

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

4
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11
12 **Abstract**

13 In this research, wastewater treatment was inspected on a pilot-scale wastewater treatment plant
14 by electrochemical techniques, electrocoagulation (EC), electroflotation (EF) and electrophoretic
15 deposition (EPD). The wastewater samples have been characterised by applying different
16 parameters to determine the optimum working conditions of the electrocoagulation reactor. Two
17 electrodes have been tested separately with an outflow coming from the primary and secondary
18 sedimentation tank. The outflows from these tanks are introduced in the EC reactor, and then EC
19 reactor efficacy is determined for the removal of; chemical oxygen demand (COD), suspended
20 solids, micropollutants and amount of coagulants in agglomerates, at different current densities.
21 The amounts of suspended solids (SS) in the influent and effluent streams were determined by the
22 membrane filtration technique. The operational applied current values range from (1–4 A) in case
23 of COD removal by Fe and Al, while for SS aggregation the applied current ranges from (0.5–3
24 A) and inflow rate is tested from 250–500 L/h. The pH of the outflows increase by an increase in

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25 the applied current and both these parameters were found a positive increase in the amount of SS
26 aggregations after EC treatment. The COD removal efficiency was found to be 56–57% and 12–
27 18% in case Fe and Al electrode respectively after EC treatment. The results showed that the
28 applied current is the most effective parameter, whereas the aluminium electrodes have produced
29 more amounts of flocs and bubbles in comparison to iron electrodes at a similar amount of current
30 density.

31

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

34 **1 Introduction**

35 The removal of anthropogenic micropollutants emitting from industrial, agricultural, domestic and
36 urban sources is one of today's major challenges. The number of such micropollutants is
37 significantly large and many of these are found in excessive quantities. Pathogenic, non-
38 pathogenic organisms, pharmaceutical and drug residues viruses and vaccines present in large
39 amount in wastewaters. These micropollutants can have direct and indirect effects on the living
40 organisms by bio magnification along the food chain. All the mentioned sources of wastewater
41 produce a huge amount of pollutants; total organic carbon, biochemical and chemical oxygen
42 demand (BOD, COD), total suspended solids, total nitrogen and total phosphorus [1, 2]. The main
43 treatments till now available for the removal of these anthropogenic pollutants and micropollutants
44 involve aerobic biodegradation, filtration, flocculation, adsorption, froth floatation and EC [3].
45 Some other methods have been applied in combination to overcome the shortcomings of the
46 individual process, including photo-electro-Fenton, electro-Fenton and electro-oxidation [4]. EC
47 is a highly studied process in this field these days. It is a multistep process which involves the

48 agglomeration of contaminants, via electrodes (coagulant) through redox reaction carried out by
49 applying an electric current. Conventional electrocoagulation process was carried out by the help
50 of inorganic chemical species as coagulants including $(\text{NH}_4)_2\text{SO}_4$ and FeCl_3 .

51

52 Various other electrochemical and photo-assisted electrochemical processes have been examined
53 for the purification of wastewater [5], whereas preference is given to the EC method. The EC
54 process as proceeds by electric current using metallic electrodes, therefore, it is an electrochemical
55 process being used for the removal of contaminants from wastewater [6, 7]. It involves the
56 conversion of hazardous organic pollutants of wastewater via redox reactions to non-hazardous
57 materials. The EC process is famous as a green technology because of its simple assembly, safety,
58 short reaction time, selective capacity, negligible usage of chemicals, easy to handle/ or operate
59 and good water purification efficiency [7, 8]. The electric current (I) supplies the necessary force
60 (electromotive) to drive the redox reactions, resultantly the particulate/ or contaminant will reach
61 the stable state; solid, that is relatively less emulsifiable, less soluble and less colloidal in
62 comparison to the equilibrium values. When this happens, the stable solids convert into
63 hydrophobic compounds/ or precipitates which can be easily separated out by different separation
64 techniques. The EC method uses a negligible amount of chemicals, therefore no need for
65 neutralization reaction as there is no secondary contamination [9].

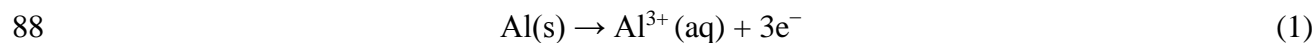
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67 Three stages of EC technology implicate in the purification of wastewater are; (1) electrolytic
68 oxidation of sample water to generate coagulants; (2) pollutant destabilization, emulsion,
69 deterioration and particle suspension; (3) agglomeration of resultant particles to generate flocs:
70 comprised of colloids entrapped sludge blanket formed from coagulation reaction [10]. These

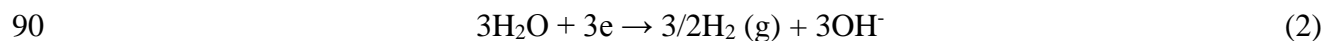
71 flocs are similar to chemical flocs and are larger, acid-resistant, bounded less water and stable,
 72 that's why can be separated out by rapid sand filtration. [11, 12]. EC process is economical as it
 73 produces relatively less amount of sludge in comparison with conventional processes [13]. There
 74 are a number of electrochemical reactions take place at anode and cathode as discussed in
 75 equations (1–7). During EC, H₂ is evolved at the cathode in the form of bubbles, discards particles
 76 by flotation known as electroflotation [14]. Numerous hydroxide of metallic electrodes (anode:
 77 Fe/ or Al) and coagulant compounds are generated from these chemical reactions to deteriorate,
 78 coagulate, and adsorb the pollutants [11]. Aluminium and iron electrodes are widely used in
 79 literature for wastewater treatment because of their increased ions production potentials [15, 16].
 80 The cations of iron (Fe⁺²) and aluminium (Al⁺³) dissolve from the relevant anode in the aqueous
 81 phase of the reactor as shown in reaction 1 and 4. Al⁺³ and Fe⁺² cationic species may form a number
 82 of monomeric and polymeric hydroxide entities during the course of electrochemical reactions
 83 relevant to each electrode. The transformation of metallo cationic species, into Al(OH)₃ (reaction
 84 3) and Fe(OH)₂ (reaction 5 and 7) hydroxides, is pH-dependent. These final species then act as
 85 micropollutant adsorbents.

86 Aluminum electrode reactions during EC:

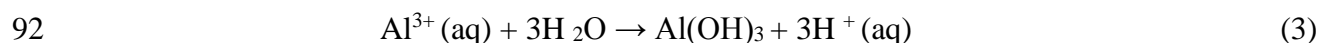
87 Anode:



89 Cathode:

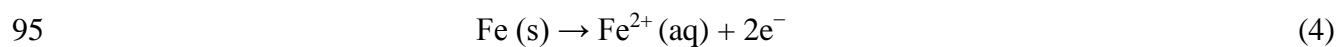


91 Overall:

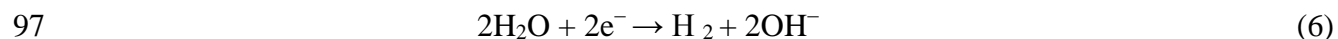


93 Iron electrode reactions during EC:

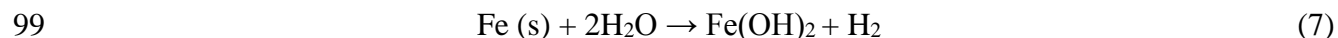
94 Anode:



96 Cathode:



98 Overall:



100

101

102 In literature, mostly efficiency of different electrodes including steel [17], Al [7], Mg [14] and Fe
103 [18] has been evaluated for the removal of COD, colour, total phosphorus and ammonia at different
104 pH and high values of current densities. There is a lack of literature regarding the complete study
105 of a suitable EC reactor, optimisation of different operating parameters, coagulant's nature effect
106 on flocs generation and calculation of the amount of suspended solids and micropollutants using
107 advanced scale wastewater treatment plant. Therefore, this study is of much worth, as it is about
108 the testing of a pilot-scale plant EC reactor for the removal of suspended solids and micropollutants
109 from the effluent's discharged from the wastewater treatment plant using green technology. Two
110 types of effluents from primary and secondary sedimentation tank have been used. Membrane
111 filtration technique (AFS) has been applied for the calculation of SS in effluents. Different flow
112 rate and current densities have been used to determine the optimum conditions and limitations of
113 this scale EC reactor. Furthermore, operational cost analysis of the EC reactor is determined to
114 verify the feasibility of the designed test EC reactor plant for micropollutant removal from the
115 wastewater.

116 **2 Experimental**

117 **2.1 Wastewater sampling and characteristic**

118 Wastewater samples were collected from a local wastewater treatment plant. Mainly two types of
119 samples depending upon their quality are used in the tests; one type was taken from the outflow of

120 primary sedimentation tank and the other was taken from the secondary sedimentation tank of the
121 wastewater treatment plant. Primary tank effluent has a higher amount of pollutant in comparison
122 to secondary tank effluent. Their wastewater characteristics are discussed in Table 1. Inflow and
123 outflow point is situated on the EC reactor from where samples were taken after constant
124 conditions of operation of the EC setup. Samples were not taken at once but with constant short
125 intervals of time, to ascertain the homogenous composition of the sample.

126 **2.2 Experimental assembly of EC reactor**

127 The experimental setup is schematically shown in Fig. 1. A 150 L open tank is used as a feed tank.
128 Some of the pollutants like suspended solids (SS) might settle during an experimental run, hence
129 the feeding 150 L tank is equipped with a stirrer for continuous stirring and maintaining a
130 consistent influent to the treatment system. The feed tank is fed with primary or secondary
131 sedimentation tank effluent one by one according to the experiment by changing the feed lines.
132 The effluent from the feed tank is fed to the EC reactor using a pump (range 250–1500 L/h) and a
133 flow meter connected in line to ensure the operating 500 L/h inflow to the EC reactor. The pilot
134 plant setup is mainly divided into two main parts. The first part consist of an EC reactor and the
135 second part consists of EF setup and EPD discs. The EC reactor is a fibre box having two chambers
136 in series provided with slits to insert the metal plates inside which act as electrodes. Each cell has
137 17 plates and on the whole, there are 34 plates of one material (either Fe or Al) used at a time in
138 the whole EC reactor. The slits inside the EC cells are fitted with plastic cylindrical objects of
139 about 3 cm in length and 3 mm in diameter to raise the height of the plates to ensure better
140 operation. The plates act as bipolar electrodes in a parallel connection and are connected to a DC
141 power supply through a polarity changer circuit. The polarity changer circuit has various time
142 settings to change the polarity on the connecting electrodes that ensure the proper functionality of

143 electrodes. In the bipolar systems, the side of the electrode facing the anode is negatively charged
144 and vice versa on the other side of the cathode. In EC reactor there is an inlet and an outlet, the
145 inlet is just a circular hole where the connecting pipe fits in to provide the inflow to the reactor.
146
147 The outlet of the reactor is provided with an overflow rectangular opening and a circular hole at
148 the bottom for the outflow. The EC reactor is provided with an auxiliary plastic glass cover, to
149 ensure a better view of the EC process inside the reactor and controlling the parameters by visually
150 viewing the process. The plates used inside the EC reactor are 300 mm × 155 mm × 3 mm of
151 dimensions and have an effective surface area of about 1.488 m² [3]. In this experiment, Al and Fe
152 electrodes are used separately by applying different values of current densities. The current density
153 applied for the Fe electrodes and Al electrodes range from 0.67–2.69 A/m² and 0.34–2.02 A/m²
154 respectively. The amount of suspended solids in case of Al electrodes agglomeration was carried
155 out nicely with lower current densities but for Fe electrodes higher current densities were required
156 for noticeable treatment. In the presence of these observations and due to economic factor, lower
157 current densities were applied for Al electrodes treatment. The inflow to the electrocoagulation
158 reactor was maintained at about 500 L/h and for some reactions 250 L/h. The pH of inflows was
159 maintained with NaOH and H₂SO₄, where required. The pH of the effluents was examined by the
160 help of a pH meter (Hanna Ins. 301). A typical pH meter consists of a special measuring probe (a
161 glass electrode) connected to an electronic meter that measures and displays the pH reading. There
162 is a bulb at the bottom of the probe, the bulb is a sensitive part of a probe that contains the sensor.
163 To measure the pH of a solution, the probe is dipped into the solution. The probe is fitted in an
164 arm known as the probe arm.

