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Electrocoagulation for COD, turbidity, ammonia and phosphate removal from municipal wastewater

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The study illustrates, viability of using electrocoagulation as a process for COD, turbidity, ammonia and phosphate removal from real municipal wastewater (MWW). Experiments were performed in batch mode at laboratory scale with stainless steel as cathode and aluminium as anode. Constant voltage of 12 V was used throughout the experiments. Effects of inter-electrode distance and current were evaluated for removal of COD, turbidity, ammonia and phosphate from MWW. Effect of inter-electrode distance was tested for this, the spacing between electrodes was fixed at 3 cm, 5 cm and 7 cm. The highest COD removal efficiency of 89% was observed at 3 cm inter-electrode distance followed by 84% and 82% at 5 cm and 7 cm, respectively. Ammonia reduction was 55%, 38% and 32% at 3 cm, 5 cm and 7 cm inter-electrode distance, respectively. Phosphate removal was 99% at 3 cm and 98% both at 5 cm and 7 cm. Turbidity removal also followed the same trend, at 3 cm maximum removal of 95.5% was recorded and at 5 cm and 7 cm 94% and 93% removal occurred, respectively. Another operating parameter studied was the influence of current by applying 0.5 A, 1 A and 2 A. Maximum COD, turbidity, ammonia and phosphate removal occurred at 2 A. COD removal of 76%, turbidity removal of 94%, ammonia reduction of 72% and phosphate reduction of 98% was recorded at 2 A which was maximum for all these parameters. From the current study, it can be highlighted that electrocoagulation is suitable for COD, ammonia, phosphate and turbidity removal. Operating cost calculated was 0.12, 0.24 and 0.48 USD/m^3 for 0.5 A,1 A and 2 A, respectively. Further studies on continuous mode operation, characteristics of sludge formed, and additional treatment needs to be carried out.

Keywords: MWW, electrocoagulation, interelectrode distance, current, COD, turbidity.

Introduction

Half of the world's population has been affected by the water crisis, which is emerging as a global environmental issue[1]. In India, numerous districts are currently facing water crisis either due to quality or quantity. To overcome this issue, emphasis is given on wastewater treatment to avoid water contamination problems and reuse of municipal wastewater to reduce burden on conventional water resources[2]. The main constituents in municipal wastewater is organic and inorganic solids, floating materials, settable solids, oil, dissolved gases, and microorganisms. There are many ways to treat wastewater as biologically, chemically or with a combination of both. Chemical treatment although are reliable but often are energy intensive and also require regular supply of metal salts or other chemical mixtures along with generation of byproducts[3]. Electrochemical processes have recently gained wider attention due to advancement of science in this area and further research is essential to improve the overall understanding of these processes for achieving highly efficient water and wastewater treatment.

Electrocoagulation (EC) is an effective method for water and wastewater treatment[4]. Electrochemical methods are widely accepted because of requirements of lower coagulant dosage, less sludge generation, easy operation with simple equipment[5]. Electrocoagulation has been widely used for purification in different industries like textile dyeing, tannery, metal laden wastewater, restaurant wastewater and potable water[6]. EC process basically works on the coagulation, flotation and electrochemistry, all these three basic processes are the backbone of electrocoagulation[7]. There is *in situ* generation of coagulants by dissolution of sacrificial anodes due to direct current flow. Aluminium electrode has been

proved to be most effective and successful electrodes by different studies[8]. Because of this, aluminium has been used as the electrode material in this study. The anode produce metal cations after its oxidation process by applying direct current, this cation involve in further reactions and produce different polymeric hydroxides of metal, which is similar to salts used in conventional coagulation[9]. The reactions when using aluminium electrode can be summarized as follows[10].

Anode reactions:

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

$$Al^{3+}_{(aq)} + 3OH^- \rightarrow Al(OH)_3 \quad (2)$$

Alkaline pH range

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

Acidic pH range

Cathode reactions leads to mainly hydrogen evolution:

$$3H_2O + 3e \leftrightarrow {}^3/_2\ H_2 \uparrow + 3OH^- \quad (4)$$

when phosphate is present as impurities, it could be removed as per following reaction[11].

Aluminium as anode when oxidizes forms many polymeric species such as $Al_{13}O_4(OH)^{247+}$, $Al_{13}(OH)^{345+}$, $Al_8(OH)^{204+}$, $Al_7(OH)^{174+}$ and $Al_6(OH)^{153+}$, which are finally transformed into $Al(OH)_3$(s) as expressed in eq. (3). These products have a large surface area which is responsible for effective adsorption of pollutants and flocs formed are separated from solution by sedimentation or H_2 flotation. Amorphous $Al(OH)_3$ (s) can exhibit large surface area and forms "sweep flocs" which are useful for fast adsorption of soluble organic compounds and also for removing colloidal particles. Ultimately these flocs are removed easily from the aqueous medium by flotation and sedimentation induced by the hydrogen bubbles generated at the cathode which is referred as electro-flotation[12]. The basic mechanisms for removal through EC are due to oxidation, absorption, reduction, coagulation, deposition, adsorption, flotation precipitation and decomposition.

