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Sustainable wastewater treatment using electrocoagulation process coupled with powdered activated carbon

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Abstract

In this research an electrochemical technique in combination with powdered activated carbon (PAC) for the removal of micropollutants by adsorption as an advanced stage purification step from effluents of pilot plant wastewater treatment plants (WWTP). The effluents of sedimentation tank comprised of wastewater plus PAC (WWPAC). The pilot plant mainly consists of two parts; the first one consists of electrocoagulation (EC) reactor and the second consists of electrophoretic deposition (EPD) discs and electroflotation (EF) setup. The electrocoagulation (EC) reactor is a fiber box consisting of two chambers and thirty four plates of one material (either Fe or Al) on the whole in one EC reactor while one cell has seventeen plates. Both types of electrodes have been tested with the outflow of sedimentation tank. The outflow from the sedimentation tank has been

entered into the EC reactor for the determination of EC reactor efficacy for the successful accomplishment of EC process at the designed pilot plant for WW treatment. The effect of different operational parameters; PAC dosage (20 mg), electrode nature (Fe and Al), current density ($0.34\text{--}2.02\text{ A/m}^2$) has been studied to find out the optimum conditions. Sludge volume index (SVI) of the sludge, thermogravimetric (TG), differential thermal analyses (DTA) and particle size distribution (PSD) of the flocs generated after the EC process has also been studied. The turbidity, pH and conductivity of effluents before and after EC treatment has also been carried out. This pilot plant research gave promising results for future work in advance wastewater treatment direction.

Keywords: Environmental pollution; Advanced wastewater treatment; powdered activated carbon; Micropollutants; Electrocoagulation and Suspended solids.

1 Introduction

The presence of organic pollutants in aquatic environment with the potential to cast adverse biological impacts, have been recognized for many years. In recent years, the authorities dealing with water has become more worried about the presence of harmful substances in water resources used for various purposes. The technical difficulties for inhibiting the discharge of micro-pollutants into the aquatic environment rise from inadequate removal in wastewater treatment plants (WWTPs) and the subsurface leakage of sewers. However, the other sources like agriculture, pharmaceutical, food additives, construction materials, cosmetics ingredients and naturally occurring substances such as hormones are also the major sources of pollutants in water bodies. The municipal WWTPs are usually designed to remove degradable organic compounds, solids and some nutrients.

Many treatments like filtration, adsorption, aerobic degradation, froth floatation, flocculation and electrocoagulation (EC) have been known till now for the removal of micropollutants and anthropogenic pollutants (Eryuruk, Un, & Ogutveren, 2018). While the EC is the most studied technique for the said purpose in WW treatment process (Sher, Hanif, Iqbal, & Imran, 2020). It is a multistep process being run by electric current for the agglomeration of micropollutants by using electrodes (coagulant) through redox reaction (Izadi, Hosseini, Darzi, Bidhendi, & Shariati, 2018). Some inorganic chemical like $(\text{NH}_4)_2\text{SO}_4$ and FeCl_3 were used as coagulants to carry out the conventional electrocoagulation processes. Powder activated carbon (PAC) is a finely grounded activated carbon which has promising effects due to its unique properties such as high surface area. It is mixed with the wastewater so that the effluent ingredients can be adsorbed on the surface of the fine activated carbon particles. Since both the natural organic matter and the micropollutants adsorb onto the activated carbon, the PAC process is best located after extensive biological treatment. When loaded with organic matter and micropollutants, the PAC has to be separated from cleaned wastewater and should be disposed of (Bazrafshan, Alipour, & Mahvi, 2016). There are numerous separation processes used for this purpose e.g. lamella separator, micro sieve, Sedimentation followed by cloth filtration, sand filtration and ultrafiltration (membrane) etc. Usually, the sedimentation of the PAC treated wastewater is possible only by the addition of coagulants and flocculating agents such as metal salts (FeCl_3 , AlCl_3 etc.) and organic polymers (Bazrafshan et al., 2016). Solid particles suspended or dissolved in water or wastewater tends to be charged negatively, so colloidal particles tend to repel each other, thus preventing the process of aggregation and settlement of these particles. It is very difficult to remove these particles from wastewater because of their small size to pass through the filter paper and they do not settle down under the action of gravity. For the effective removal of micropollutants, the colloids need to be

aggregated to increase in size to stay on the filtration media. Precipitation of a soluble product occurs when antisolvent is added to reduce the solubility of desired compound. So that precipitates can be easily separated by different physical treatments. The use of coagulants is done to bring about particle's agglomeration (coagulation). Coagulants are selected depending upon the electric charge on particles and the nature of the particles such as their affinity to adsorb water.