165 **2.3 Characterisation of wastewater samples**

166 Wastewater from the primary sedimentation tank was treated with Fe and Al bipolar electrodes to
167 examine the COD reduction efficacy of the test pilot plant at the above mentioned variable current
168 densities [6]. The COD of samples were calculated by the help of spectrophotometer (Hach Dr
169 5000, USA). While samples collected from the secondary sedimentation tank were examined for
170 their amount of SS substances before (inflow) and after (outflow) EC treatment. The outflow of
171 secondary tank had not been tested for COD removal, because of the characteristic lower level of
172 COD. The effluent from the primary sedimentation tank is treated at higher current densities to see
173 the effect of electrocoagulation on the COD removal efficiency. Similar to COD measurement,
174 amount of SS substances in samples were inspected by using separate Fe and Al coagulant, at
175 different current densities and resultant pH values to identify the conditions at which the EC set-
176 up work efficiently. The measurement of SS substances in the samples was done by using
177 membrane separation technique, according to DIN 19643-2 and is called after filtration solids
178 (AFS) [19].

179
180 According to DIN 19643-2 [19], the filtration done by a pre-weighed filter for a specific volume
181 of each sample at air pressure up to 6 bars. Some important steps are described here. Filtration is
182 done by a specific nitrogen pressure of 5 bars and during filtration the filter paper allows only the
183 particles which have the diameter of less than 0.45 μm . After filtration, the wet filter paper is again
184 dried in the oven for 1 hour at 105 °C temperature. It evaporates all the water content and collects
185 only the solids with it, which needs to reside in the desiccator to cool it down for 30 minutes.
186 Finally, again measuring the weight of the filter paper, the total SS are calculated in the treated
187 wastewater. Each sample was subjected to AFS setup three times and the average value was taken

188 for the experimental calculation. The AFS of inflow and outflow samples were made to compare
189 the quantitative analysis on the basis of flocs formation and coagulant dissolution.

190
191 After the calculation of AFS, the amount of coagulants dissolved during the generation of flocs
192 was calculated experimentally and theoretically to observe the reliability of the process.
193 Experimentally, to calculate the amount of Fe and Al coagulation dose in the outflows specifically
194 in the case of used pilot plant setup was a major task of this study. For the total amount of Fe in
195 the outflow, spectrophotometric analysis is performed while for Al in the outflow gravimetric
196 analysis are done [20]. Moreover, the theoretical amount of Fe and Al coagulation dose is
197 calculated according to equation (1) [21].

198
199
$$C_x = \frac{M_{total,x}}{V_{total,liquid}} = \frac{I \times t \times m_x \times N}{Q \times t} = \frac{I \times m_x \times N}{Q} \quad (1)$$

200
201 where C_x = dosage of released metal (Fe or Al) from the outlet of the electrolysis cell (mg/L);
202 $M_{total,x}$ = total mass of released metal (mg) by Faradays law within the retention time (t, s); $V_{total,liquid}$
203 = total volume of the effluent (L) within the retention time: I = applied current (amperes); N =
204 number of channels in the electrolysis cell; Q = flow rate (L/s), and m_x = electrochemical equivalent
205 of the metals. The electrochemical equivalent of aluminum metal is (0.093 mg/C) and of iron metal
206 is (0.193 mg/C).

207
208 The particle-size distribution (PSD) of a powder, or granular material, or particles dispersed in
209 fluid, is a list of values or a mathematical function that defines the relative amount, typically by
210 mass, of particles present according to size. The samples from the effluent streams after the EC

211 process with Fe and Al electrodes are subjected to PSD characterization. During the course of
212 experiments, every time when the samples were subjected to PSD, 1 litre sample was taken in
213 order to examine the resulting flocs in detail. The samples were taken separately for Fe and Al
214 electrodes at their respective amount of applied current densities. PSD is then performed with the
215 help of a laser granulometer, the Mastersizer 3000 from Malvern [22]. With this device, particles
216 can be measured with a size of 0.01 to 3500 microns. For the measurement, two light sources, one
217 red at 632.8 nm and other blue at 470 nm were used. At last, the operational cost analysis for the
218 optimum operating current densities of Fe and Al electrode was also performed to verify the
219 feasibility of the designed test EC reactor plant for micropollutant and suspended solid removal
220 from the effluents (equation 2). While the energy consumption for this process was determined by
221 using equation (3) [8].

$$222 \quad \text{Operating cost} = \text{energy cost} + \text{material cost} \quad (2)$$

$$223 \quad \text{Energy consumption} = \frac{V \times I}{Q} \quad (3)$$

224 Where "V" is the voltage applied on EC reactor, "I" is the current applied in Amperes on the EC
225 reactor and "Q" is the flow of the influent through the EC reactor in m³/h. Energy consumption is
226 then expressed in Wh/m³.

227

228 **3. Results and discussion**

229 **3.1 Treatment from primary sedimentation tank**

230 **3.1.1 COD removal efficiency of Fe and Al electrodes**

231 Applied current or current density is the most important operational parameter, in electrochemical
232 EC process. To test the role of current in COD removal, different current densities have been used

233 in case of Fe and Al electrode at the same initial pH of 7.30 and a flow rate of 500 L/h. In the case
234 of Fe electrode, basically, two types of settings were used. In the first run, the current was applied
235 at 3 A (current density= 2.02 A/m²) and in the second run current applied was 4 A (current density=
236 2.69 A/m²). It was observed with the samples that the flocs were formed within 5 minutes after the
237 samples were placed in the Imhoff cones (Fig. 2) and the COD was also measured afterwards. The
238 settling of the flocs formed at 3 A are shown in Fig. 2 as a function of time for settling. It can be
239 seen clearly from Fig. 2 that after 30 minutes of settling, the amount of solids in the outflow was
240 noticeably settled to an amount of 32 mL/L. On the other part, the suspension in the inflow was
241 still very stable and was nowhere near settling during the 30 minutes of reaction. The greenish
242 colour of the outflow after EC process indicated that the flocs formed contain purely Fe(OH)₂
243 which shows the dissolution of Fe electrodes in the form of Fe (II), also showed by [23-25].

244

245 The outflow samples were tested for the amount of COD removal, the results as shown in Fig. 3
246 that with 3 A, COD was removed from 233 to 100 mg/L and with 4 A, COD was removed from
247 250 to 109 mg/L. This shows a significant amount of COD removal from the water of the primary
248 sedimentation tank up to 56–57 % via Fe electrocoagulation as shown in Fig. 6, and also shows
249 that increase of current did not make a significant difference in the COD removal, as it is also
250 mentioned in the [26], that more COD removal with Fe electrodes was achieved at lower currents.
251 There have been studies where high removal of organic matter was obtained with iron electrodes,
252 such as [27] and [28]. However, the electric charge per volume has been really high .e.g; [28]
253 measured 70.5% removal of COD (from 254 to 75 mg/L) of COD from paper mill effluent when
254 they used 17280 C /L.

255

256 While in case of Al electrode, wastewater from primary sedimentation tank was introduced in the
257 EC reactor, the current was applied in the range of 1–2 A and the corresponding current density
258 values were 0.67-1.34 A/m². The COD removal as shown in Fig. 4 with 1 A applied current was
259 decreased from 121 to 107 mg/L and with 2 A current was decreased from 105 to 86 mg/L. The
260 sample treated with 1A applied current was put in the Imhoff apparatus to see the settling effects
261 of the solids and watched for a 30 minutes interval. It can be seen in Fig. 5 that there was no
262 significant colour imparted to the outflow and the settling with a half-hour interval was quite good
263 indicating the amount of flocs produced which is almost doubled before and after EC process. The
264 results showed a COD removal in the range of 12–18% which is almost 3 folds less than the COD
265 removal achieved in the case of Fe electrodes as shown in Fig. 6. It is stated in [24, 26] that Fe
266 electrodes are more efficient than Al electrodes in colour and COD removal.

267

268 **3.2 Treatment from secondary sedimentation tank**

269 **3.2.1 Effect of current density on AFS in Fe coagulation**

270 When Fe electrodes were used and the wastewater from secondary sedimentation tank was
271 introduced in the EC reactor, the current was applied in the range of 1-3 A and current density was
272 changed from 0.67–2.02 A/m². Different operating conditions had been opted for secondary
273 sedimentation tank effluent analyses as because of their changed characteristics. Higher current
274 densities were applied in primary sedimentation tank analysis in comparison with lower values of
275 current densities of secondary sedimentation tank analysis. This is because the higher amount of
276 contaminants were detected in the primary sedimentation tank, while very little values were
277 observed in secondary tank effluents Table 1. According to AFS amounts found in the inflow and
278 outflows after the EC reactor, it can be seen in Fig. 7 that the amount of AFS produced is directly

279 proportional to the amount of current applied. The higher amounts of outflows in comparison to
280 the inflows indicated that a large amount of electrode dissolution happened during the course of
281 the EC process. Also, it is showed in [23, 29] that dissolution of the Fe electrodes is the primary
282 reaction at anodes during typical current densities when the pH approaches neutral values. Each
283 experiment was run thrice to check the reproducibility of the results and named as WW-Fe-01,
284 WW-Fe-02 and so on. Similar was the case with Al coagulation.

285

286 In Fig. 8 coagulation dose of Fe, theoretical amount of Fe and AFS in the outflows are compared
287 against the different current densities values. The measured amounts of Fe in the outflow based on
288 the equation (7), where one mole of Fe (s) reacts with two moles of water to produce one mole of
289 $\text{Fe}(\text{OH})_2$. Here it was assumed first that all the iron electrodes produce $\text{Fe}(\text{OH})_2$ precipitates which
290 were dirty green in colour and the amount of which were then tested by Hach Lange test cuvettes,
291 to prove the fact that the whole amount of Fe which produced as coagulation dose by Fe electrodes
292 is in the form of $\text{Fe}(\text{OH})_2$.

293

294 According to the results, current density has a direct effect on the dissolution rate in the studied
295 range (0.67–2.02 A/m^2 or 7.19–21.58 C/L) (Fig. 9). Iron dissolved at the rate of 2.93 mg Fe/C,
296 while the theoretical value for dissolution of Fe (II) is 3.09 mg Fe/C. It can be concluded that iron
297 dissolved in Fe (II) form and dissolution followed Faraday's law. This was consistent with the
298 results of other researchers [23-25]. This result is very important because it is established that Fe
299 (II) is a poor coagulant and should be oxidized to Fe (III) form before it is employed to remove
300 organic matter [1].

