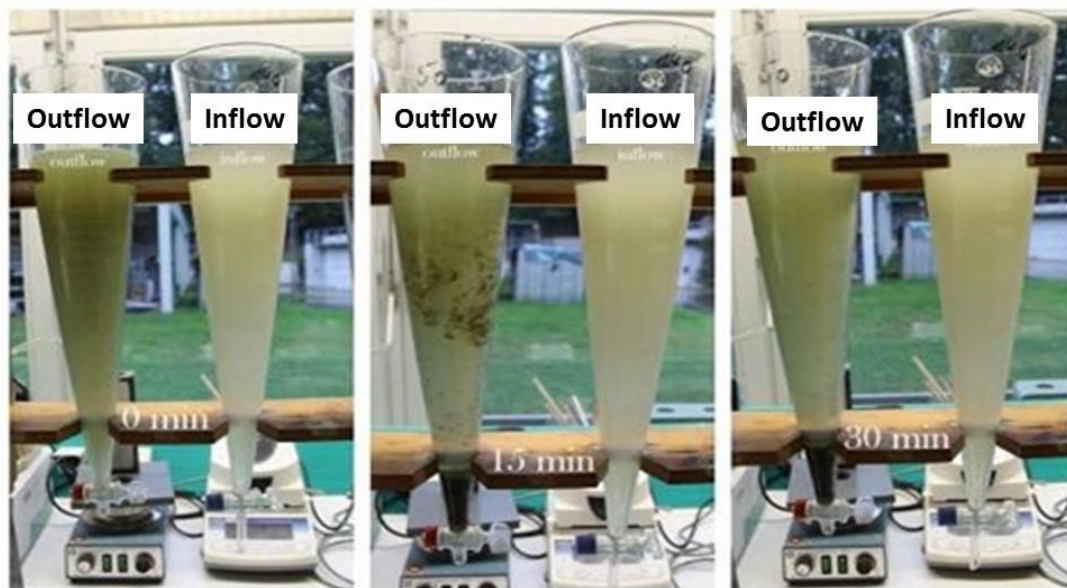


Fig. 2. Change of colour with Fe electrodes before and after EC treatment with time.

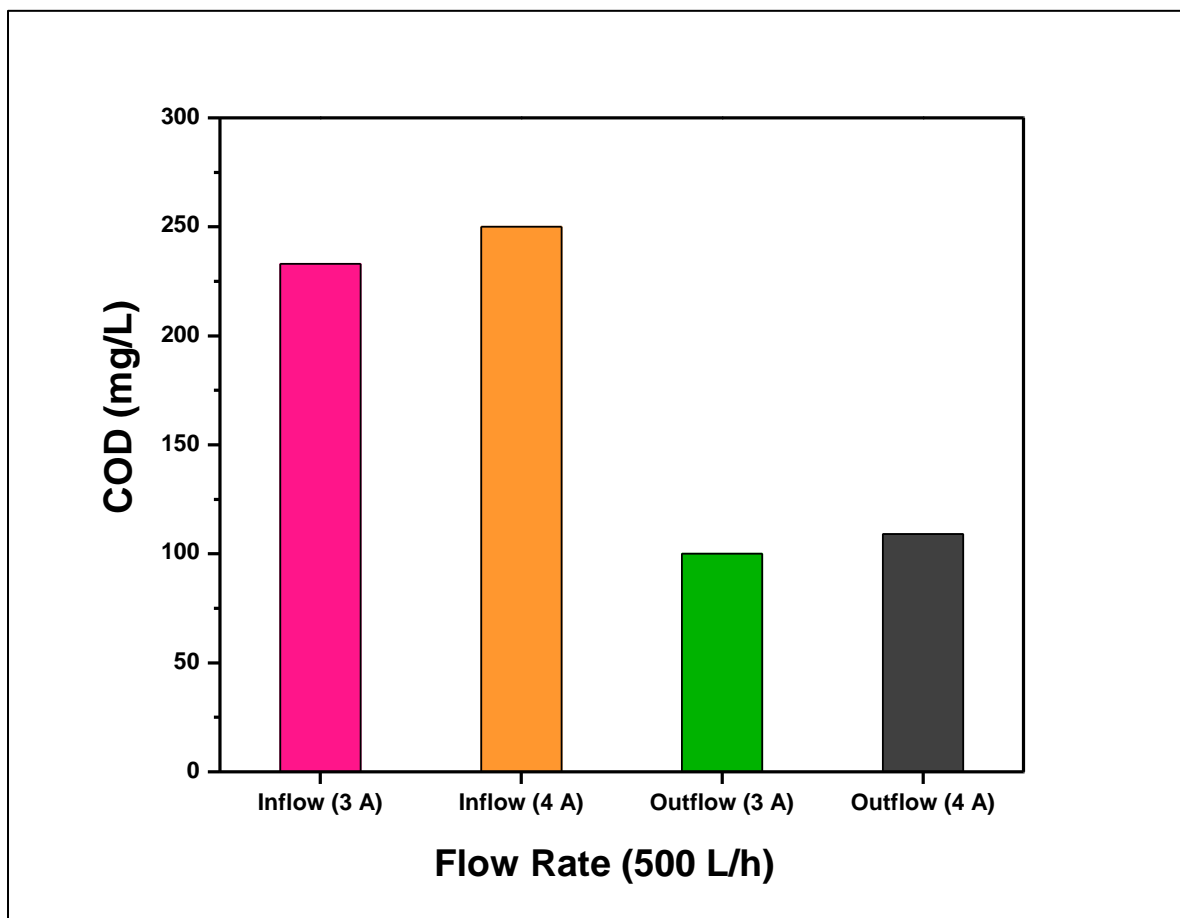


Fig. 3. COD removal of wastewater from primary sedimentation tank using Fe electrodes.

