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Implications of advanced wastewater treatment: electrocoagulation and electroflocculation of effluent discharged from a wastewater treatment plant

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Highlights

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

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Graphical Abstract



Abstract

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

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

1 Introduction

The removal of anthropogenic micropollutants emitting from industrial, agricultural, domestic and urban sources is one of today's major challenges. The number of such micropollutants is significantly large and many of these are found in excessive quantities. Furthermore, pathogenic, non-pathogenic organisms, pharmaceutical and drug residues viruses and vaccines present in large amount in wastewaters. These micropollutants can have direct and indirect effects on the living organisms by bio magnification along the food chain. All mentioned sources of wastewater produce a huge amount of pollutants; organic carbon, biochemical oxygen demand (BOD) and chemical oxygen demand (COD), suspended solids (SS), total nitrogen and phosphorus contents [1, 2]. The main treatments till now available for the removal of these anthropogenic pollutants and micropollutants involve aerobic biodegradation, filtration, flocculation, adsorption, froth floatation and electrocoagulation (EC) [3]. However, some other methods have been applied in combination to overcome the shortcomings of an individual process including; photo-electro-Fenton, electro-Fenton and electro-oxidation [4]. EC is a highly studied process in this field these days. It is a multistep process, which involves the agglomeration of contaminants via electrodes (coagulant) through redox reaction carried out by applying an electric current. Conventional electrocoagulation process was carried out with the help of inorganic chemical species as coagulants including (NH₄)₂SO₄ and FeCl₃.

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

Three stages of EC technology implicate in the purification of wastewater are as: (1) electrolytic oxidation of sample water to generate coagulants; (2) pollutant destabilization, emulsion, deterioration and particle suspension; (3) agglomeration of resultant particles to generate flocs: comprised of colloids entrapped sludge blanket formed from coagulation reaction [10]. These flocs are similar to chemical flocs and are larger, acid-resistant, bounded less water and stable, that's why can be easily separated out by rapid sand filtration [11, 12]. The EC process is economical as it produces relatively less amount of sludge in comparison with conventional processes [13]. The electrochemical reactions take place at anode and cathode as discussed in equations (1–6). During EC, H₂ is evolved at cathode in the form of bubbles, that discards particles by flotation known as electroflotation [14]. Furthermore, numerous hydroxide of metallic electrodes (anode: Fe/ Al) and coagulant compounds are generated from these chemical reactions to deteriorate coagulate and adsorb pollutants [11]. Aluminium and iron electrodes are widely used

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

Aluminum electrode reactions during EC:

Anode:

$$Al(s) \rightarrow Al^{3+}(aq) + 3e^{-}$$
(1)

Cathode:

$$3H_2O + 3e \rightarrow 3/2H_2(g) + 3OH^-$$
 (2)

Overall:

$$Al^{3+}(aq) + 3H_2O \rightarrow Al(OH)_3 + 3H^+(aq)$$
(3)

Iron electrode reactions during EC:

Anode:

$$Fe(s) \rightarrow Fe^{2+}(aq) + 2e^{-} \tag{4}$$

Cathode:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^-$$
(5)

Overall:

$$Fe(s) + 2H_2O \rightarrow Fe(OH)_2 + H_2 \tag{6}$$

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

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

2 Experimental

2.1 Wastewater sampling and characteristics

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

2.2 Experimental assembly of EC reactor

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

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

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

2.3 Characterisation of wastewater samples

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

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

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

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

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

$$Cx = \frac{Mtotal, x}{Vtotal, liquid} = \frac{I \times t \times mx \times N}{Q \times t} = \frac{I \times mx \times N}{Q}$$
(1)

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

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

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

$$Operating \cos t = energy \cos t + material \cos t$$
(2)

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

Where V is the voltage applied on EC reactor, I is the current applied in Amperes on the EC reactor and Q is the flow of the influent through the EC reactor in m^3/h . The nergy consumption is expressed in Wh/m³.