301

302 In order to test the volumetric inflow parameters of the EC reactor, a set of experiments is done
303 with a low flow rate of about 250 L/h, the results of which are compared to the inflow rate of 500
304 L/h. The amount of AFS obtained during different flow rates is shown in Fig. 10. The results in
305 Fig. 10 clearly show that when the inflow rate is decreased from 500 to 250 L/h, the amount of
306 AFS is increased to almost doubled, provided the current applied remains constant. These results
307 also support the arguments, that if charge per litre volume of inflow is increased the amount of
308 AFS and hence the coagulation dose of Fe is also increased linearly. In other words, if inflow rate
309 to EC reactor has an indirect effect on the AFS produced provided the current density is kept
310 constant. But this rapid agglomeration couldn't be helpful, as it needs more coagulation dose and
311 might not be cost effective. Therefore, this parameter was not further optimised.

312 **3.2.2 Effect of current density on AFS in Al coagulation**

313 When Al electrodes were used and the wastewater from secondary sedimentation tank was
314 introduced in the EC reactor. The current was applied in the range of 0.5–1.0 A and current density
315 changed from 0.34–0.67 A/m². The amount of AFS found in the inflow and outflow; before and
316 after the EC process shown in Fig. 11 indicates that the current applied has a direct effect on the
317 amount of AFS produced. The electrodes dissolution is the primary reaction at anodes, the same
318 is the case with aluminium electrodes.

319

320 In Fig. 12 coagulation dose of Al, theoretical amount of Al and AFS amounts in the outflows are
321 presented corresponding to the range of current densities. The measured amount of Al based on
322 the reaction (1–3), where one mole of Al_(s) reacts with three moles of water to produce Al(OH)₃
323 and afterwards polymerized to other hydroxide species [6]. On the first instance, it is assumed that
324 the aluminium electrodes produce on the whole Al(OH)₃ which is then experimentally proved by

325 the gravimetric test. The test was performed at 1 A current and 0.67 A/m² current density applied
326 to the EC reactor. The amount of Al based on the gravimetric measurement, AFS by the EC process
327 and calculated theoretically by following equation (1).

328
329 According to the results, the current passing through the EC reactor was directly proportional to
330 the dissolution rate [10] and followed Faraday's law. In the studied range (0.34–0.67 A/m² or 2.45–
331 4.82 C/l), Fig. 13 the dissolving rate of Al measured was 1.27–2.39 mgAl/C, whereas the
332 theoretical rate according to Faraday's law was 2.22 mgAl/C. It can be concluded that the amount
333 of dissolution of Al at lower values of current densities was lower than the theoretical values but
334 approaches equal to theoretical values at higher current densities. When the experiments run for
335 Al electrodes at 250 L/h, the same results were obtained as shown in Fig. 10.

336 **3.2.3 Outflows pH**

337 In addition to the applied current, another factor that influences the reactions inside the EC reactor
338 is pH. Literature confirms that pH of the outflows changes in comparison to inflows as reaction
339 taking place at cathode (H₂ evolution) in EC reactor, moreover it also depends on inflow's pH and
340 type of selected anode [1, 5, 6]. In the previous section, it is noted that with an increase in current
341 density amount of outflow AFS is increased. pH factor was also observed, before and after the EC
342 process under the applied operational current densities and resultant AFS for both electrodes. The
343 pH of outflows is observed a correspondent increase with current density for all experiments [6].
344 In all the experiments performed in both the series, with Al and Fe electrodes, the pH was slightly
345 increased in the outflows. The inflow's pH for all run at variable applied current, in case of Fe is
346 7.07, and while in case of Al is 7.24. The increase of pH was more in the case of Fe electrodes
347 (Fig. 14) than to Al electrodes (Fig. 15), is in accordance with applied current values.

348 **3.2.4 Flocculation analyses**

349 In the EC process, hydrogen bubbles are produced on the cathodes (reaction 2 and 6) [30]. The
350 production of visible hydrogen bubbles on the electrodes indicates that the EC process is going
351 well. Electrodes material and current density both have an effect on the bubble size and amount.
352 It is observed that very small hydrogen bubbles are produced in both the cases when aluminium
353 or iron electrodes are used [5, 10], according to these, smallest hydrogen bubbles are produced
354 during EC process at neutral or acidic pH. Current density has a direct effect on the bubbles
355 amount, the more the current density, the more bubbles were produced on the electrodes. The gas
356 bubbles produced can carry some of the flocs to the top of the surface of the electrodes, and then
357 the layer can be removed from there in the form of foam.

358
359 The flocs layers produced due to the hydrogen bubbles can be seen in Fig. 16. It shows the layers
360 formed during the EC operation with Fe electrodes and aluminium electrodes simultaneously. The
361 sludge formed during the EC process is settleable and during the initial test with only the wastewater
362 from the secondary sedimentation tank, it was figured out that sludge sometimes settles down in
363 the EC chambers. So, it was decided to empty the chambers and refill it with water at the end of
364 each day operation. So each time the water is replaced from within the EC cell, the sludge moved
365 out of the EC reactor. At the first instance, a little amount of sludge deposited on in the chambers
366 of the EC cell.

367
368 The samples from the effluent streams after the EC process with Fe and Al electrodes are subjected
369 to PSD analysis in the laser granulometer. Fig. 17 shows particle size distribution (PSD) for
370 samples in which Fe and Al electrodes are used for coagulation at 0.5–2 A applied current values.

371 This PSD analysis shows the cumulative volume % of the particles against the particle diameter in
372 μm . From Fig. 17, it confirms that a higher amount of current produced the bigger amount of flocs
373 with both electrodes, which later on could be separated by secondary separation technique. In the
374 case of Fe electrodes while increasing applied current more amount of flocs are observed while no
375 such increment has been observed in the case of Al electrode. The comparison of PSDs of Al with
376 Fe showed that, at the same value of applied current, Al generates more flocs than Fe.

377 **3.3 Cost analysis of optimum current densities**

378 The operating cost for the current EC process is calculated with equation (2). The energy
379 consumption is then expressed in Wh/m^3 . The energy consumption costs for Fe and Al electrodes
380 at 2 A and 1 A operation are found to be 0.03 €/m^3 and 0.02 €/m^3 . From this it is observed that
381 energy consumption is a direct relation with applied current, the greater the applied current the
382 higher will be the consumption of energy [5]. Based on the measured coagulant doses of Fe and
383 Al electrodes as described in the previous section, the amounts of Fe and Al electrodes consumed
384 per day were found $0.011 \text{ Kg}/\text{m}^3$ and $0.036 \text{ Kg}/\text{m}^3$ at 2 A for Fe and 1 A for Al. Based on these
385 optimum conditions, the used metal plates in the EC reactor can last 86 and 99 days respectively
386 for Fe and Al electrodes at its full capacity of operation at 2A and 1A. The material cost of Fe, Al
387 operating plant at 2 A, 1 A was calculated to be 0.01 €/m^3 and 0.02 €/m^3 respectively. According
388 to reaction (6), the operating cost of the EC operation for Fe and Al electrodes at the mentioned
389 current values of 2 A and 1 A is calculated as 0.04 €/m^3 and 0.03 €/m^3 respectively.

390

391 **4. Conclusions**

392 This study is dealing with the wastewater treatment on a pilot-scale plant set up by electrochemical
393 technology. The electrocoagulation and electroflotation techniques were applied with a revived
394 design to meet the need of the process to remove suspended solids and micropollutants out of the
395 wastewater stream. By using various settings of applied currents, the process is tested for the better
396 quality of effluent using water from the outflow of primary sedimentation tank and treated water
397 from the outflow of secondary sedimentation tank, and results are optimized for a better
398 downstream separation process. Wastewater from the primary sedimentation tank is treated both
399 with Fe and Al electrodes. In the case of Fe electrodes, more than 50% of COD removal is observed
400 which was about 12–18% with Al electrodes. The coagulation dose of Fe and Al electrodes are
401 measured, on the basis of AFS produced and according to Faraday's Law. With both Fe and Al
402 electrodes, the coagulation dose went on increasing with increasing current density. The best
403 setting for Fe and Al electrodes in terms of current and current densities for effluent from
404 secondary sedimentation tank are 3 A ($2.02 A/m^2$) and 1 A ($0.67 A/m^2$) respectively. The higher is
405 the coagulation dose, the bigger are the flocs in size and hence can easily settle down in
406 downstream sedimentation tank. The operating cost of operation for Fe and Al electrodes at the
407 most feasible mentioned current values of 2 A and 1 A are calculated as 0.04 € /m^3 and 0.03 € /m^3
408 respectively. The work provided good results and comparison with the past work done in this
409 regard by other researchers and provided some new space and enhancements, which can be done
410 to the process for continuing research in this regard. In the future, combination of electrode
411 materials and natural coagulants shall be a good option for efficiency improvement of this reactor
412 at pilot scale.

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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

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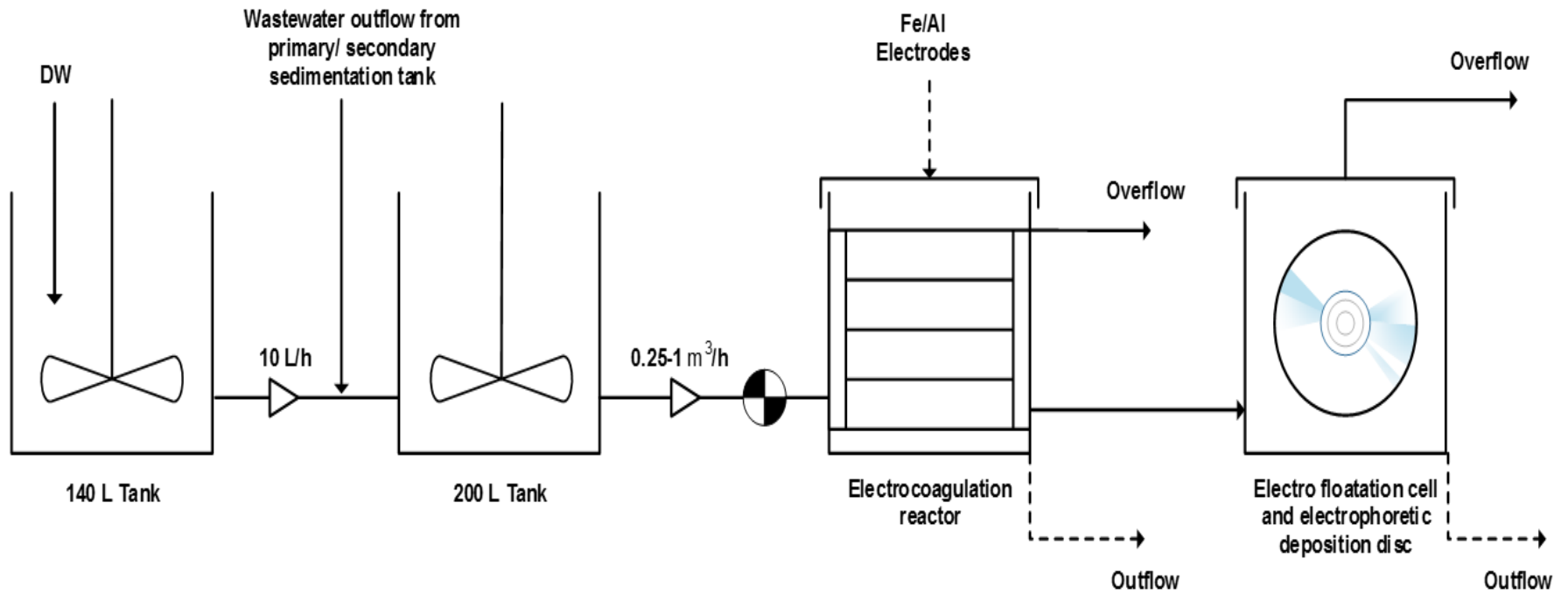


Fig. 1. Electrocoagulation pilot plant process flow diagram.