The EC process has been used for the removal of wastewater pollutants as it is an electrochemical process, proceeds by the passage of electric current from the metal electrodes (coagulants) (Hashim et al., 2019). This process consists of the transformation of water retaining hazardous pollutants to the non-hazardous materials by redox reactions. Due to the simple assembly, selective capability, short reaction period, easiness of handling, safety of the process, less use of chemicals and good purification efficiency, EC has been recognized as a famous green technology (Hashim et al., 2019; Papadopoulos et al., 2019). To carry out the redox reaction, necessary force (electromotive) is supplied by the electric current (I) which convert the soluble contaminants to the stable state that are comparatively less soluble, less colloidal and less emulsifiable than the previous contaminants at equilibrium values. By this process the soluble contaminants are converted into hydrophobic compound or precipitates which are easily separated out by the use of various separation techniques. By the use of EC method, no secondary contaminants form because of the use of very less amount of chemicals so there is no need of neutralization reaction which is the common advantage of this process (Ensano et al., 2019).

The EC technology comprised of three purification stages such as; (1) generation of coagulants by the electrolytic oxidation of sample wastewater; (2) destabilization of contaminants, deterioration emulsification and particle suspension; (3) for the generation of floc, the agglomeration of resultant particles consisting of colloids entangled in sludge layer formed after coagulation reaction

(Elazzouzi, El Kasmi, Haboubi, & Elyoubi, 2018). These flocs having similarity with chemical flocs, therefore they can be easily separated out by the sand filtration process due to their larger size, stability, containing less amount of water and acid resistance (Omwene, Kobya, & Can, 2018; Tanyol, Ogedey, & Oguz, 2018). A number of electrochemical reactions are involved in this process which occur at anode and cathode in the sequence as mentioned in equations (1–7). The electro-floatation process occurs during EC process which involves evolution of H₂ gas at cathode in the form of bubbles which discards particles by flotation (Devlin et al., 2019).

These chemical reactions are carried out to generate metallic electrodes' (anode: Fe or Al) hydroxides and coagulants for the destruction, coagulation and adsorption of pollutants for wastewater (Tanyol et al., 2018). From the reaction 1 and 4 it is obvious that the metal cations (Fe²⁺ and Al³⁺) get dissolved from the relevant anode to the aqueous phase of the reactor. These metal ions can further form various monomeric and polymeric hydroxides during electrochemical process to each electrode. The conversion of metallic cations into Al(OH)₃ and Fe(OH)₂ hydroxides in (reaction 3 and 6) are pH dependent reactions. Therefore, the final species formed act as adsorbents for micro-pollutants.

The reactions during EC process at aluminum electrode are as:

Anode:

Cathode:

Overall:

Reactions at iron electrode during EC are:

123 Anode:

125 Cathode:

127 Overall:

129

130 In literature, purification of colour, ammonia, chemical oxygen demand (COD) and phosphorus
131 by using various electrodes such as Mg (Devlin et al., 2019), Fe (Chou, Wang, & Chang, 2009),
132 Al (Hashim et al., 2019) and steel (Mahmad, Rozainy, Abustan, & Baharun, 2016) have been
133 evaluated at different pH and high current values, also PAC has been used for this purpose along
134 with all these electrodes (Bonvin, Jost, Randin, Bonvin, & Kohn, 2016; Mailler et al., 2015). This
135 study is performed at pilot plant scale with electrocoagulation process in combination with PAC
136 using different current densities and coagulants. The objectives are focused to study the role of
137 current densities, amount of PAC and coagulant's nature in the production of flocs and EC process
138 at this pilot plant and the evaluation of optimised conditions for this process. All experiments were
139 conducted individually using Fe and Al coagulants/electrodes and then amount of suspended solids
140 (SS) is also calculated. Furthermore, the technique of membrane filtration has also been used for
141 the determination of SS amount in the wastewater.

142 **2 Experimental setup**

143 **2.1 Wastewater sampling**

144 Wastewater sample from the wastewater treatment plant was taken for the experiments. The
145 wastewater characteristics from secondary sedimentation tank are elaborated in Table 1. After

maintaining constant operating conditions of EC setup, the inflow and outflow points were fixed for obtaining homogenous samples every time. To make sure the constant homogeneous sample's composition, samples were taken at short intervals of time instead taking all sample at once.

2.2 Powdered activated carbon (PAC) dosing

In two ways, activated carbon can be used; powder form of granular form. PAC usually has a size less than 0.074 mm diameter (200 mesh). The bulk density of PAC is ranging from 360–740 kg/m³. The total surface area of PAC is 800–1800 m²/g. The typical characteristics of PAC are given in Table 2. The 20 mg/L dosage of PAC was used for each run of electrocoagulation process with Fe and Al. PAC slurry is prepared in a 200 L closed tank. The amount which has to be added in the mixing tank in order to achieve 20 mg/L concentration in the outflow of mixing tank was calculated to be about 2 g/L in the PAC slurry. The wastewater stream coming to the mixing tank was adjusted accordingly at about 1000 L/h while the PAC slurry rate is adjusted at about 10 L/h with the help of a small pump for an open mixing tank of volume 150 L. The PAC added to the PAC slurry tank was used from a storage tanker placed outside the lab at ambient temperature.