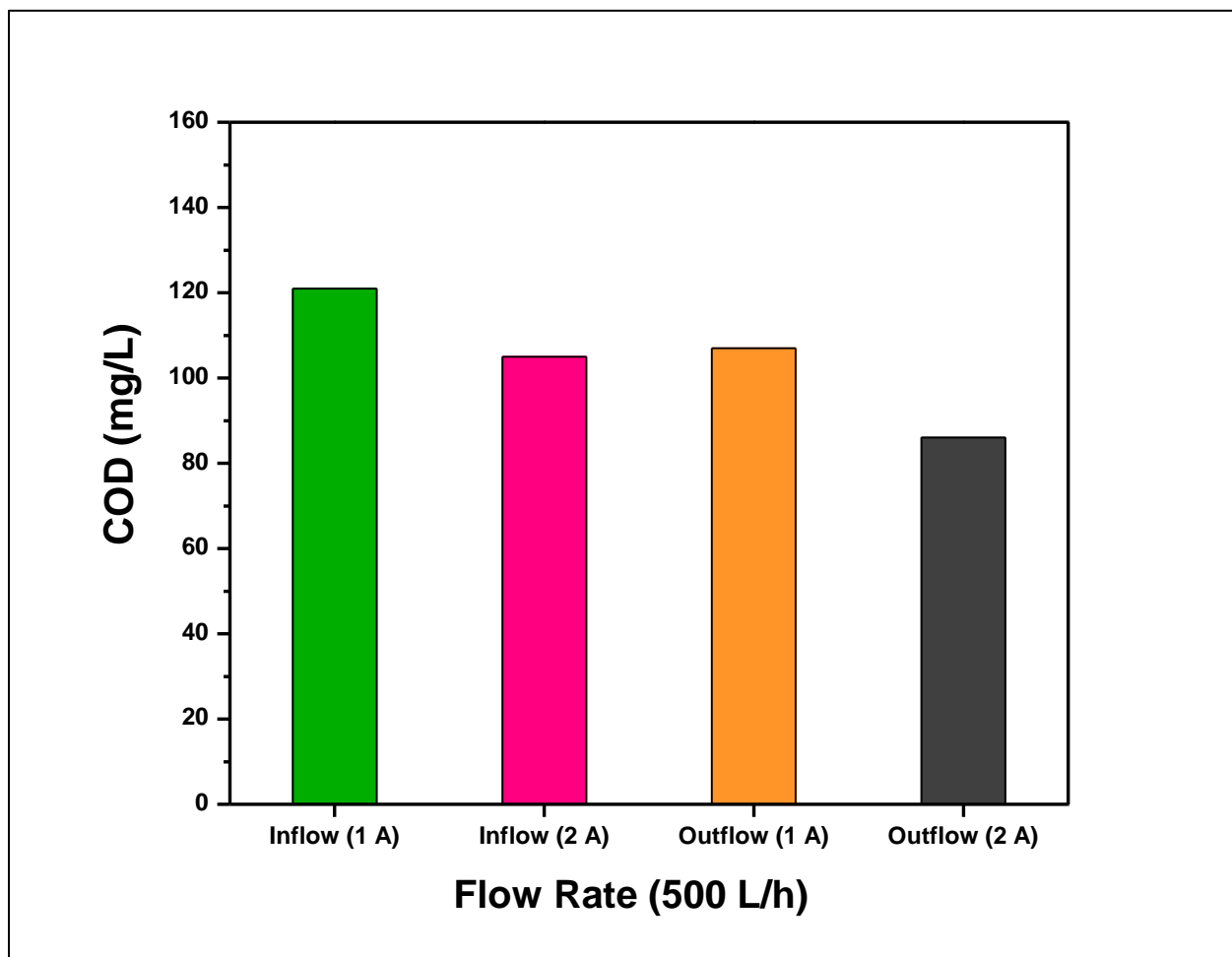


Fig. 4. COD removal of wastewater from the primary sedimentation tank using Al electrodes.

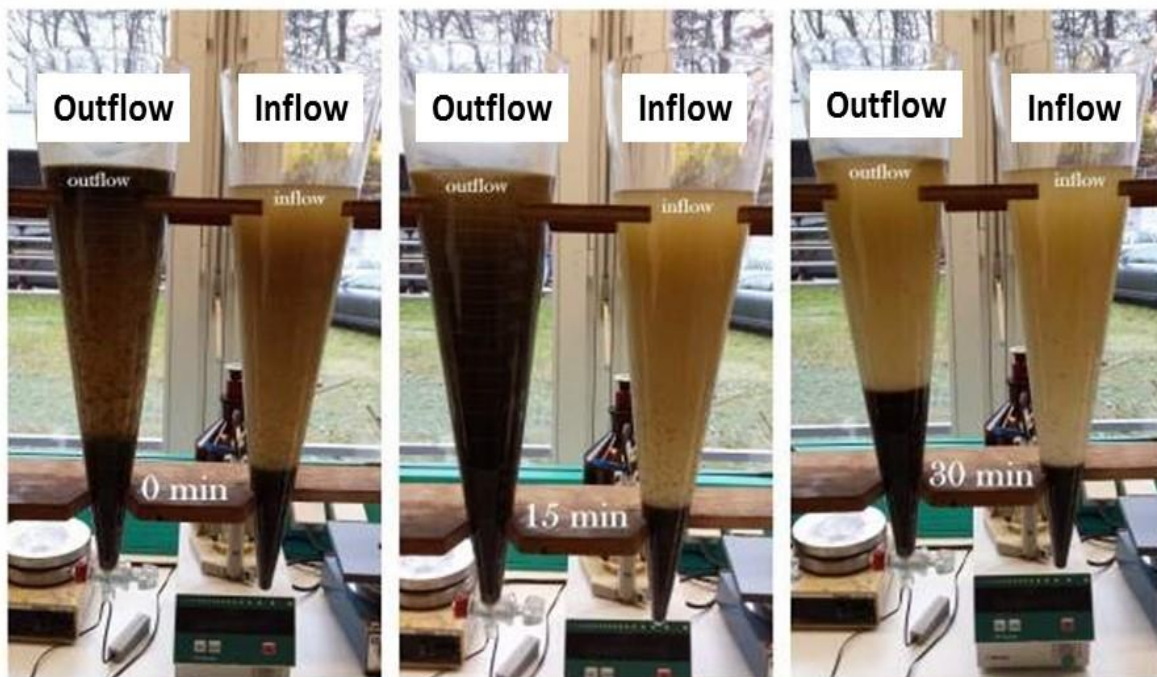


Fig. 5. Change of colour with Al electrodes before and after EC treatment with time.