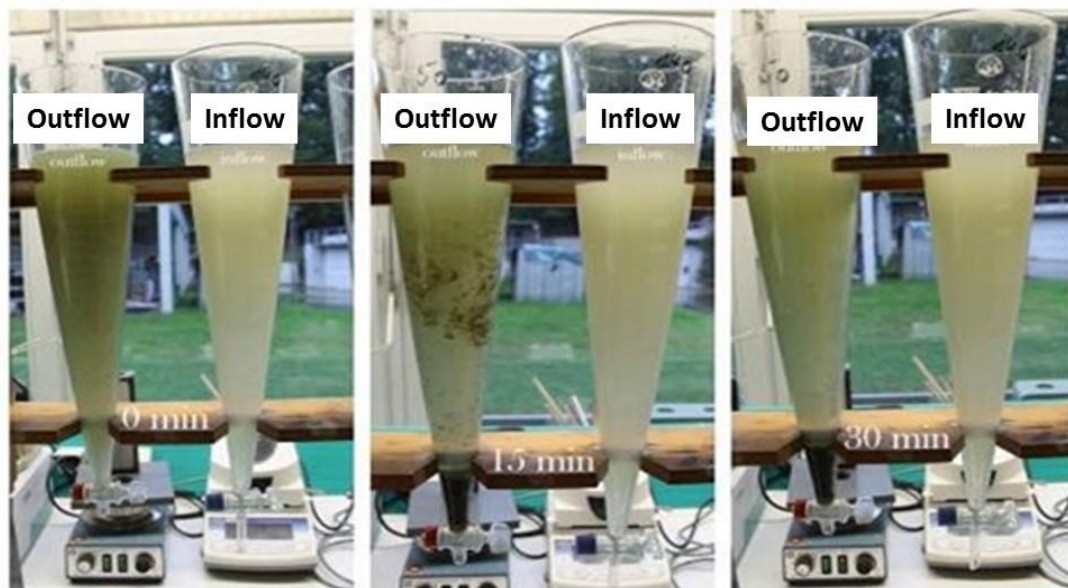


Fig. 2. Change of colour with Fe electrodes before and after EC treatment with time.

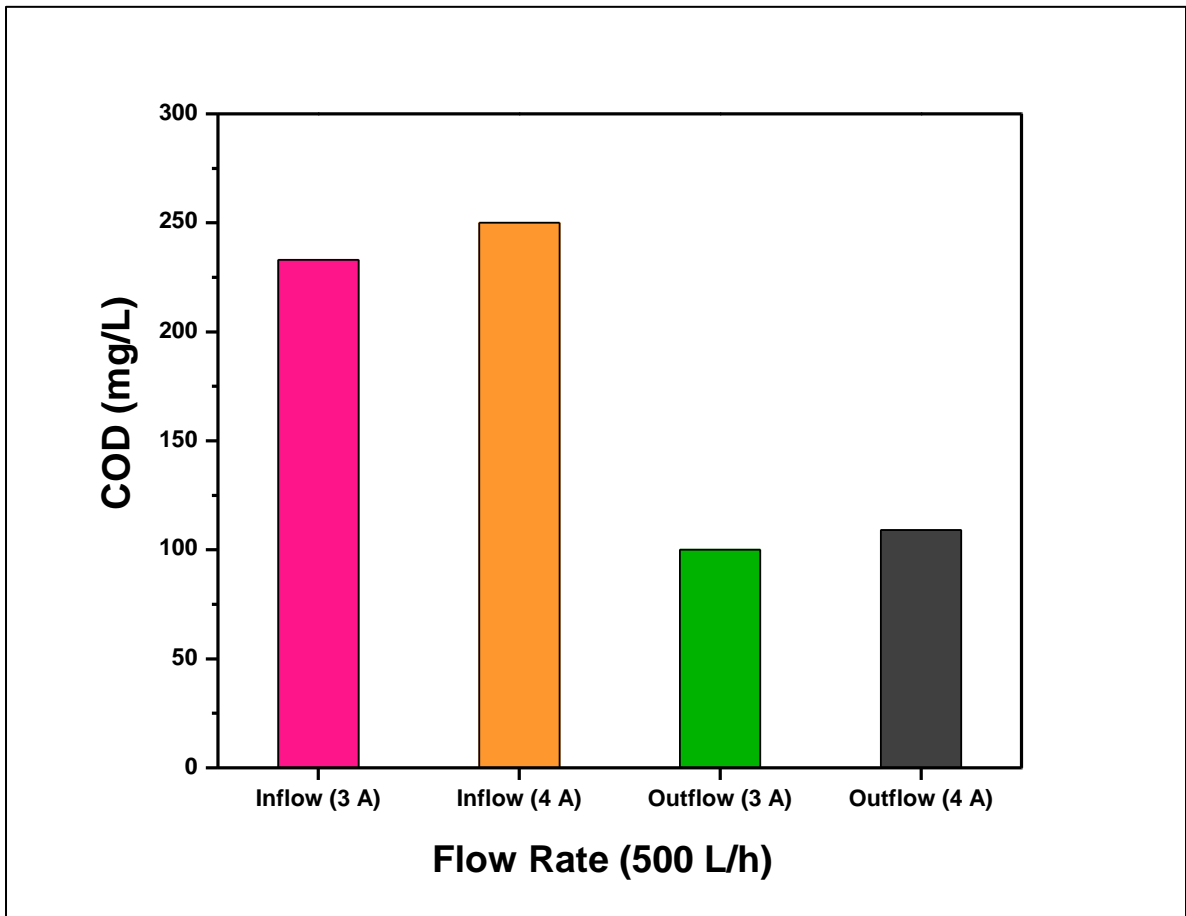


Fig. 3. COD removal of wastewater from primary sedimentation tank using Fe electrodes.

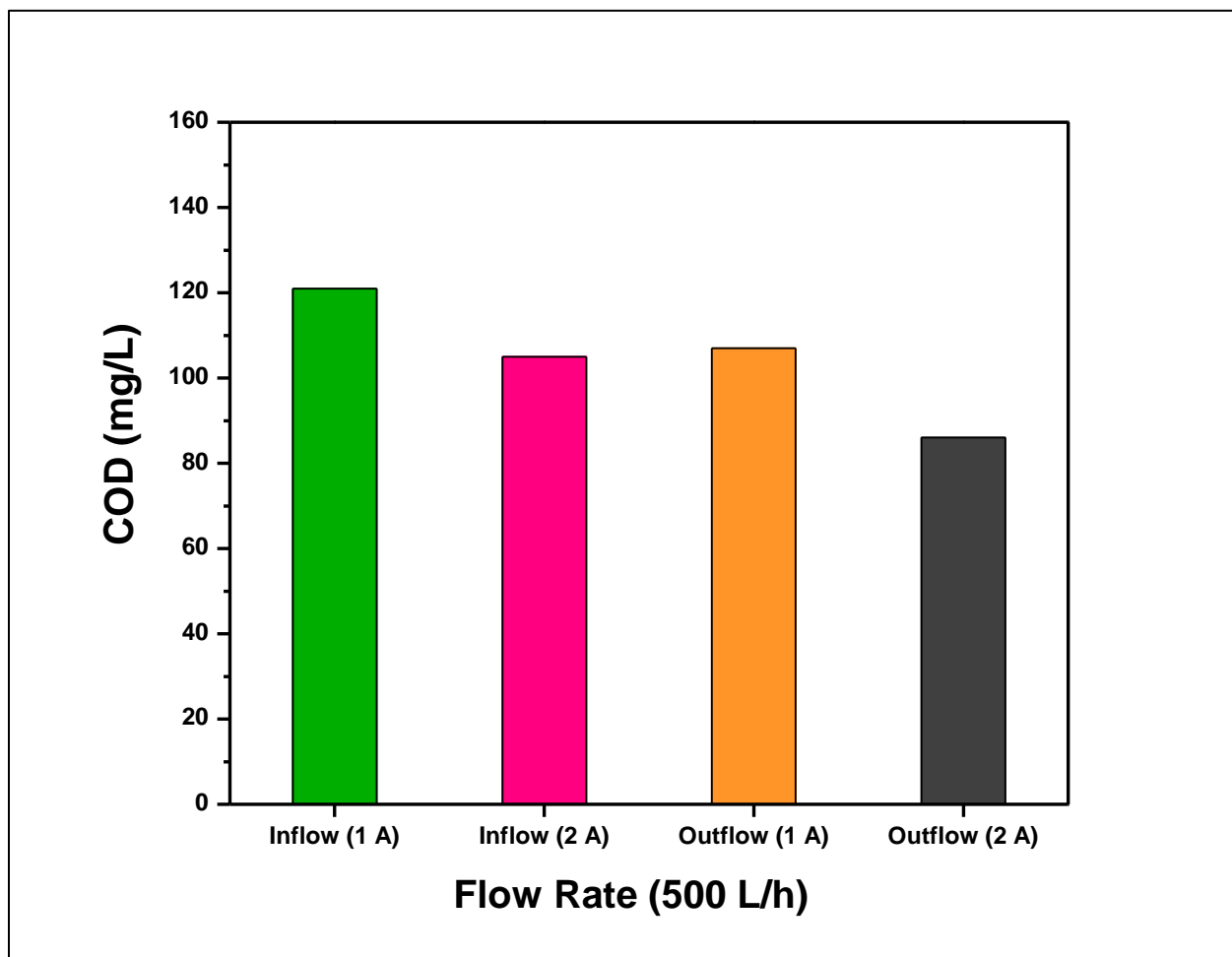


Fig. 4. COD removal of wastewater from the primary sedimentation tank using Al electrodes.