2.3 Pilot plant setup

The experimental setup of EC plant is represented in **Fig. 1**. A feed/ mixing open tank is used for this experiment. Some contaminants settle down during the experiment like suspended solids (SS), so for the maintenance of consistent inflow rate and continuous stirring, a stirrer is fitted in the feeding tank (150 L). PAC slurry is made in a 200 L closed tank, also equipped with a mechanical stirrer for a homogeneous outflow concentration of the PAC slurry to the feed tank. PAC Slurry tank is connected to the feed tank/mixing tank through a small pump (range 8–12 L/h) operating at about 10 L/h and pumping in PAC at about 2 g/L in the feed tank. A pump (250–1500 L/h) is used to feed effluent to the EC reactor from the feed tank and for maintaining 500 L/h inflow to EC

reactor, a flow meter is attached in line. The pilot plant setup comprises of two main sections: first part has an EC reactor while the second part has EF setup and EPD discs as shown in **Fig. 1**.

The EC reactor is made up of fibre box, consists of two chamber attached in series having slits inside for the insertion of metal plates performing the function of anodes and cathodes. There are total thirty four plates of one material (Fe or Al) in the whole reactor from which 17 plates are present in each cell. For the better operation of system, the height of plates is increased to about 3 cm and 3 mm in length and diameter respectively by attaching slits inside the EC cells by using plastic cylindrical objects in EC reactor. Furthermore, the plates are connected to DC power supply via polarity changer circuit. In this connection, plates play the role of bipolar electrodes in parallel circuit. Moreover, the polarity changer circuit has the control to change the polarity of electrodes at various time intervals and hence equally consuming all the electrodes. The electrode's side facing anode is negatively charged and vice versa on the opposite side of metal electrode. To enter the inflow into the reactor, the EC reactor is provided with a circular inlet hole where connecting pipes are attached.

A rectangular opening at reactor's outlet, a circular hole at the bottom of reactor is fitted for the overflow of water and an auxiliary glass lid is used for EC reactor covering to ascertain a better inside view of the process and for the control of parameters by visually observing the procedure. The electrode plates inside the reactor constitute an effective surface area of about 1.49 m^2 with the dimensions of $300 \times 155 \times 3 \text{ mm}$. Different values of current densities were selected for Al and Fe electrodes in this experiment such as $0.67\text{--}1.34 \text{ A/m}^2$ and $0.34\text{--}0.67 \text{ A/m}^2$ ranges current densities were used for Fe and Al electrodes respectively. The results showed that the Al electrode provided more agglomeration of suspended solids at the lower densities of current as compare to

Fe electrode which required higher densities of current for noticeable results. The Al electrode was selected for this treatment as observed previously, due to the requirement of lower densities of current and economic factor. The electrocoagulation inflow rate was kept at 500 L/h. The solutions of NaOH and H₂SO₄ were used for the maintenance of inflow pH on requirement. The settings of the EC reactor for Fe and Al derived coagulation of WWPAC is inserted in Table 3.

2.4 Membrane filtration technique

The membrane separation process was used for the calculation of suspended solids in the sample by following the protocol of DIN 19643-2 (Dudziak, Wyczarska-Kokot, Łaskawiec, & Stolarczyk, 2019). After treatment, they called as after filtration solids (AFS). The filtration was carried by using the filter papers which allowed only those particles to pass on having the diameter of <0.45 µm at the 5 bars specific nitrogen pressure followed by drying the wet filter papers in oven at 105 °C for about 1 hour which evaporated all moisture content and left only solid residues on the paper. These solid particles were kept in desiccator for cooling down for 30 minutes. Finally, the weight of filter papers was measured for the calculation of total suspended solids in the treated wastewater. Furthermore, each sample was taken to AFS setup three times for taking the average experimental calculated values. On the basis of coagulant dissolution property and floc formation ability, the quantitative analysis was performed by the comparison of AFS of inflow and outflows of samples. For the observation of the reliability of the treatment, the amount of coagulants was measured experimentally and theoretically which was added during the experiment for flocs generation and this was done after AFS calculation. The experimental calculation of optimum dosing amount of Al and Fe coagulants in outflows is the major goal of this study. Spectrophotometric analysis was done for the calculation of total Fe amount in outflow and gravimetric analysis was performed for

the measurement of Al amount in outflow (Baierle et al., 2015). While theoretical measurement of Fe and Al coagulants' doses is done by using Eq. (7) (Heidmann & Calmano, 2010).

$$C_x = \frac{m_{total,x}}{V_{total,liquid}} = \frac{I \times t \times m_x \times N}{Q \times t} = \frac{I \times m_x \times N}{Q} \quad (7)$$

where C_x (mg/L) represents the released metal amount (Fe or Al) after the treatment from the electrolysis cell; $m_{total,x}$ (mg) is the mass of metal released after process by Faraday's law in retention time; $V_{total,liquid}$ (L) shows the total effluent's volume within the range of retention time; I (A) is the current applied; N shows the channels' number; Q (L/s) is the rate of flow and m_x represents the electrochemical equivalent for metals. For Al and Fe metals, electrochemical equivalent values are (0.093 mg/C) and (0.193 mg/C) respectively.