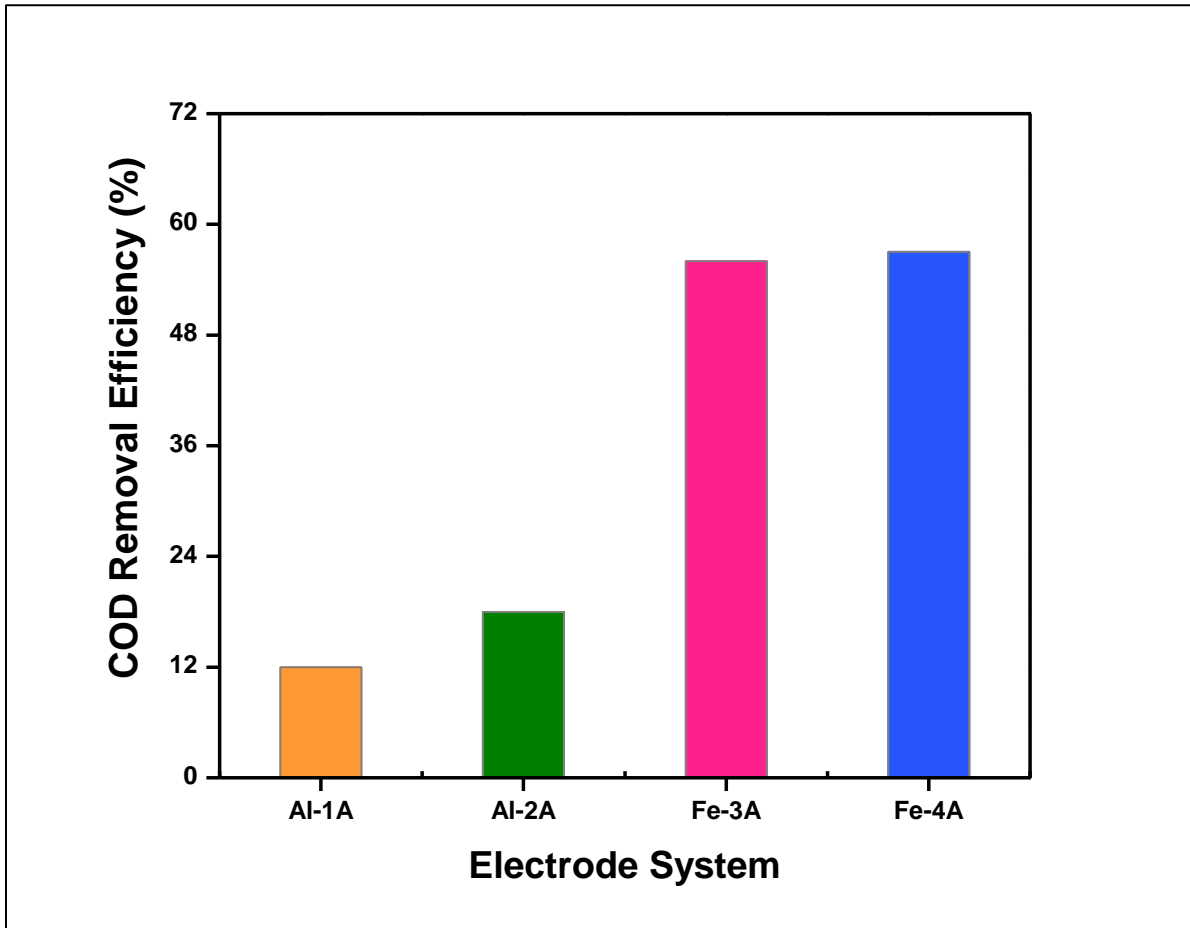


Fig. 6. The COD removal efficiency of Fe and Al electrodes at selected current densities.

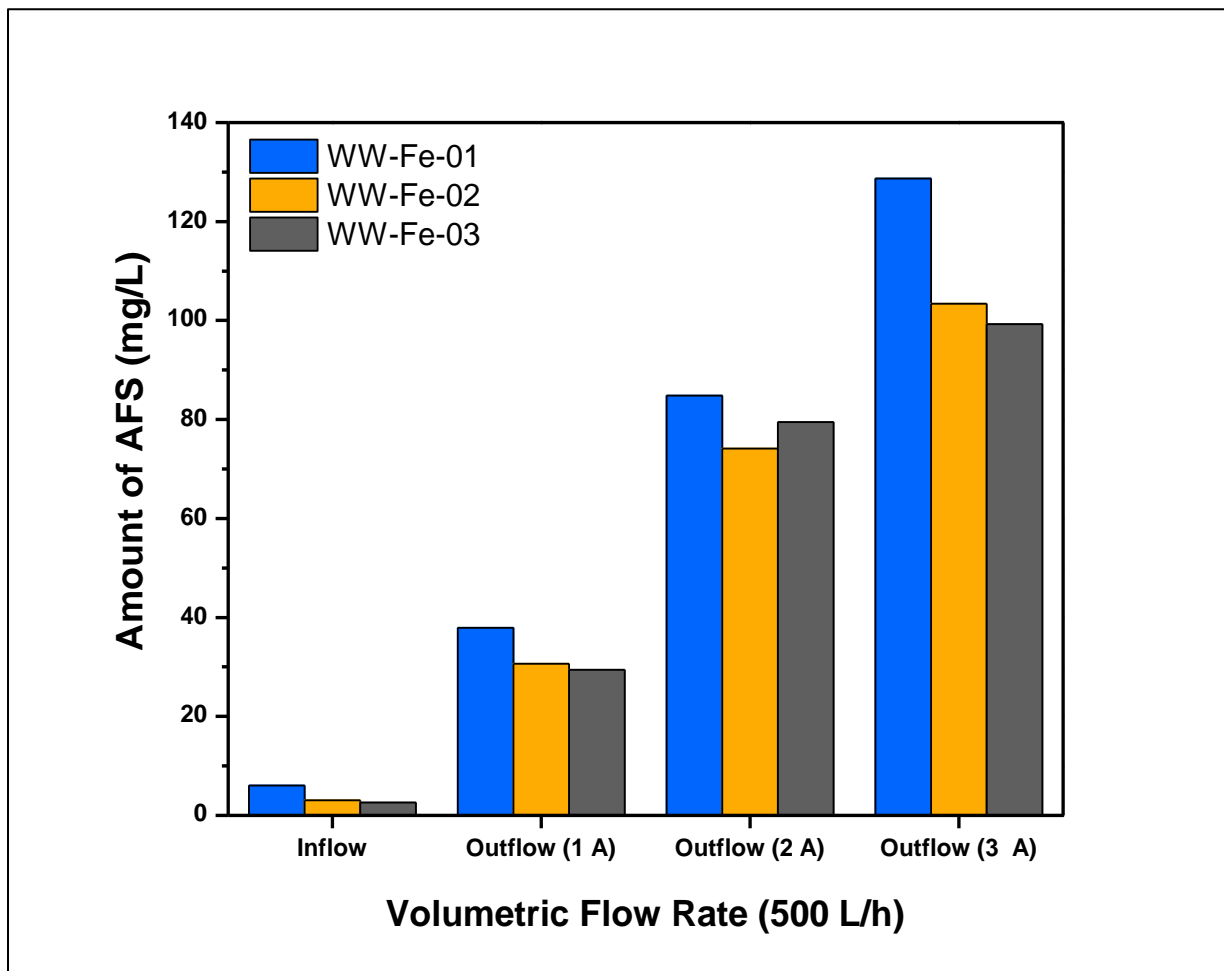


Fig. 7. Amount of AFS after applying current from 1 to 3 A on the EC reactor.