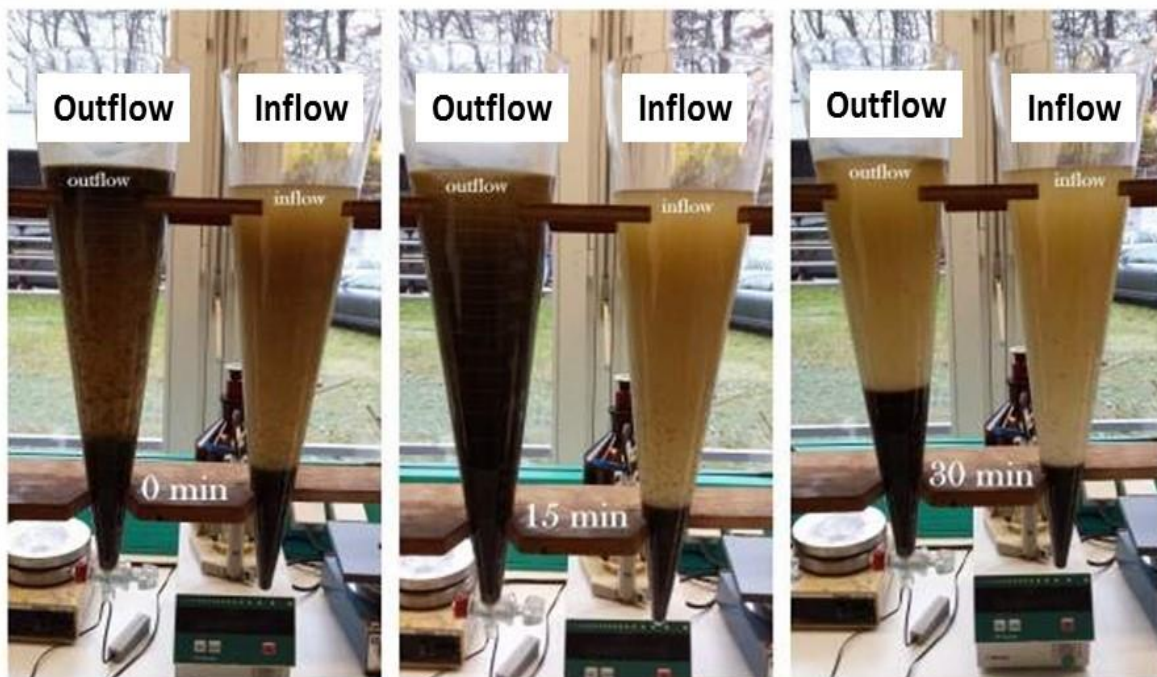


Fig. 5. Change of colour with Al electrodes before and after EC treatment with time.

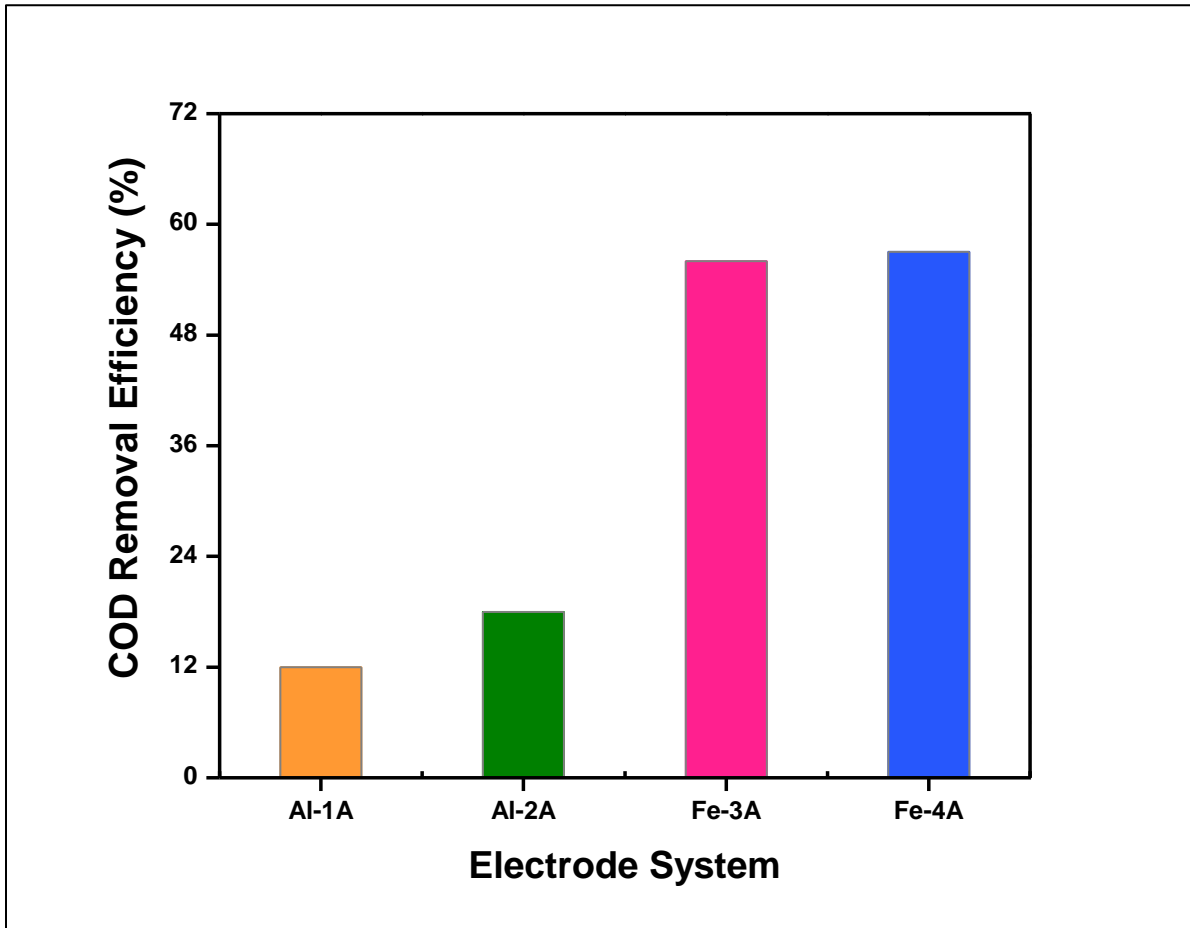


Fig. 6. The COD removal efficiency of Fe and Al electrodes at selected current densities.

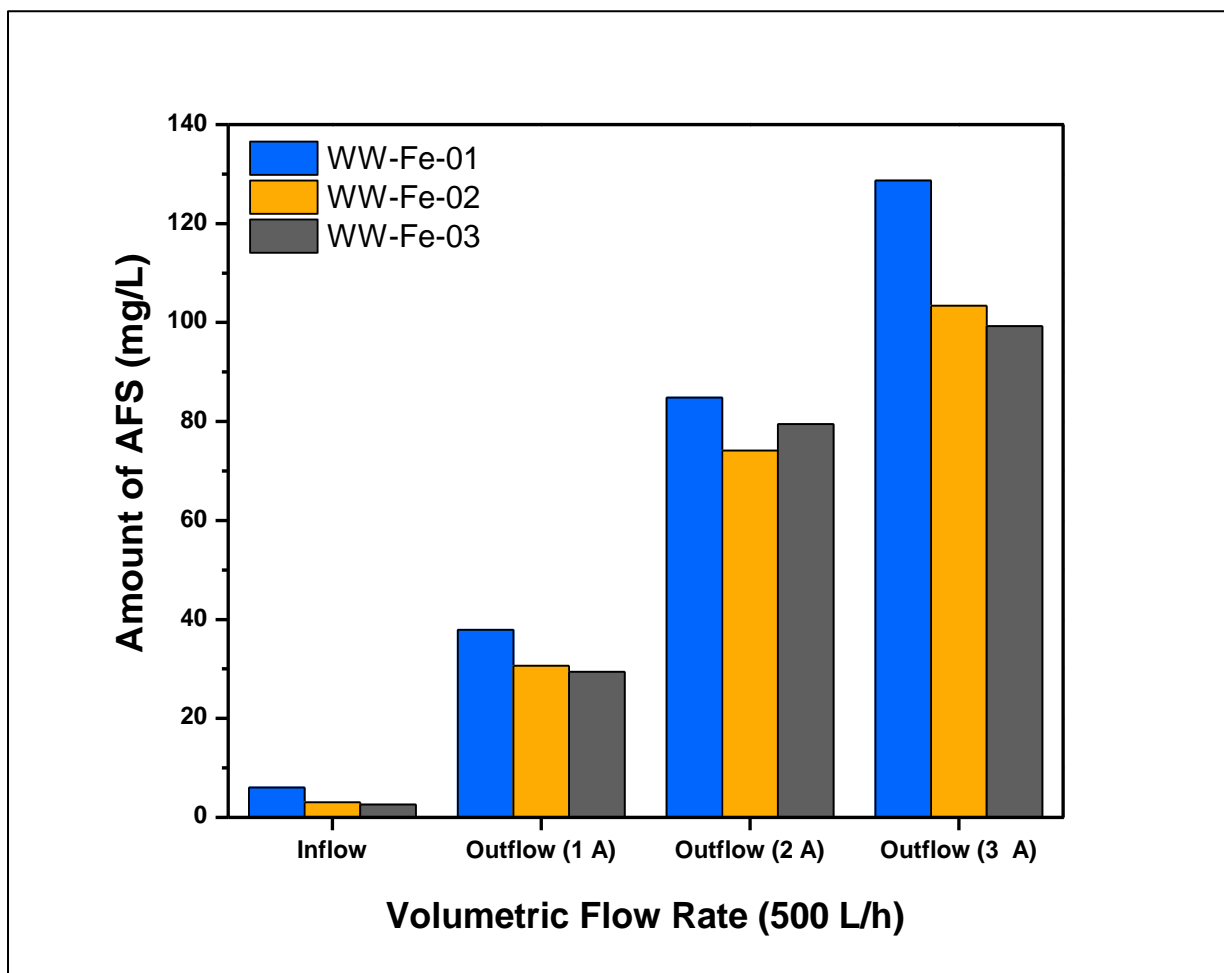


Fig. 7. Amount of AFS after applying current from 1–3 A on the EC reactor.

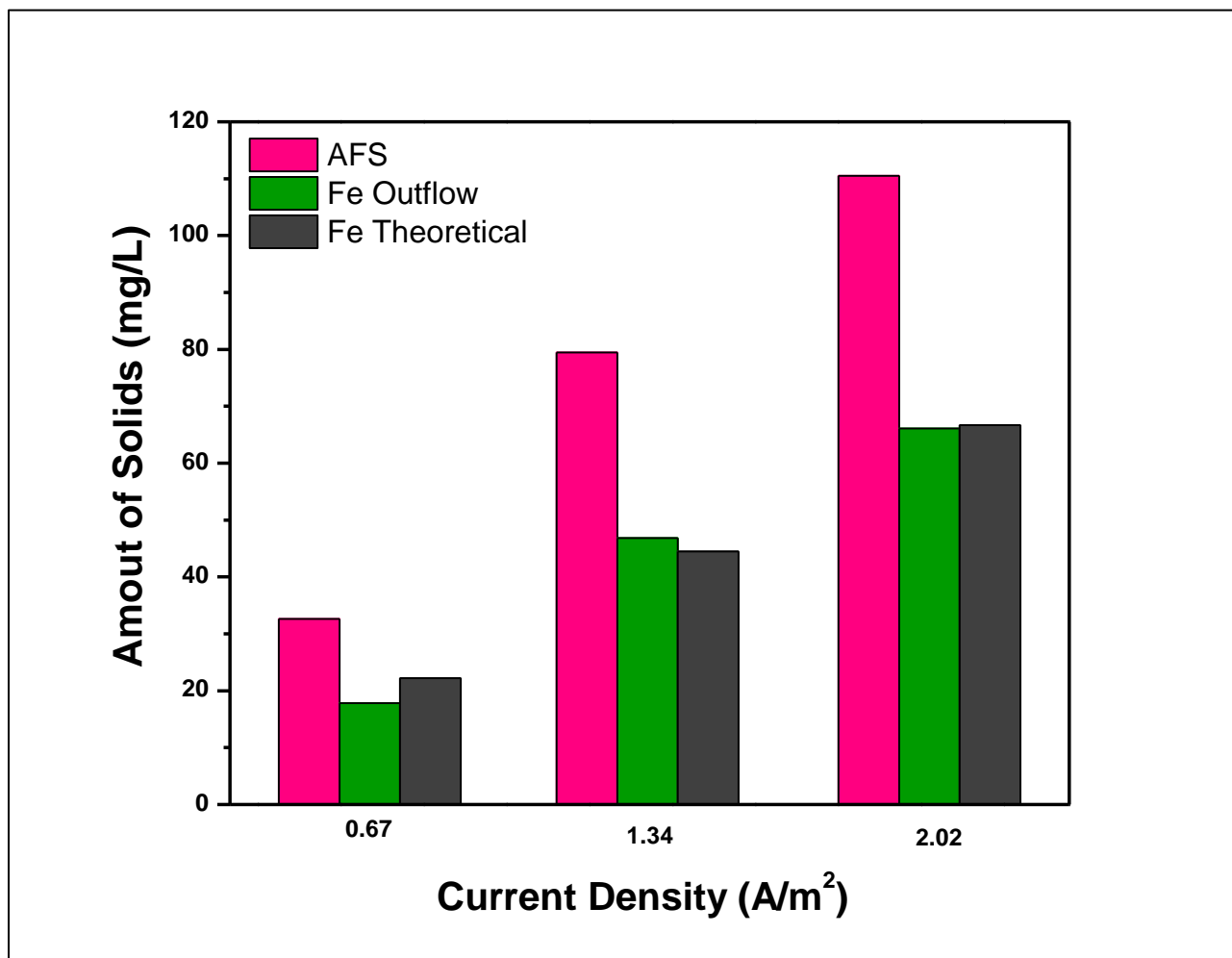


Fig. 8. Amount of AFS, Coagulation dose of Fe and theoretical amount of Fe in the outflow against various amount of current densities.