3. Results and discussion

3.1 Treatment from primary sedimentation tank

3.1.1 COD removal efficiency of Fe and Al electrodes

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

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

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

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

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

3.2 Treatment from secondary sedimentation tank

3.2.1 Effect of current density on AFS in Fe coagulation

When Fe electrodes were used and wastewater from secondary sedimentation tank was introduced in the EC reactor, the current was applied in the range of 1–3 A and current density was changed from 0.67 to 2.02 A/m². Different operating conditions were opted for secondary sedimentation tank effluent analyses because of their changed characteristics. Higher current densities were applied for primary sedimentation tank analysis in comparison with lower values of current densities for secondary sedimentation tank analysis. This is because of higher amount of contaminants were detected in the primary sedimentation tank, while very little values were observed in secondary tank effluents (Table 1). According to AFS amounts found in the inflow and outflows after the EC reactor. It can be seen in Fig. 7 that the amount of AFS produced is directly proportional to the amount of current applied. Furthermore, higher amounts of outflows in comparison to inflows indicated that a large amount of electrode dissolution happened during the course of the EC process. In addition, it showed that dissolution of Fe electrodes is the primary reaction at anodes during typical current densities when pH approaches neutral values [23, 29]. Each experiment was run thrice to check the reproducibility of the results and named as WW-Fe-01, WW-Fe-02 and so on. Similar was the case with Al coagulation. In Fig. 8 coagulation dose of Fe, theoretical amount of Fe and AFS in outflows are compared against different current densities. The measured amounts of Fe in outflow is based on reaction (6), where one mole of Fe (s) reacts with two moles of water to produce one mole of Fe(OH)₂. It was assumed that first, all iron electrodes produce $Fe(OH)_2$ precipitates which were dirty green in colour and their amounts were then tested by Hach Lange test cuvettes, to prove the fact that the whole amount of Fe that produced as coagulation dose by Fe electrodes is in the form of Fe(OH)₂.

According to the results, current density has a direct effect on the dissolution rate in the studied range $(0.67-2.02 \text{ A/m}^2 \text{ or } 7.19-21.58 \text{ C/L})$ (Fig. 9). Iron dissolved at the rate of 2.93 mg Fe/C, while the theoretical value for dissolution of Fe (II) is 3.09 mg Fe/C. Therefore, it can be concluded that iron dissolved in Fe (II) form and dissolution followed Faraday's law. This was consistent with the results of other researchers [23-25]. These results are very important to establish the fact that Fe (II) is a poor coagulant and should be oxidized to Fe (III) form before it is employed to remove organic matter [1].

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

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

3.2.2 Effect of current density on AFS in Al coagulation

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

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

According to results, the current passing through the EC reactor was directly proportional to the dissolution rate [10] and followed Faraday's law. In the studied range ($0.34-0.67 \text{ A/m}^2$ or 2.45-4.82 C/l), Fig. 13 shows dissolving rate of Al measured was 1.27-2.39 mgAl/C, whereas the

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

3.2.3 Outflows pH

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

3.2.4 Flocs production analyses

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

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

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

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

3.3 Cost analysis of optimum current densities

The operating cost for the current EC process is calculated with Eq. (2). The energy consumption is then expressed in Wh/m³. Furthermore, the energy consumption costs for Fe and Al electrodes at 2 A and 1 A operation are found to be 0.03 €/m^3 and 0.02 €/m^3 respectively. In addition, it is observed that energy consumption is a direct relation with applied current, greater the applied current the higher will be the consumption of energy [5]. Based on the measured coagulant doses of Fe and Al electrodes as described in the previous section, the amounts of Fe and Al electrodes 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 on these optimum conditions, the used metal plates in the EC reactor can last for 86 and 99 days for Fe and Al electrodes with its full capacity of operation at 2A and 1A respectively. Moreover, the material cost of Fe, Al operating plant at 2 A, 1 A was calculated to be 0.01 € /m³ and 0.02 € /m³ respectively. According to reaction (5), the operating cost of EC operation for Fe and Al electrodes at mentioned current values of 2 A and 1 A is calculated to be 0.04 € /m³ and 0.03 € /m³ respectively.

4. Conclusions

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

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

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Variable	Wastewater from primary sedimentation tank	Wastewater from secondary sedimentation tank
рН	7.30	7.30
Conductivity ($\mu S/cm$)	1350	850-1100
Suspended solids (mg/L)	100–150	1.4–6.6
$\operatorname{COD}\left(mg/L\right)$	233.2	20
Total Phosphorous (<i>mg/L</i>)	5	0.45
Total Nitrogen (mg/L)	40.9	11.5

 Table 1. Wastewater characteristics.

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.