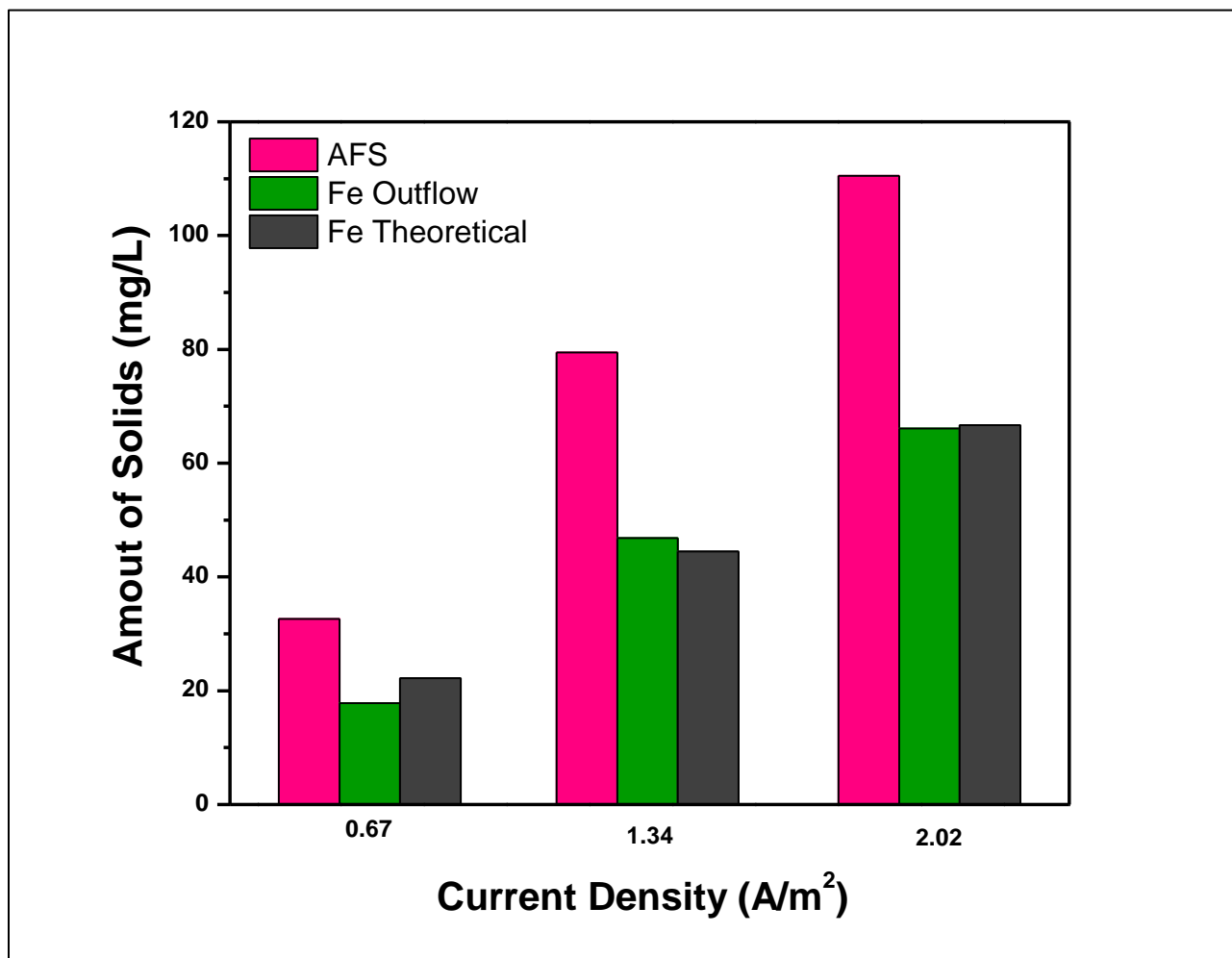


Fig. 8. Amount of AFS, coagulation dose of Fe and theoretical amount of Fe in the outflow against various amount of current densities.

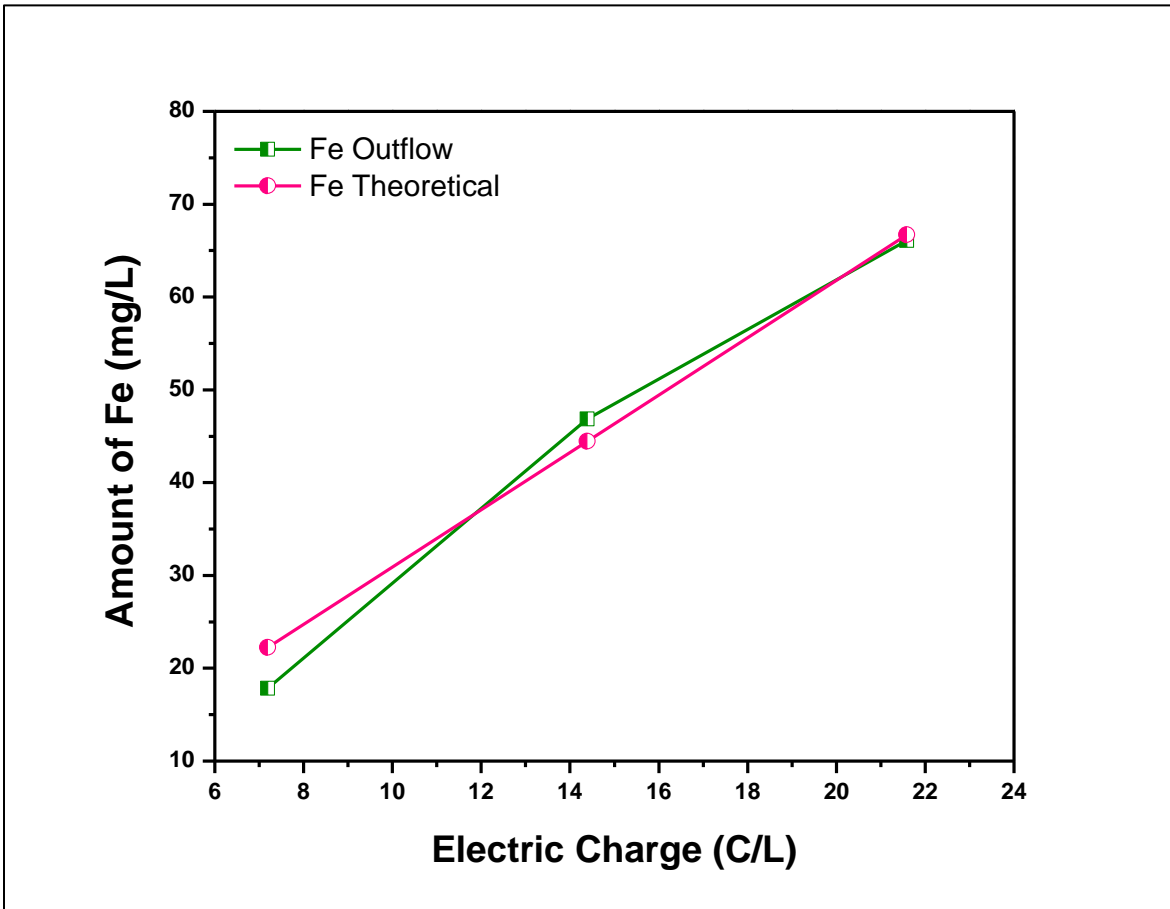


Fig. 9. Amount of Fe in the outflow and theoretical amount of Fe against charge per litre of influent stream.