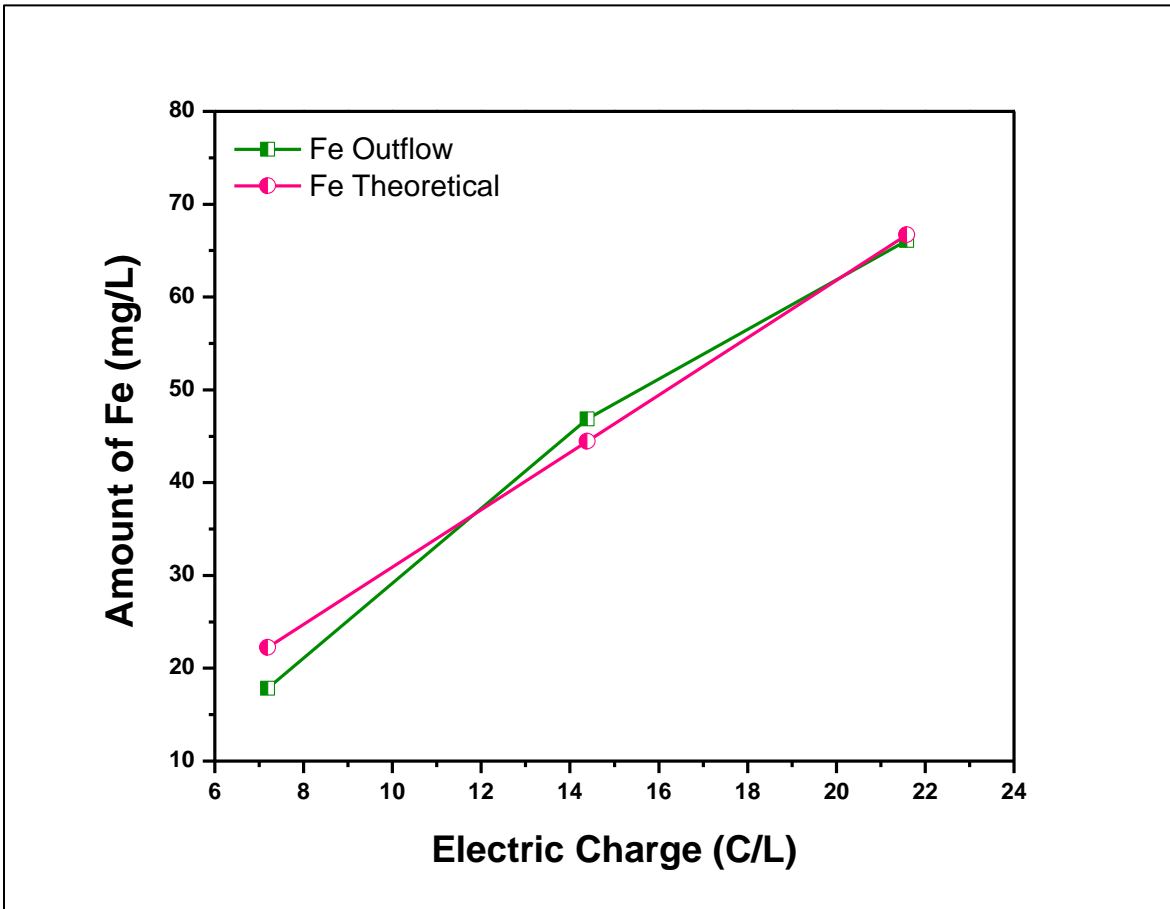


Fig. 9. Amount of Fe in the outflow and theoretical amount of Fe against the charge per litre of influent stream.

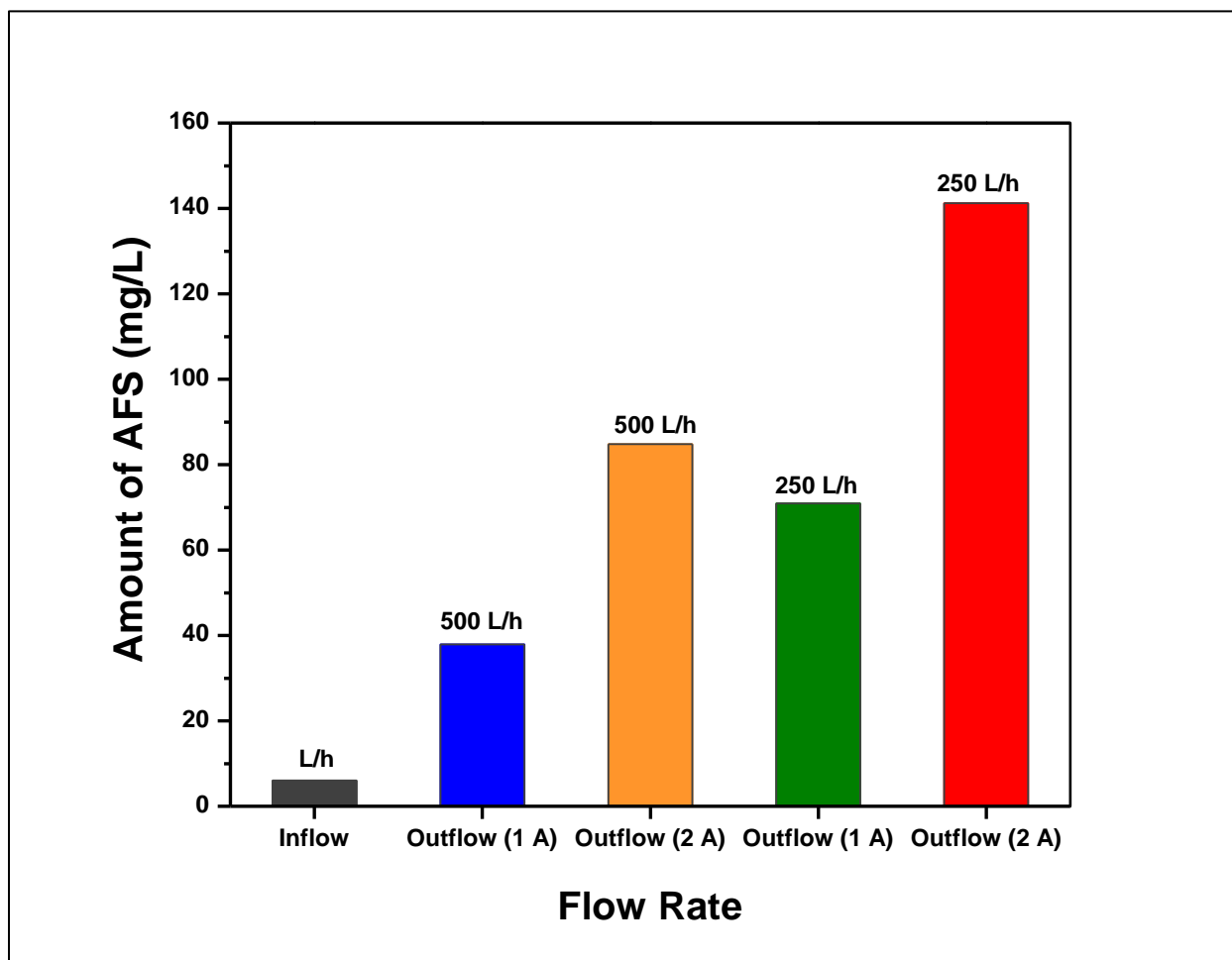


Fig. 10. Amount of AFS determined in the outflow after varying the current from 1–2 A and volumetric inflow rate from 250–500 L/h through the EC reactor.

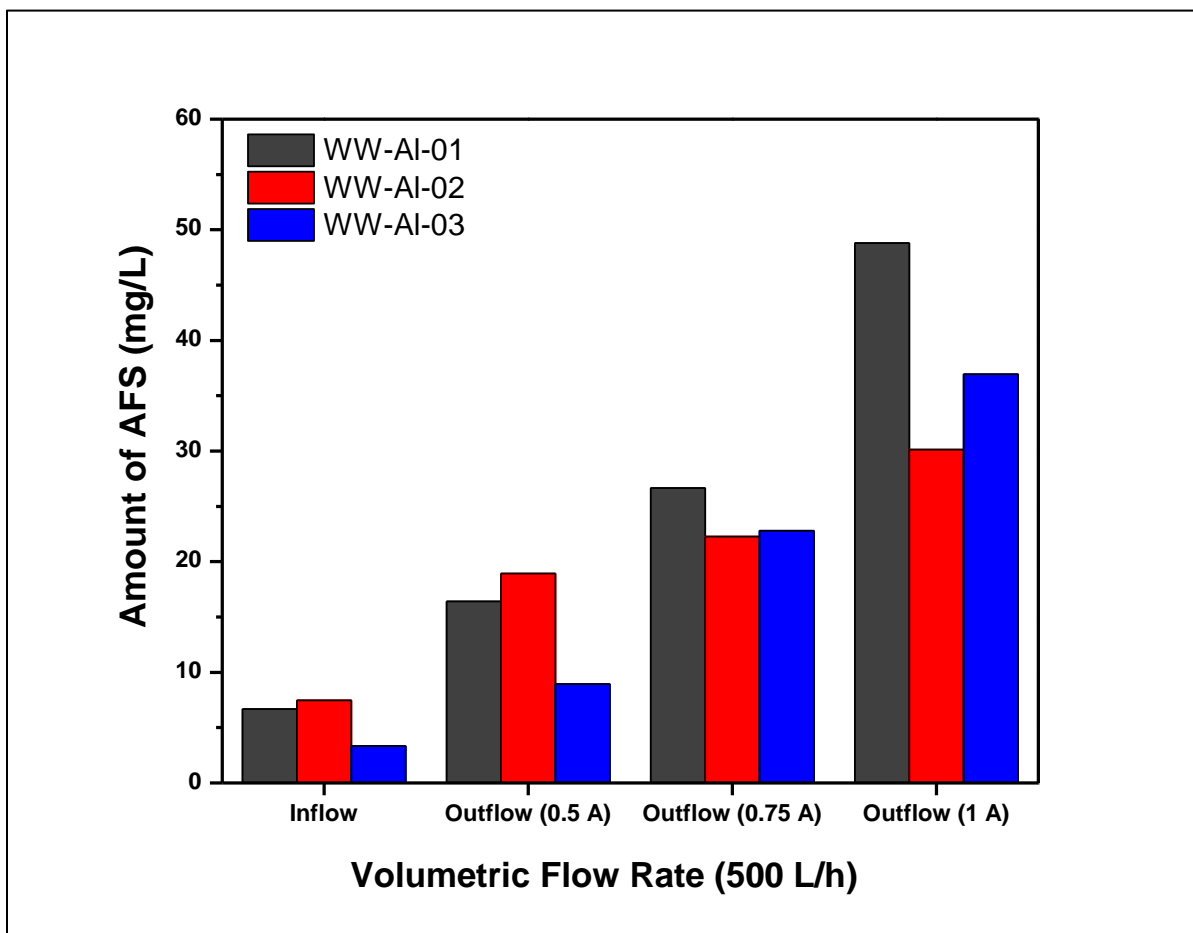


Fig. 11. Amount of AFS determined in the outflow after applying current at 0.5 A, 0.75 A and 1 A through the EC reactor.