To characterise the sludge development, sludge volume index (SVI) that is the volume (mL) occupied by 1 gram of sludge after settling in 1 L amount of treated liquor for 30 min was measured. The SVI can be calculated by using Eq. (8) (Papadopoulos et al., 2019).

$$SVI = (\text{Settled volume of sludge} \times 1000) / \text{MLSS} \quad (8)$$

where MLSS is the mixed liquid suspended solids and used for the calculation of SVI.

2.5 Flocs characterisation

The change in physical or chemical properties of materials/ flocs after EC process was carried out by thermal gravimetric analyser (TGA) which consists of a sample pan attached with a precision balance. The pan is kept inside the furnace at constant heating during the experiment. Moreover, the mass of sample is examined during the course of experiment. A sample of pure gas controls the sample environment. However, the nature of gas can be inert or reactive that can flow over the sample or eliminates through an exhaust. The equipment used for thermal gravimetric analysis

was Netsch STA 409 C developed by Netsch Gerätebau GmbH. It combines differential thermal analysis (DTA) and TGA, so allowing simultaneous measurement of thermal properties and mass change (Peng et al., 2016).

A list of values of mathematical function defining the relative amount by mass according to the size of particles dispersed in liquid in the form of powder or granular material is known as the particle size distribution (PSD). PSD characterization is done of the effluent after EC treatment with metal electrodes (Fe and Al). Further to observe the flocs formed after the EC process in detail, 1 litre of sample is separated during the experiments every time. The samples were separately taken to the electrodes (Fe and Al) with their respective current densities. The laser granulometer, the Mastersizer 3000 from Malvern is used to perform PSD (Lee & Gagnon, 2016). This device can be used to measure the particles' size of 0.01 to 3500 microns range. Two sources of light were used for measurement; red and blue lights of the wavelengths of 632.8 nm and 470 nm respectively.

2.6 Measurement of pH and turbidity

Turbidity was measured using an electronic turbidity meter. A pH meter an electronic device was used for the measurement of pH of the samples before and after EC process. A pH meter usually consists of two parts: a measuring glass probe and an electronic meter for the measurement of pH which is displayed in digits.

3 Results and discussion

It is an established fact that concentration of 10–20 mg PAC per litre of wastewater and average retention time of 15–30 minutes can remove efficiently the micropollutants from the wastewater (Kårelid, Larsson, & Björleinius, 2017). The sedimentation of colloidal micropollutants with the

PAC is possible by the addition of coagulants and flocculants (FeCl_3 , AlCl_3 etc.) and organic polymers. Because of the small particle size in combination with the high flow rates, the separation of PAC loaded with micropollutants and suspended solids is a difficult task. Besides the cost of these chemical coagulants, they have a negative effect such as they lower the adsorption capacity of PAC by blocking surface pores. Electrocoagulation process can remove the side reaction of lowering adsorption capacity of PAC and also have several other operational advantages in wastewater treatment. In EC electrochemical process, the amount of current density plays a vital role. Further, different values of current densities were used at Fe and Al electrodes to test the role of current density on coagulation process of solid particles at the flow rate of 500 L/h.

3.1 EC process with Fe coagulation

After entering the wastewater from sedimentation tank to the EC reactor having Fe electrodes, PAC was mixed in the amount of 20 mg/L to the inflow water at the current range of 1–2 A and current density of 0.67–1.34 A/m^2 . The **Fig. 2** shows that the AFS production is dependent on the amount of applied current and it can also be concluded from the AFS amount established in inflow and outflow of EC reactor. The indication of dissolution of metal electrode is obtained by higher amounts of outflows than in the inflows during the EC process. Furthermore, it is can be assumed that Fe electrode dissolution is the main reaction which occurs at anodes during specific amount of current densities when pH become neutral (Sasson, Calmano, & Adin, 2009).

In the **Fig. 3**, theoretically calculated amounts of Fe coagulant, Fe and AFS obtained from outflows were compared with the current densities values taking it as a variable. Reaction (6) shows the amount of Fe obtained at outflow of EC reactor in which by the reaction of 1 mole of Fe (s) and 2 moles of water produces 1 mole of $\text{Fe}(\text{OH})_2$. It was supposed that complete amount of metal in Fe electrode was consumed for the production of $\text{Fe}(\text{OH})_2$ precipitates with dirty green appearance.

After adding PAC amounts, new flock were formed causing the color change from dirty green precipitates of Fe(OH)₂ to dark black.

A new set of experiments were designed to know and verify the fact about the produced Fe(II) and consequently Fe(OH)₂. When the outflow containing Fe(OH)₂ was treated with enough amount of 25% 1M HCl, the new product formed Fe(Cl)₂ was fully soluble in water and made a clear solution, which is then subjected to AFS again. The reaction took place while treating the outflows produced by Fe electrodes with HCl is shown in reaction (9).