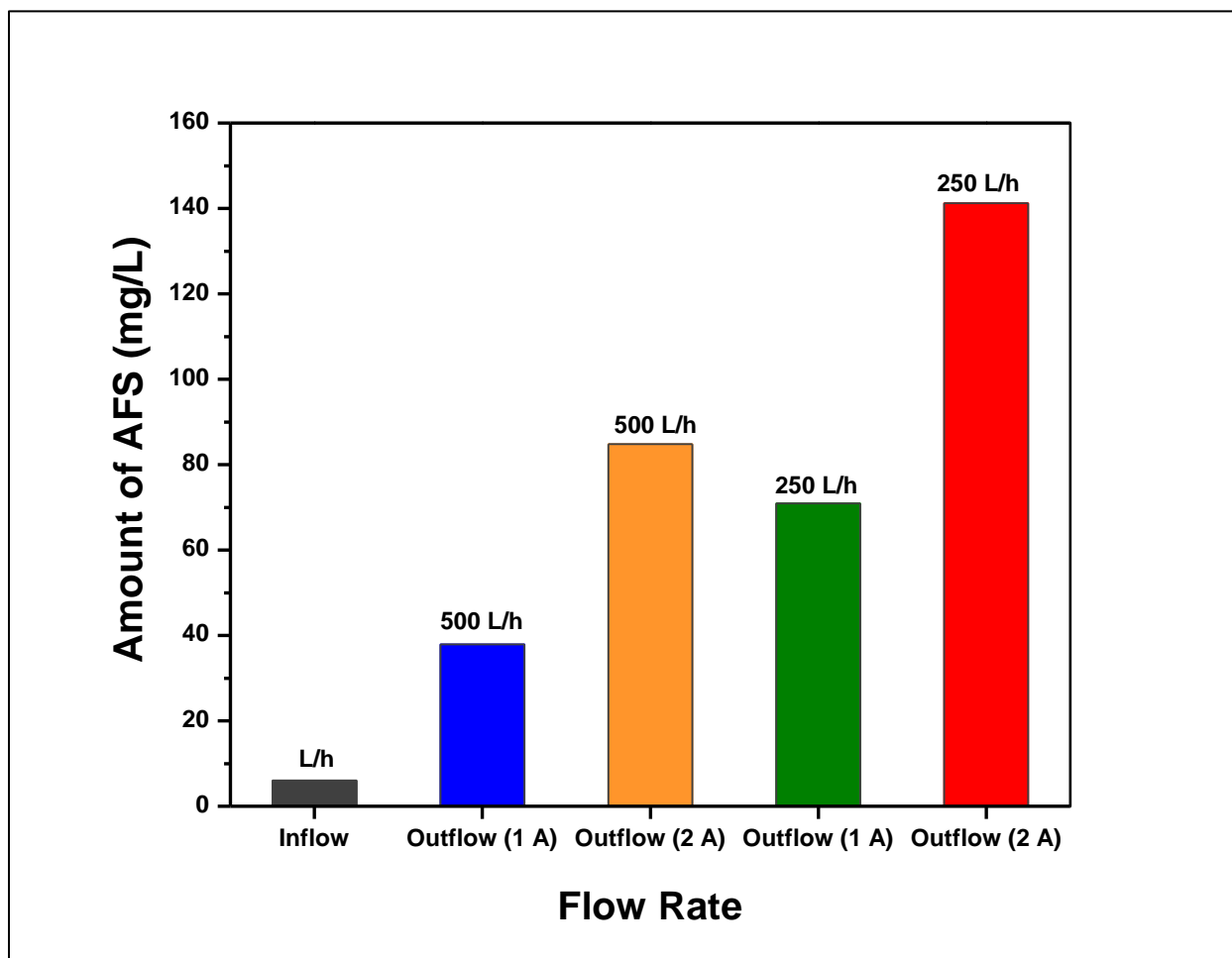


Fig. 10. Amount of AFS determined in the outflow after varying the current from 1 to 2 A and volumetric inflow rate from 250 to 500 L/h through the EC reactor.

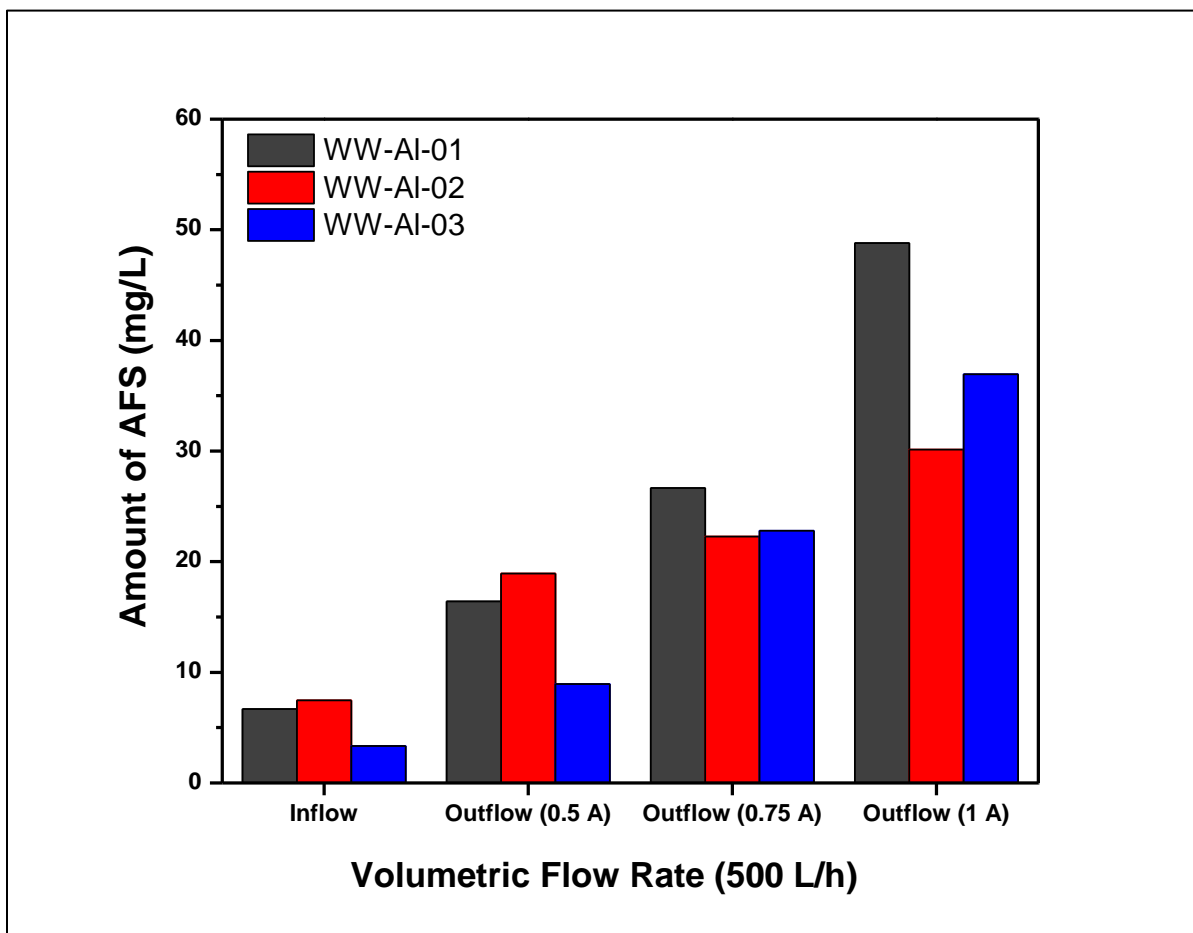


Fig. 11. Amount of AFS determined in the outflow after applying current at 0.5 A, 0.75 A and 1 A through the EC reactor.