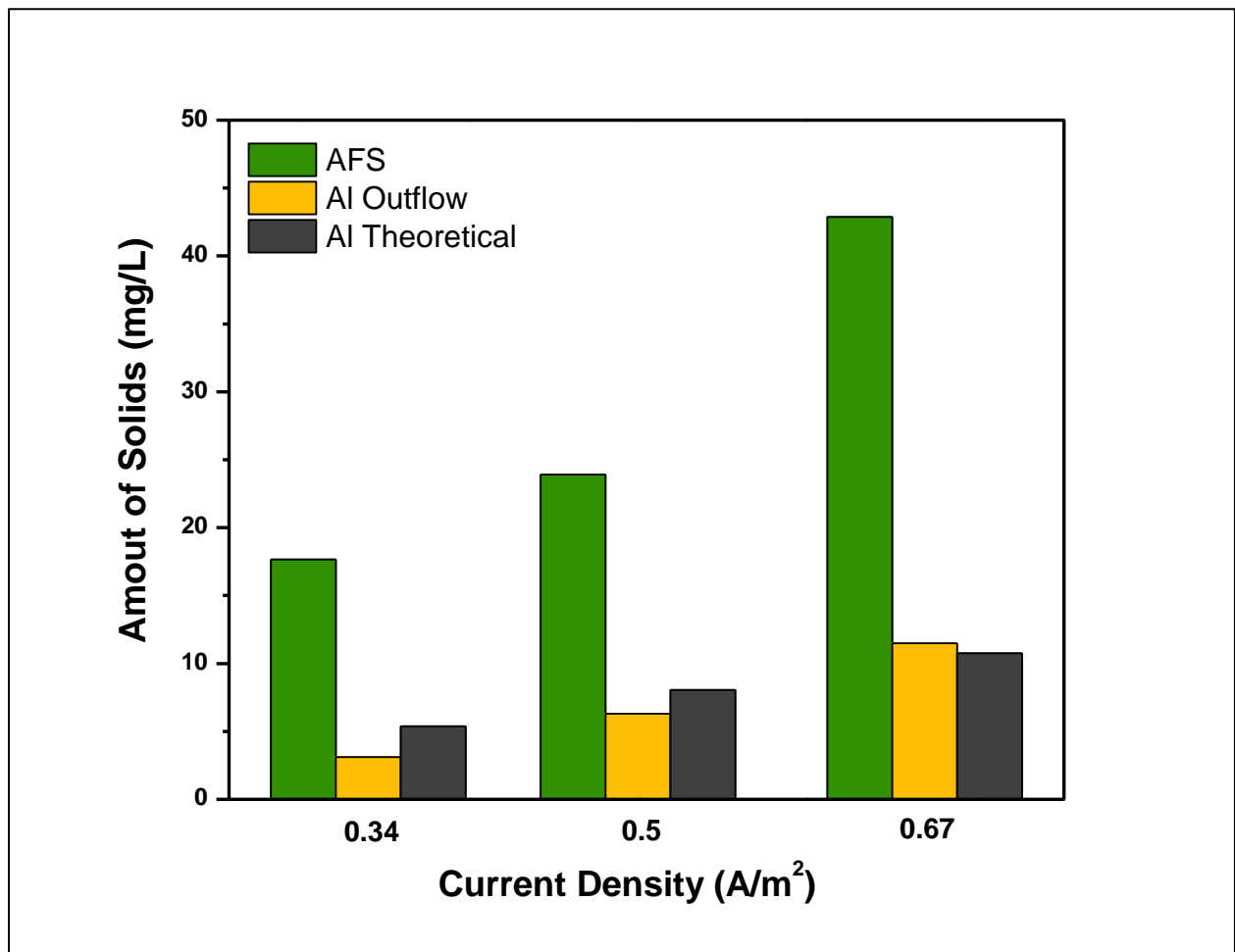


Fig. 12. Amount of AFS, Coagulation dose of Al and theoretical amount of Al in the outflow against various amount of current densities.

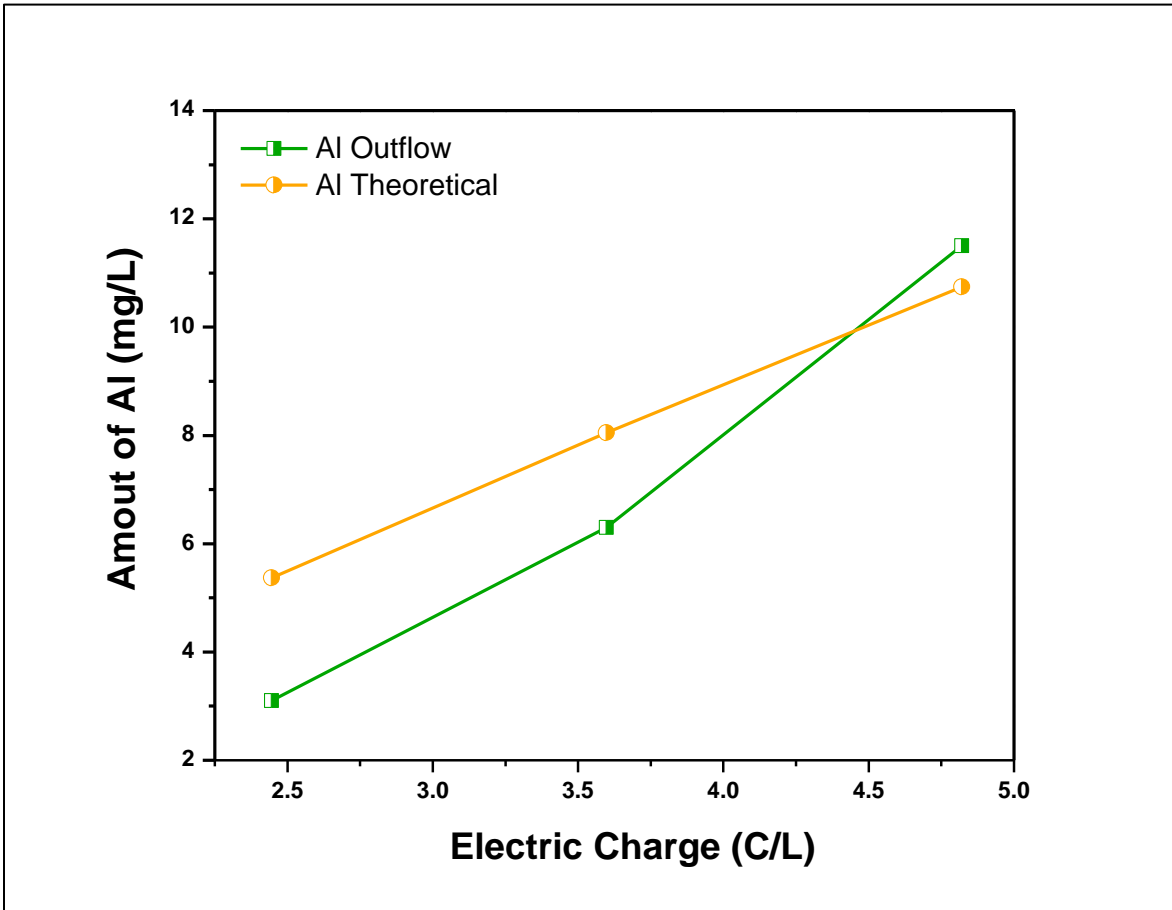


Fig. 13. Amount of Al in the outflow and theoretical amount of Al against the charge per litre of influent stream.

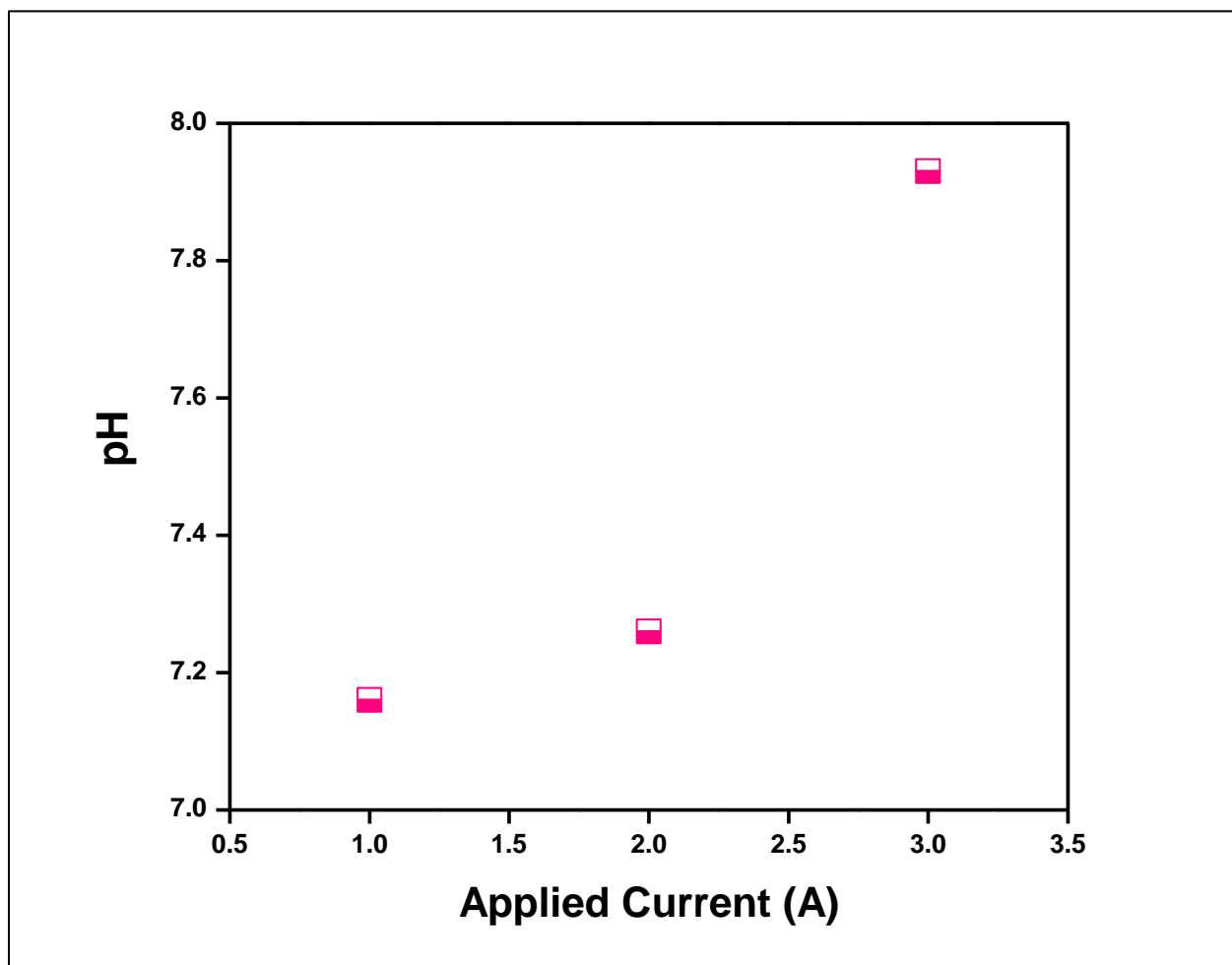


Fig. 14. Effect of applied current on outflow pH in the case of Fe electrodes EC treatment.