The objective of this study was to understand the application of batch mode EC for the conventional pollutants removal from municipal wastewater. Experiments were performed to see the effects of operating parameters like electrode spacing and current on efficiency of COD, turbidity, ammonia and phosphate removal.

Materials and methods

Wastewater:

Municipal wastewater (MWW) was collected from B.I.T. Mesra, Ranchi, Jharkhand, where wastewater originated from offices, hostels, faculty and staff quarters. Grab method was used for sampling wastewater on daily basis.

Experimental setup:

Electrocoagulation experiments were performed in a borosilicate glass beaker of 1 L capacitywith working volume of 0.8 L. Lab reactor unit consisted of aluminium electrode as anode and stainless steel as cathode. DC power was used in the experiments to supply constant voltage of 12 V. Due to oxide formationon electrodes it gets passivated over time and to prevent this, electrodes were first cleaned with sandpaper followed by distilled water and then washed again with distilled water after immersing for 10 min in hydrochloric acid (5 *M*) solution. The dried electrodes were weighed before and after every run[13].

Experimental procedure:

Batch experiments were performed in laboratory. The metal plate acting as anode and cathode were arranged parallelly acting as monopolar electrodes, having an inter-electrode distance of 3 cm, 5 cm and 7 cm. The dimensions of both the electrodes were 7 cm×6.5 cm×0.4 cm and total effective electrode area was 91 cm^2. Reactor contents were agitated by using a magnetic stirrer (Make: REMI) with the agitation speed of ~350 rpm. Experiments were performed for 120 min while at every 15 min interval samples were collected and then analysed. The settled samples were analysed for different parameters. During all the experiments, neither electrolyte (e.g. NaCl) was added externally nor pH was adjusted. All the experiments were performed in triplicates.

Analytical procedure:

Wastewater analysis was performed as per the procedures detailed in APHA[14]. A Hanna (HI 98130) multiparameter water quality instrument was used for the measurements of pH, and electrical conductivity. A turbidity meter was used to measure turbidity. Open reflux dichromate method was used for COD measurement[14]. Ammonia and phosphate were analysed using phenate and stannous chloride method, respectively.

Operating cost:

To calculate operating cost of the electrocoagulation pro-

cess electrical energy and electrode material consumption is an important consideration (Thakur *et al.* 2017). These two factors were calculated according to eq. (6):

$$\text{Operating cost} = aC_{\text{energy}} + bC_{\text{electrode}} \quad (6)$$

Here C_{energy} and $C_{\text{electrode}}$ are quantities per m^3 of water. The unit prices *a* and *b* were taken rate as per current Indian market: *a* = electrical energy price of Rs. 5.75/kW h for Jharkhand rural areas and *b* = materials of electrode price of Rs. 135/kg for Al. Since in this experiment 30 g of aluminium was used, aluminium price here was Rs. 4.05. Consumption of electrical energy was calculated using eq. (7) (Kobya *et al*. 2016).

$$C_{\text{energy}} = \frac{U \times i \times t_{\text{EC}}}{v} \quad (7)$$

Here, *U* is the applied voltage (v), *i* the applied current (A), *t* is the treatment time (h), *v* is the volume (m^3) of water used for treatment. Electrode consumption can be calculated as per eq. (8).

$$C_{\text{electrode}} = \frac{i \times M_{\text{w}} \times t_{\text{EC}}}{F \times v \times z} \quad (7)$$

Here, $C_{\text{electrode}}$ is the consumption of electrode(kg/m^3), *z* is the number of electron transferred M_{w} is molecular mass of aluminium (26.98 g/mol) and *F* is Faraday's constant (96487 C/mol).

Results and discussion

Municipal wastewater characteristics are presented in Table 1. Municipal wastewater is of moderate strength as evident from the BOD and COD concentration.

Table 1. Physico-chemical characteristics of municipal wastewater (n = 3, ±S.D.)