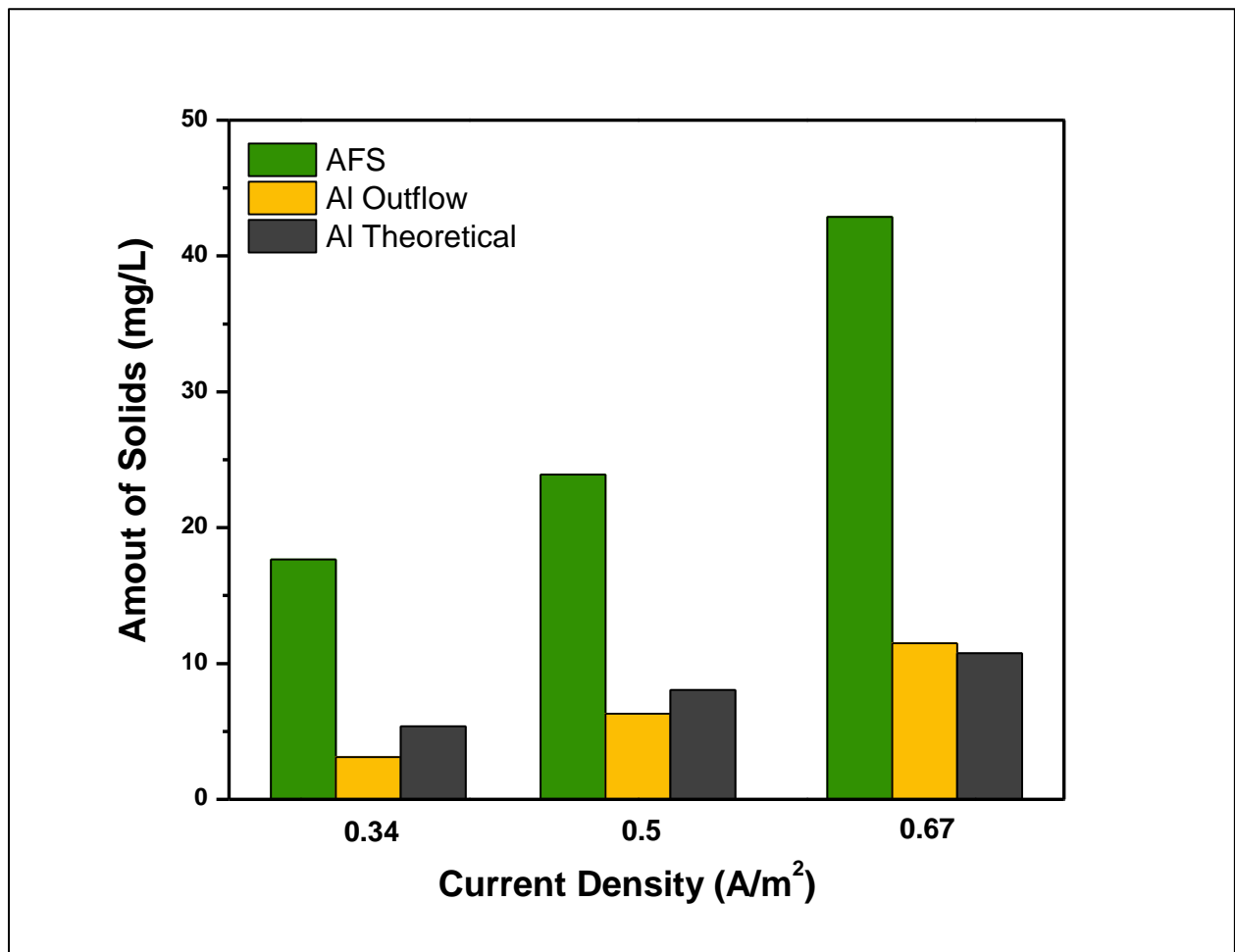


Fig. 12. Amount of AFS, coagulation dose of Al and theoretical amount of Al in the outflow against various amount of current densities.

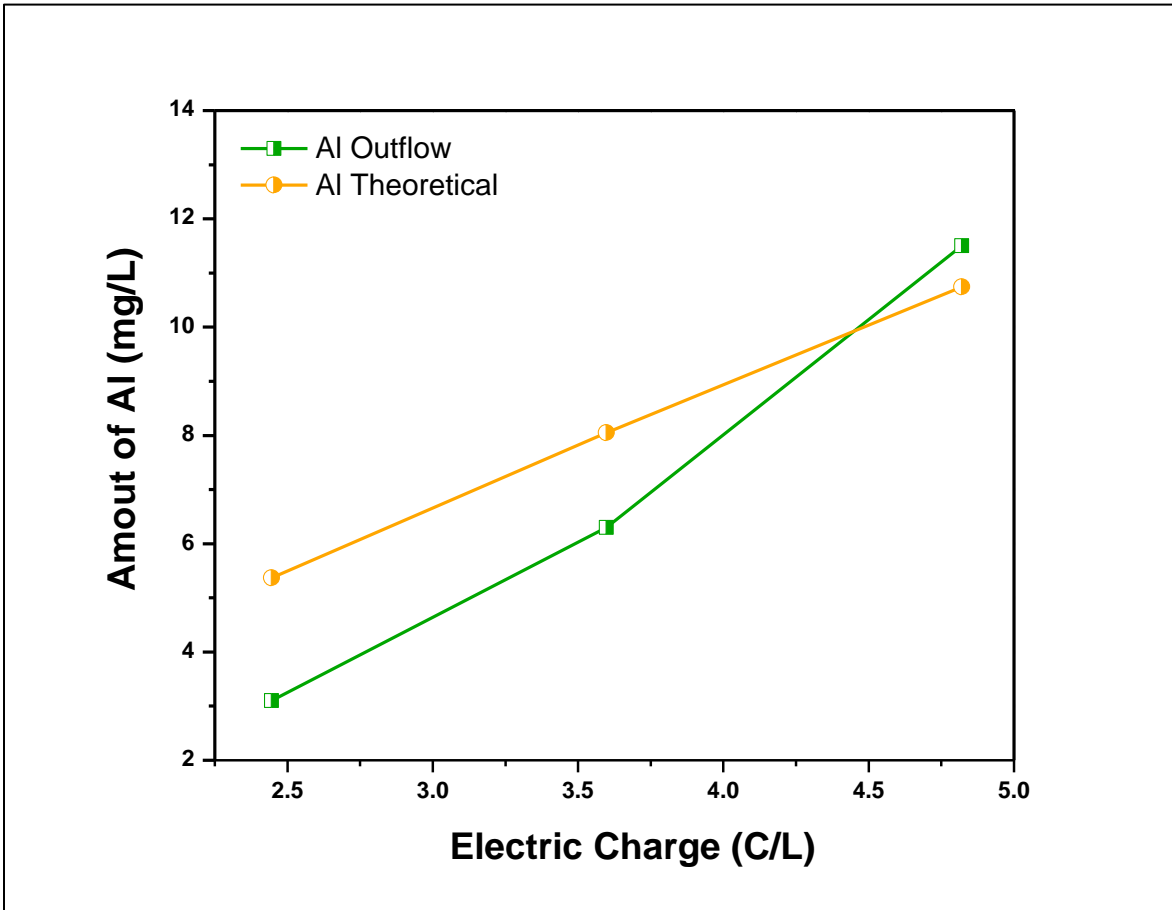


Fig. 13. Amount of Al in the outflow and theoretical amount of Al against the charge per litre of influent stream.