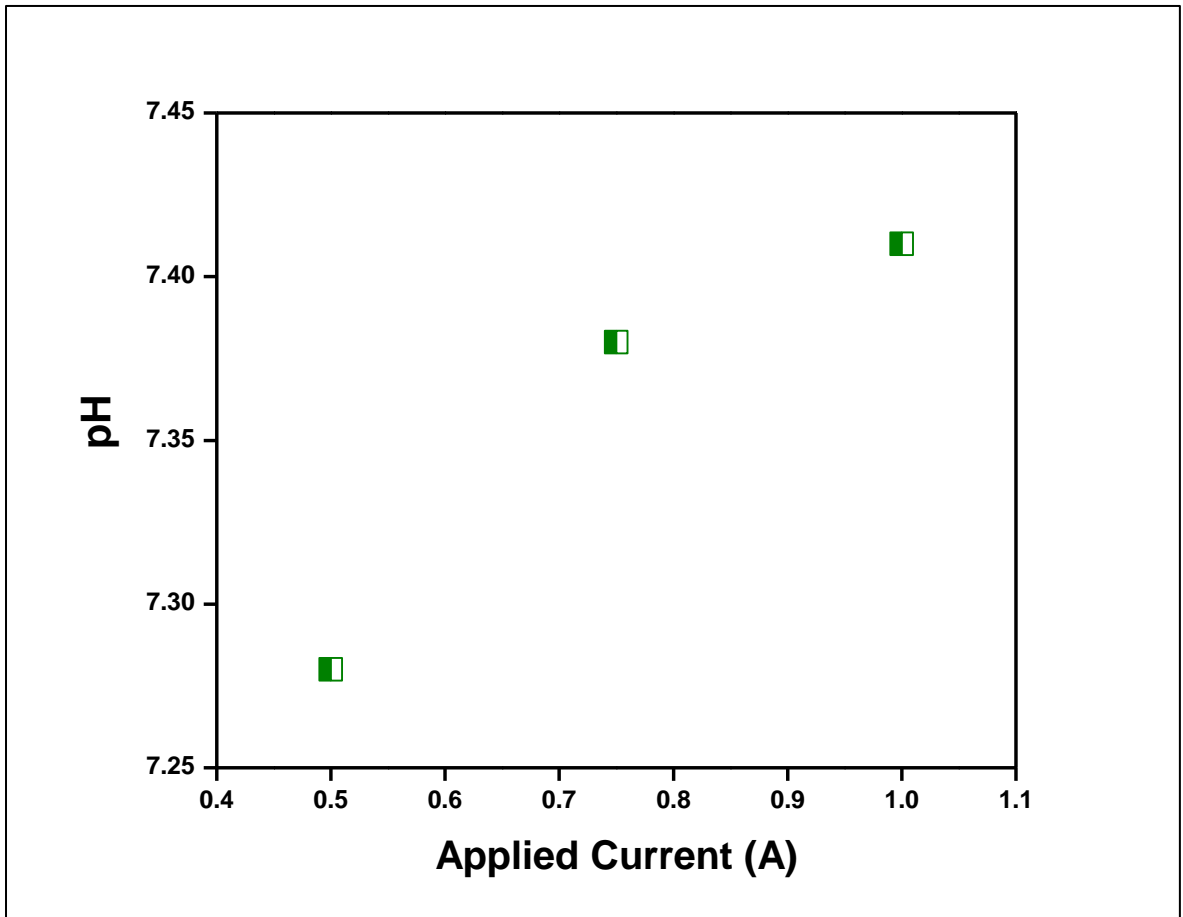


Fig. 15. Effect of applied current on outflow pH in the case of Al electrodes EC treatment.

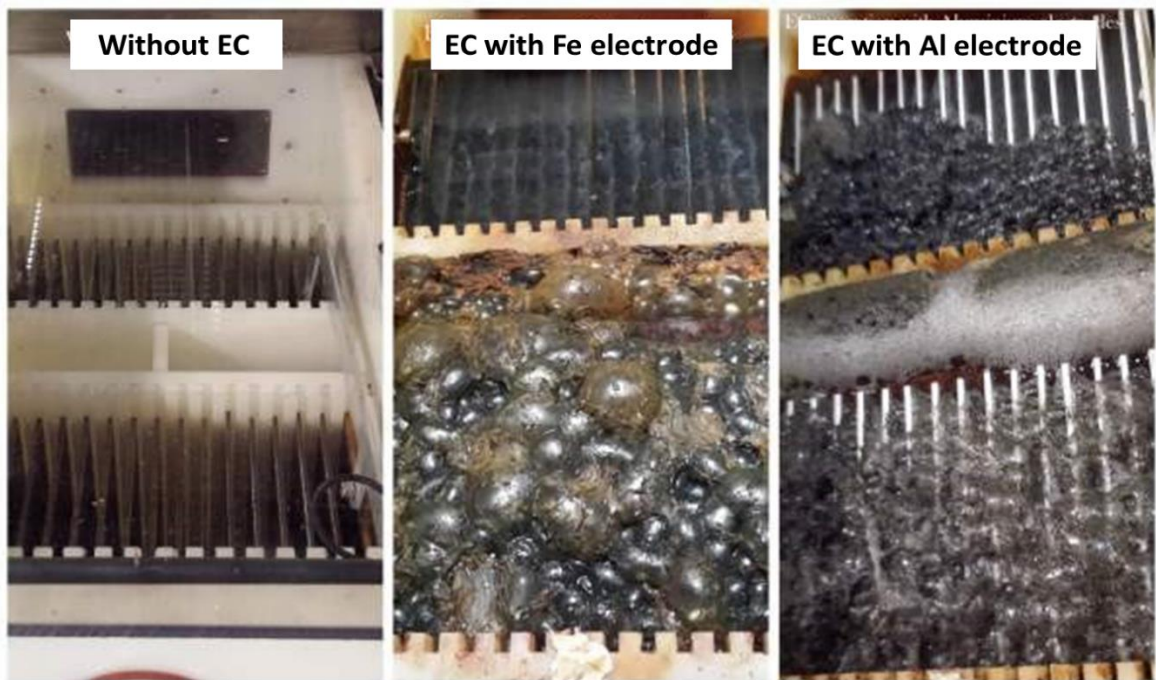


Fig. 16. Flocs layers formed due to hydrogen bubbles with or without EC treatment.

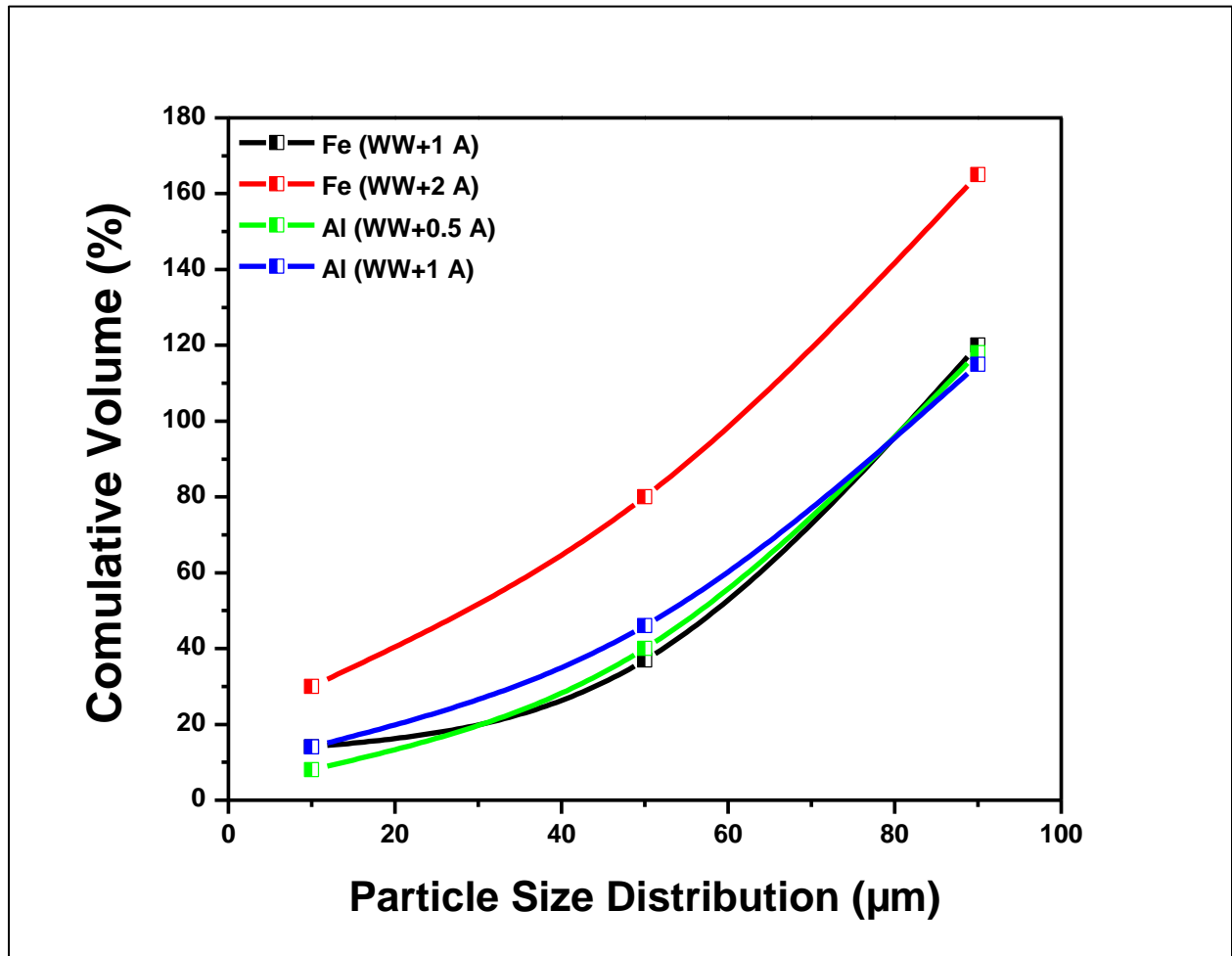


Fig. 17. The particle size distribution of Fe and Al flocs generated by EC reactor.