As shown in **Fig 4**, the red shaded area in the bars represents the amount of AFS after treating with HCl and the normal height of the bars (green shaded) represents the amount of AFS produced without treating with HCl. Furthermore, it can be clearly seen that the amounts of AFS produced after treating with HCl are approximately the same amount as it is in the inflow stream before the EC process as shown in **Fig. 2**. This fact proved the calculations for the Fe amount in the outflow streams based on the fact that the whole amount of produced precipitates are of Fe(OH)₂. That on the treatment with HCl becomes soluble in solution while AFS of HCl treated outflow gave the amount which have to be introduced into the inflow to EC process for this specific set of experiments.

3.2 EC process with Al coagulation

When wastewater was entered into EC reactor (with Al electrodes) from the sedimentation tank, PAC was mixed in amount of 20 mg/L into the inflow with the current and current density in the

range of 0.5–1 A and 0.34–0.67 A/m² respectively. The AFS amount was measured before and after the EC process present in the inflow and outflow and represented in **Fig. 5** which also shows that the AFS amount production is directly proportional to the applied current. The dissolution of metal electrodes is the basic reaction occurring at anodes, here the same case is with Fe electrodes.

The amount of Al coagulant dose, theoretically calculated amount of Al and AFS produced with the corresponding current density range are shown in **Fig. 6**. The measurement of amount of Al is shown by the reactions (1–3) in which 1 mole of Al and 3 moles of water reacts to form Al(OH)₃ which polymerized to form hydroxide compounds (Izadi et al., 2018). The assumption is made that whole Al electrode consume up to produce Al(OH)₃ and this assumption is proved experimentally by the reaction with HCl.

In this experiment as well, the outflows obtained after treating with EC based on 0.5 A, 0.75 A and 1 A current settings, were treated with HCl to verify the fact that the whole amount which was produced by Al electrodes during EC process was Al(OH)₃. When the outflows containing Al(OH)₃ was treated with enough amount of 25% 1 M HCl, the new product formed AlCl₃ which was fully soluble in water and made a clear solution, which is then subjected to AFS again. The reaction took place while treating the outflows produced by Al electrodes with HCl is shown in reaction (10).

As shown in the **Fig. 7**, the red shaded area in the bars represents the amount of AFS after treating with HCl and the normal height of the bars (blue shaded) represents the amount of AFS produced

without treating with HCl. Moreover, it can be clearly noticed that the produced amount of AFS after treating with HCl are approximately the same amount as was introduced in the inflow stream before the EC process as shown in **Fig. 5**. This fact proved the calculations for the Al amounts in the outflow streams based on the fact that the whole amount of produced precipitates are of Al(OH)_3 , which on treatment with HCl become soluble in solution and the AFS of the HCl treated outflow gives us the amount which have to be introduced in the inflow to EC Process for this specific set of experiments.

3.3 Settling characteristics of sludge

The **Fig. 8** (a) and (b) show the settled volume of sludge after two hours of settling, for Fe and Al electrodes at different applied current settings for the EC operation with wastewater and PAC. The sludge volume index of the sludge produced by Fe and Al electrodes according to Eq. (9) while operating with wastewater and PAC is measured at various current amounts. The Sludge volume index (SVI) of sludge produced by Al and Fe electrodes operation with wastewater is incorporated in Table 4. The SVI was used by R. Katal et al., (Katal & Pahlavanzadeh, 2011) for the settling characteristics of EC sludge produced by Al and Fe electrodes.

According to Zodi et al., textile wastewater treatment by EC process. The SVI values generated by Fe and Al EC process found to be varies with current density are in the range of 0–215 mL/g at 50–200 A/m² (Zodi, Potier, Lapique, & Leclerc, 2009). This research is designed at pilot plant scale, and the EC process was run in combination with PAC dosage, the obtained results of SVI with Fe and Al EC process with WWPAC effluents were very interesting. To some extent similar results of SVI were obtained but at lower values of current densities. The SVI found by the designed pilot plant operations is 23–208 mL/g and 58–312 mL/g for Al and Fe electrodes

respectively. However, the sludge produced by iron was denser and in the form of a compact layer, while the sludge produced by Al electrodes was observed to be fluffy and light weight.

3.4 Outflow effluent analysis for pH and turbidity

Other factor such as pH also plays a vital part in the reactions of EC reactor in addition to the effect of current applied. The effect of pH is also confirmed by the literature which states that pH of wastewater changes after passing through the EC reactor due to the cathode reactions (H_2 evolution). It also depends on the selection of anode and initial pH of inflow water (Eryuruk et al., 2018; Izadi et al., 2018; Yavuz & Ögütveren, 2018). The increase in AFS amount in the outflow was observed by the increase of current density in the previous section. The effect of pH was observed on the inflow and outflow of the EC reactor and on the production of AFS at the applied conditions of current density for both type of electrodes. The outflow's pH was increased correspondently with the increase in current density in all the experiments (Izadi et al., 2018). The outflow had a slight increase in pH with both electrodes (Fe and Al) in both series of all the experiments.