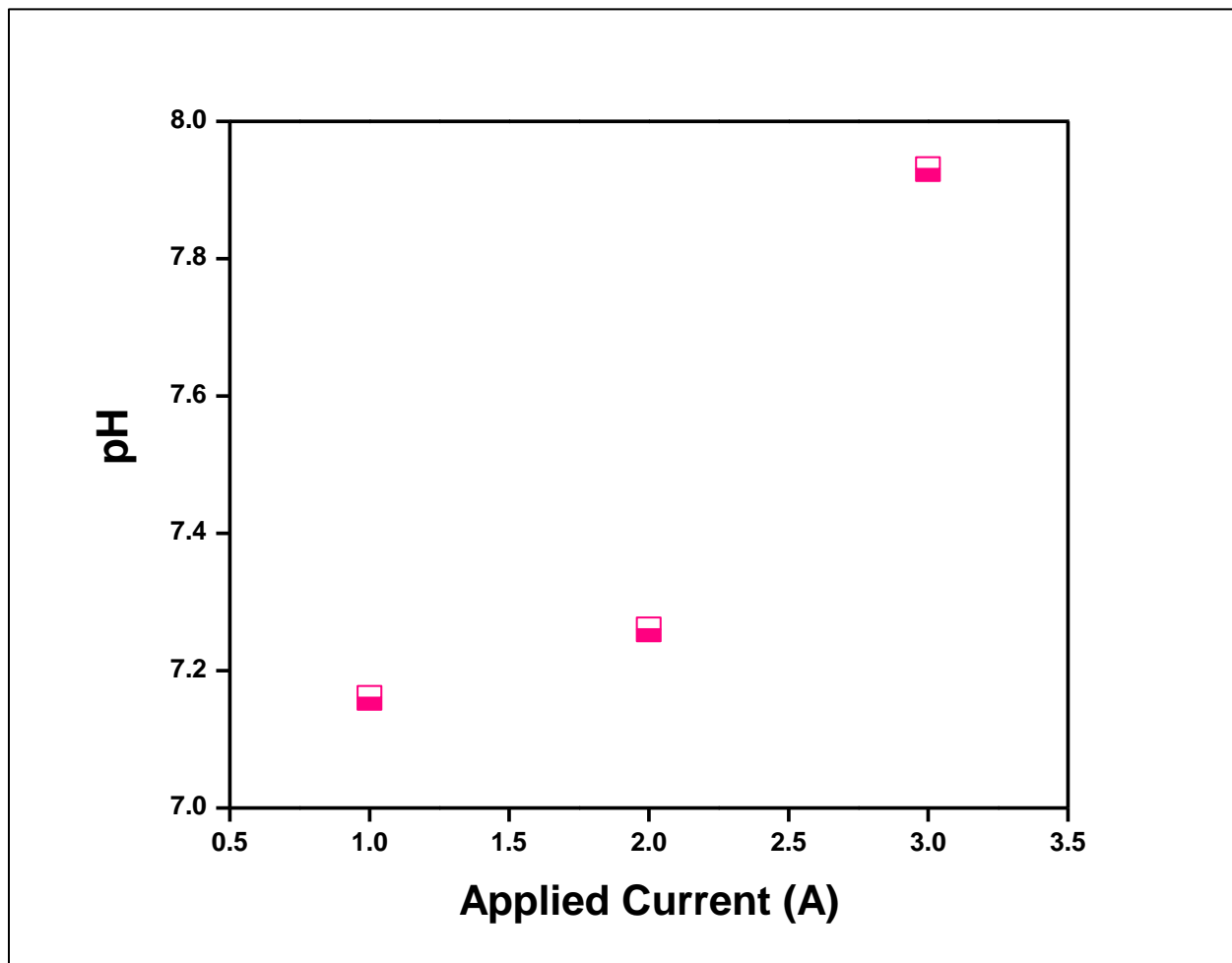


Fig. 14. Effect of applied current on outflow pH in the case of Fe electrodes EC treatment.

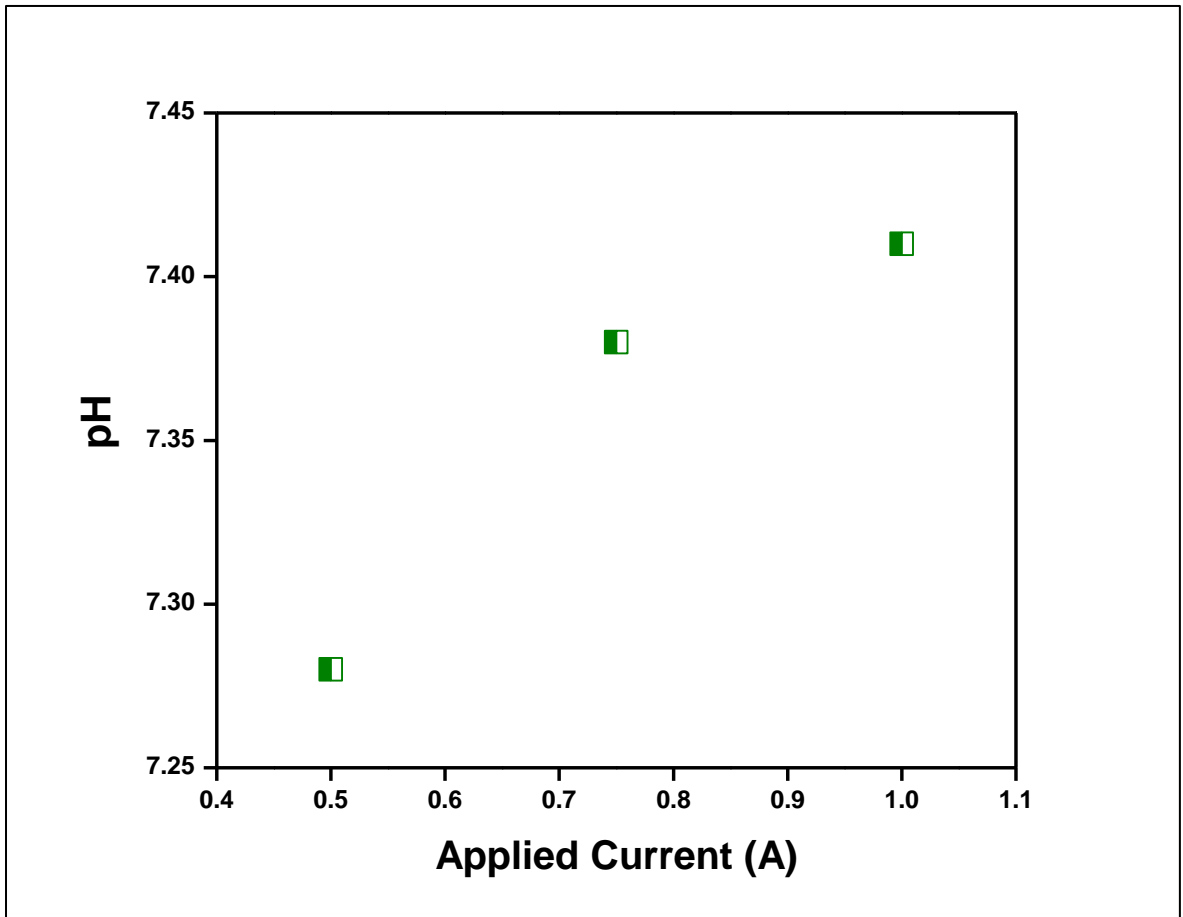


Fig. 15. Effect of applied current on outflow pH in the case of Al electrodes EC treatment.