The pH of the reaction solution changes during the course of EC process and this is a well-known side reaction of the process. The pH increase was observed during the whole set of experiments performed in two series with Fe and Al electrodes. However, the mechanism of EC is highly dependent on the conductivity of the aqueous medium. In the whole set of experiments done with metal electrodes (Al and Fe), the conductivity remained almost similar before and after the EC process. Therefore, it can be mentioned that the conductivity remained constant over the EC process. It can be observed that the turbidity of the effluents is increased after EC process. The turbidity of the sample increases after the EC process because of the hydroxide formation. No

matter which metal has been used, the turbidity varies directly as the current density increases. However, in this study the turbidity generated with Al electrodes is higher than Fe electrodes and a sharp rise is observed in turbidity values at same current **Fig. 9** (Sasson et al., 2009).

3.5 Flocs production investigation

3.5.1 Thermogravimetric analysis

Thermogravimetric analysis (TGA) was made to understand the thermal behaviour of the flocs produced from EC process. Thermogravimetric (TG) and differential thermal analysis (DTA) plots for iron and aluminum flocs are shown in **Fig. 10**. The DTA signal which is represented the amount of released energy and that also corresponds to the maximum amount at approximately about 550 °C and hence confirming the exothermic reaction nature of the tested substances. TGA and DTA analysis for iron and aluminium flocs were done. Observing these plots, there are clearly four zones visible that represent the mass loss of the flocs while exhibiting the analysis. In the first zone, which corresponds to the 25–150 °C, the usual mass loss happened for both the **Fig. 10 (a)** and **(b)** flocs (Sahu, Mazumdar, & Chaudhari, 2019).

The first zone here corresponds to the mass loss on the basis of moisture removal or drying of the matter. In the second zone, where the temperature corresponds to 150–350 °C and the mass loss can be assigned as oxidation, decomposition or transformation of the easily degradable substances. In the third zone, the behaviour of the Fe and Al flocs is quite different, depending upon the corresponding products formed during the EC process. The mass loss in case of Fe flocs corresponding to temperature range of 300–430 °C is mainly related to the decomposition of Fe(OH)_3 , which is a highly exothermic reaction according to DTA signals generated. On the other hand, the mass loss in case of Al flocs corresponding to temperature range of 300–470 °C is mainly

related to the decomposition of $\text{Al}(\text{OH})_3$ and the energy release was even higher as compared to the Fe flocs. The more mass loss in case of the Al flocs indicates that there are more easily degradable salts of Al present in the Al flocs. In the zone of rest matter or residual non-degradable substances, the Fe flocs tend to have more solid metal residues than Al flocs.

The amount of rest matter for Fe flocs is more than that of Al flocs, showing that the amount of coagulation dose was also higher in case of Fe flocs, as the sample was taken at about 2A and the lesser amount of residues in the Al flocs is also justified on the basis that the lesser current was applied to take the sample, corresponding to 1A. As a conclusion for the TG and DTA graphs, at optimum conditions defined for the PAC series, which are 2 A for the Fe flocs and 1 A for the Al flocs, both types of flocs exhibits the same sort of behaviour in the initial phases, but in the main degradation phase of the coagulant salts formed during the course of the process, Al flocs exhibit more mass loss in comparison to the Fe flocs and Fe flocs are found to be giving more residual matter in the end in contrast to Al flocs.

3.5.2 Particle size distribution

For PSD analysis by using laser granulometer, the samples were taken from outflow streams after EC reactor process having PAC, Fe and Al electrodes. The results are shown in **Fig. 11**, when Fe and Al were used for the purpose of coagulation at the current values of 0.5–2 A. In PSD analysis, the cumulative volume (%) is shown against the diameter (μm) of particles. **Fig. 11** confirms that the bigger flocs were produced with the higher amount of current application which could be easily separated by secondary sedimentation processes. The reason for the bigger size of flocs with operation of wastewater and PAC by Al electrodes can be the result of the special structure of Al complexes formed during the EC process.

4 Conclusions

The design of electrocoagulation (EC) technique is revived to fulfil the needs of this method for the removal of micropollutants from wastewater. Moreover, an advanced research of PAC is used in combination with EC. By changing the amount of current applied, the selected process is tested for obtaining the improved quality water from the outflow of EC pilot scale reactor. The results are compared for a good downstream separation method. On the basis of the Faraday's Law and the amount of AFS produced, the optimum doses of Fe and Al coagulants are calculated. The dosing amount of coagulants increased by increasing the current density with Fe and Al metal electrodes. The best results obtained for Al and Fe metal electrodes for variable range of current and current densities are 1 A (0.67 A/m²) and 2 A (1.34 A/m²) respectively for the highest amount of AFS production. Various other analyses of pH, turbidity, conductivity, TGA, DTA, SVI, PSD have been performed to deeply study the whole process, before and after the reaction. From all analyses mentioned before, better results with lower values of current density were obtained by Al electrodes. Furthermore, all these experiments are performed on the designed EC pilot plant and gained remarkable results which are a promising opportunity for other researchers to use similar electrodes system and natural coagulant material at this test plant in future. Additionally, this pilot plant research could have future work in advance wastewater treatment such as using other electrodes system and natural coagulant materials.

Declarations

Ethical Approval

Not applicable

Consent to Participate

Not applicable

Consent to Publish

Not applicable

Authors Contributions

Farooq Sher; Tahir Rasheed: Conceptualization, Methodology, Software Farooq Sher; Sania Zafar Iqbal.: Data curation, Writing- Original draft preparation. Tahir Rasheed: Visualization, Investigation. *Farooq Sher*: Supervision.: Kashif Hanif; Fatima Zafar: Software, Validation.: Farooq Sher; Tahir Rasheed; Jasmina Sulejmanović; Eder C. Lima: Writing- Reviewing and Editing,

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Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Availability of data and materials

Not applicable

References

- Baierle, F., John, D. K., Souza, M. P., Bjerk, T. R., Moraes, M. S., Hoeltz, M., . . . Schneider, R. C. (2015). Biomass from microalgae separation by electroflotation with iron and aluminum spiral electrodes. *Chemical Engineering Journal*, 267, 274-281.
- Bazrafshan, E., Alipour, M. R., & Mahvi, A. H. (2016). Textile wastewater treatment by application of combined chemical coagulation, electrocoagulation, and adsorption processes. *Desalination and Water Treatment*, 57(20), 9203-9215.
- Bonvin, F., Jost, L., Randin, L., Bonvin, E., & Kohn, T. (2016). Super-fine powdered activated carbon (SPAC) for efficient removal of micropollutants from wastewater treatment plant effluent. *Water research*, 90, 90-99.

- Chou, W.-L., Wang, C.-T., & Chang, S.-Y. (2009). Study of COD and turbidity removal from real oxide-CMP wastewater by iron electrocoagulation and the evaluation of specific energy consumption. *Journal of hazardous materials*, 168(2-3), 1200-1207.
- Devlin, T. R., Kowalski, M. S., Pagaduan, E., Zhang, X., Wei, V., & Oleszkiewicz, J. A. (2019). Electrocoagulation of wastewater using aluminum, iron, and magnesium electrodes. *Journal of hazardous materials*, 368, 862-868.
- Dudziak, M., Wyczarska-Kokot, J., Łaskawiec, E., & Stolarczyk, A. (2019). Application of Ultrafiltration in a Swimming Pool Water Treatment System. *Membranes*, 9(3), 44.
- Elazzouzi, M., El Kasmi, A., Haboubi, K., & Elyoubi, M. (2018). A novel electrocoagulation process using insulated edges of Al electrodes for enhancement of urban wastewater treatment: Techno-economic study. *Process Safety and Environmental Protection*, 116, 506-515.
- Ensano, B. M. B., Borea, L., Naddeo, V., Belgiorno, V., de Luna, M. D. G., Balakrishnan, M., & Ballesteros Jr, F. C. (2019). Applicability of the electrocoagulation process in treating real municipal wastewater containing pharmaceutical active compounds. *Journal of hazardous materials*, 361, 367-373.
- Eryuruk, K., Un, U. T., & Ogutveren, U. B. (2018). Electrochemical treatment of wastewaters from poultry slaughtering and processing by using iron electrodes. *Journal of cleaner production*, 172, 1089-1095.
- Hashim, K. S., Al Khaddar, R., Jasim, N., Shaw, A., Phipps, D., Kot, P., . . . Alawsh, R. (2019). Electrocoagulation as a green technology for phosphate removal from River water. *Separation and Purification Technology*, 210, 135-144.
- Heidmann, I., & Calmano, W. (2010). Removal of Ni, Cu and Cr from a galvanic wastewater in an electrocoagulation system with Fe-and Al-electrodes. *Separation and Purification Technology*, 71(3), 308-314.
- Izadi, A., Hosseini, M., Darzi, G. N., Bidhendi, G. N., & Shariati, F. P. (2018). Treatment of paper-recycling wastewater by electrocoagulation using aluminum and iron electrodes. *Journal of Environmental Health Science and Engineering*, 16(2), 257-264.
- Kårelid, V., Larsson, G., & Björleinius, B. (2017). Effects of recirculation in a three-tank pilot-scale system for pharmaceutical removal with powdered activated carbon. *Journal of environmental management*, 193, 163-171.
- Katal, R., & Pahlavanzadeh, H. (2011). Influence of different combinations of aluminum and iron electrode on electrocoagulation efficiency: Application to the treatment of paper mill wastewater. *Desalination*, 265(1-3), 199-205.
- Lee, S. Y., & Gagnon, G. A. (2016). Comparing the growth and structure of flocs from electrocoagulation and chemical coagulation. *Journal of Water Process Engineering*, 10, 20-29.
- Mahmad, M. K. N., Rozainy, M. M. R., Abustan, I., & Baharun, N. (2016). Electrocoagulation process by using aluminium and stainless steel electrodes to treat total chromium, colour and turbidity. *Procedia Chemistry*, 19, 681-686.
- Mailler, R., Gasperi, J., Coquet, Y., Deshayes, S., Zedek, S., Cren-Olivé, C., . . . Caupos, E. (2015). Study of a large scale powdered activated carbon pilot: Removals of a wide range of emerging and priority micropollutants from wastewater treatment plant effluents. *Water research*, 72, 315-330.