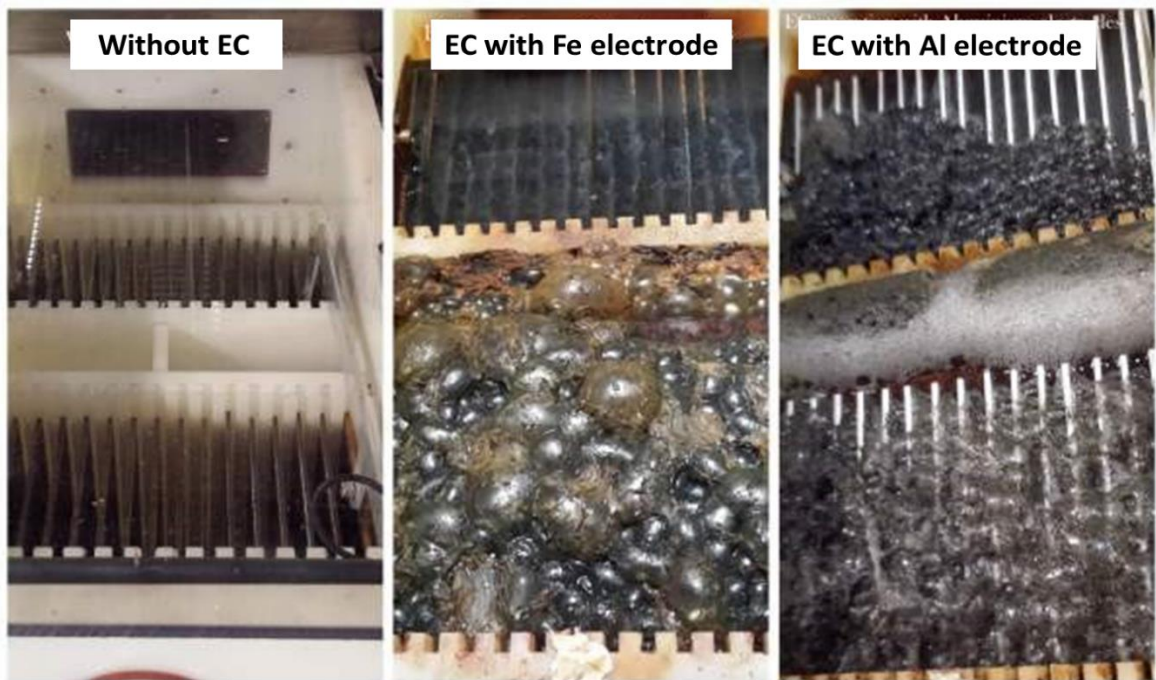


Fig. 16. Flocs layers formed due to hydrogen bubbles with or without EC treatment.

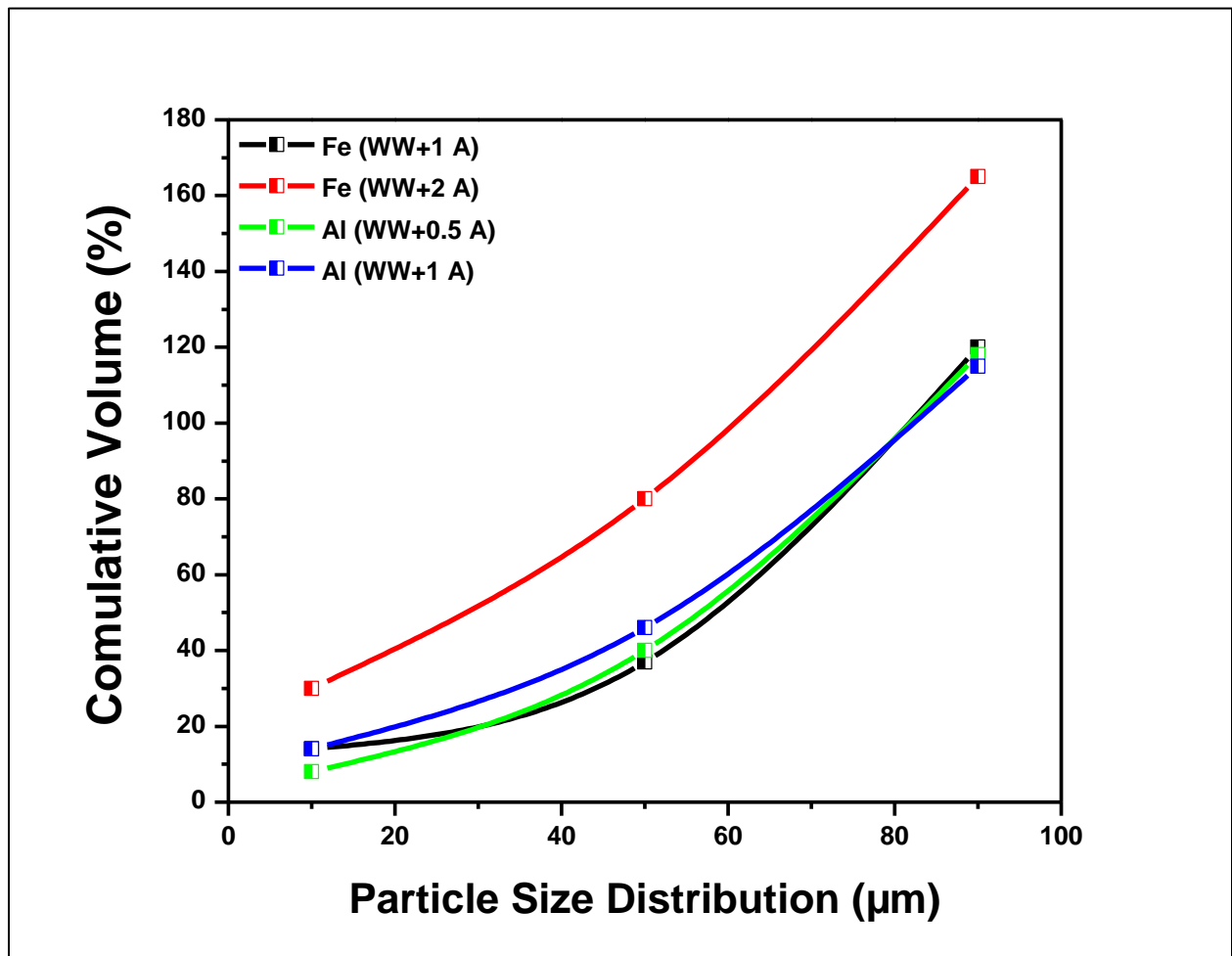


Fig. 17. The particle size distribution of Fe and Al flocs generated by EC reactor.