- Omwene, P. I., Kobya, M., & Can, O. T. (2018). Phosphorus removal from domestic wastewater in electrocoagulation reactor using aluminium and iron plate hybrid anodes. *Ecological engineering*, 123, 65-73.
- Papadopoulos, K. P., Argyriou, R., Economou, C. N., Charalampous, N., Dailianis, S., Tatoulis, T. I., . . . Vayenas, D. V. (2019). Treatment of printing ink wastewater using electrocoagulation. *Journal of environmental management*, 237, 442-448.
- Peng, C., Zhai, Y., Zhu, Y., Xu, B., Wang, T., Li, C., & Zeng, G. (2016). Production of char from sewage sludge employing hydrothermal carbonization: char properties, combustion behavior and thermal characteristics. *Fuel*, 176, 110-118.
- Sahu, O., Mazumdar, B., & Chaudhari, P. (2019). Electrochemical treatment of sugar industry wastewater: process optimization by response surface methodology. *International journal of environmental science and technology*, 16(3), 1527-1540.
- Sasson, M. B., Calmano, W., & Adin, A. (2009). Iron-oxidation processes in an electroflocculation (electrocoagulation) cell. *Journal of Hazardous Materials*, 171(1-3), 704-709.
- Sher, F., Hanif, K., Iqbal, S. Z., & Imran, M. (2020). Implications of advanced wastewater treatment: Electrocoagulation and electroflocculation of effluent discharged from a wastewater treatment plant. *Journal of Water Process Engineering*, 33, 101101.
- Tanyol, M., Ogedey, A., & Oguz, E. (2018). COD removal from leachate by electrocoagulation process: treatment with monopolar electrodes in parallel connection. *Water Science and Technology*, 77(1), 177-186.
- Yavuz, Y., & Ögütveren, Ü. (2018). Treatment of industrial estate wastewater by the application of electrocoagulation process using iron electrodes. *Journal of environmental management*, 207, 151-158.
- Zodi, S., Potier, O., Lapique, F., & Leclerc, J.-P. (2009). Treatment of the textile wastewaters by electrocoagulation: Effect of operating parameters on the sludge settling characteristics. *Separation and purification Technology*, 69(1), 29-36.

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Table 1. Characteristic of wastewater from secondary sedimentation tank.

Variable	Values
pH	7.03–7.21
Conductivity (µS/cm)	850–1100
Suspended solids (mg/L)	1.4–6.6
COD (mg/L)	20
Total Phosphorous (mg/L)	0.45
Total Nitrogen (mg/L)	11.5

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Table 2. Properties of powdered activated carbon.

Parameters	Activated carbon
Total surface area (m ² /g)	800–1800
Bulk density (kg/m ³)	360–740
Particle size range (mm)	5–50
Mean pore radius (Å)	20–40
Iodine number	800–1200
Abrasion number	70–80
Ash (%)	6
Moisture (%)	3–10

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Table 3. Settings of EC reactor via Fe and Al treatment.

Electrodes	Parameter settings		
	Current (A)	Voltage (V)	PAC (mg)
Fe	1	38	20
	2	58	20
Al	0.5	26	20
	0.75	37	20
	1	58	20

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550 **Table 4.** SVI of sludge produced by Al and Fe electrodes operation with wastewater.

Electrode	Current	Current density	MLSS	Settled volume of	SVI
type	(A)	(A/m²)	(mg/L)	sludge (mL/L)	(mL/g)
Al	0.5	0.34	22.2	0.5	23
	1	0.67	58	6	103
	2	1.34	86.7	18	208
Fe	1	0.67	22.4	1.3	58
	2	1.34	79.5	18	226
	3	2.02	131.4	41	312

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Table 5. Operational parameters of pH, turbidity and conductivity.

Sample	Setting	Current (A)	pH	Turbidity (NTU)	Conductivity ($\mu\text{s}/\text{cm}$)
Inflow	WWPAC-Fe	–	7.03	5	1019
Outflow	WWPAC-Fe	1	7.26	29	991
Inflow	WWPAC-Fe	–	7.03	5	1019
Outflow	WWPAC-Fe	2	7.36	35	1007
Inflow	WWPAC-Al	–	7.10	8	964
Outflow	WWPAC-Al	0.5	7.21	29	991
Inflow	WWPAC-Al	–	7.1	8	964
Outflow	WWPAC-Al	0.75	7.52	31	988
Inflow	WWPAC-Al	–	7.10	8	964
Outflow	WWPAC-Al	1	7.52	40	985

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