Sl. No.	Parameters	Values
1.	pH	6.9±0.4
2.	EC (mS/cm)	0.7±0.3
3.	TDS (mg/L)	480±50
4.	COD (mg/L)	460±120
5.	BOD (mg/L)	250±80
6.	Turbidity (NTU)	45±10
7.	Phosphate (mg/L)	12±2
8.	Ammonia (mg/L)	45±4

Effects of inter-electrode distance:

Inter-electrode spacing plays vital role in electrocoagulation. When inter-electrode distance was increased ohmic potential of a cell also decreases. When sample conductivity is low inter-electrode spacing also affects electrolysis energy consumption. The distance between electrodes was varied at 3 cm, 5 cm and 7 cm to determine the effect of inter-electrode spacing. The highest removal efficiency was observed at 3 cm inter-electrode distance. It is well known that electrical energy consumption decreases with less inter-electrode distance, which reduces motion resistance and improves process efficiency[15]. Different electrode spacing resulted in significant differences in removal efficiencies. The distance between electrodes has a direct effect on the IR-drop, which can be altered by decreasing the inter-electrode distance. It is obvious that when short inter-electrode distance between the electrodes are used lower removal efficiencies of the pollutants from water can occur because several phenomena can be affected (e.g. coagulation, flocculation, precipitation and electro flotation). These effects impact the flocs formation and their precipitation avoiding the formation of aggregates because the high electrostatic effect hinders the particles collision. However, more gap between electrodes affect the formation of flocs[16]. From the Fig. 1(a) it can be observed that for all the different electrode distance there was an increasing trend of pH with increasing electrolysis time. A slight decrease in conductivity during electrocoagulation is shown in Fig. 1(b). It is known that conductivity decreases as a function of electrolysis time[17]. At three different inter-electrode distance there was 95% turbidity removal at 3 cm, and 94% and 93% removal at 5 cm and 7 cm, respectively (Fig. 1c).

From Fig. 1(d) is evident that there is effect of inter-electrode distance on COD removal. It was observed that at 3 cm inter-electrode distance there was 89% COD removal and at 5 cm and 7cm electrode distance COD removal was 84% and 82%, respectively. Higher COD removal is due to adsorption of organic matter on the $Al(OH)_3$ flocs formed *in situ* during the reaction. Zodi *et al.*[18] also found 50% reduction in COD during 60 min of treatment and 66% after 90 min time using Al electrode. There was 55% ammonia removal at 3 cm inter-electrode distance and 38% and 32%, respectively at 5 cm and 7 cm inter-electrode distance (Fig. 1e).

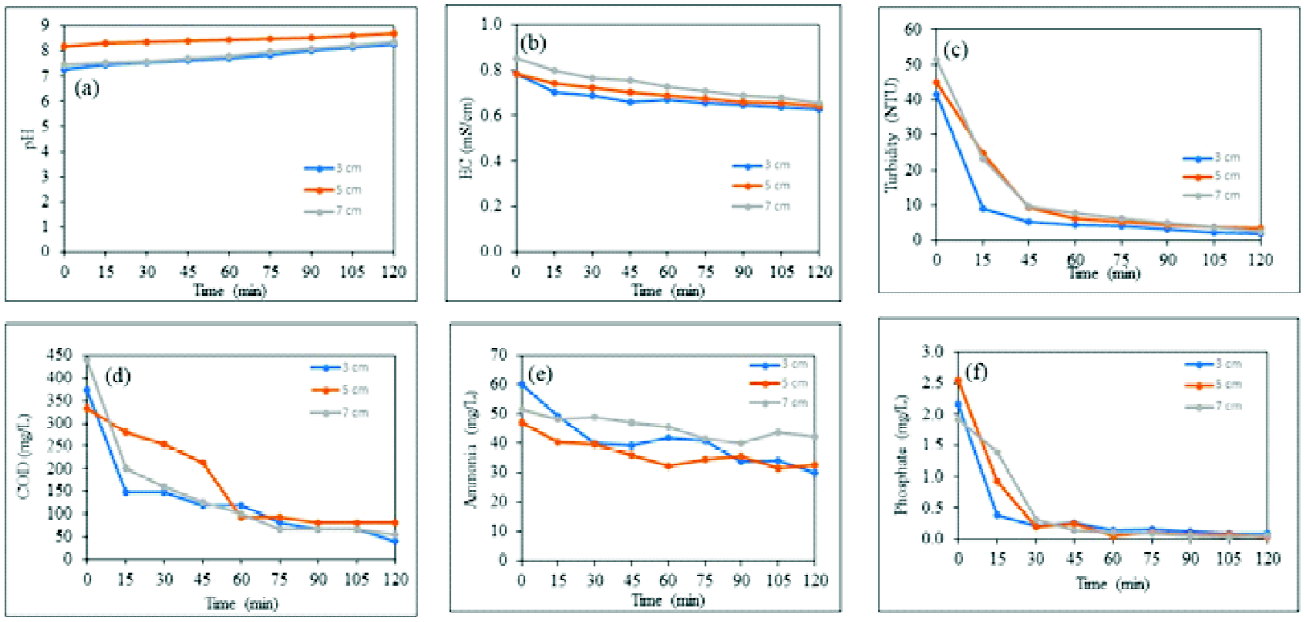

Fig. 1. Effect of inter-electrode distance on (a) pH, (b) EC, (c) turbidity, (d) COD, (e) ammonia and (f) phosphate removal with time.

There was also significant difference of inter-electrode distance on phosphate removal. Fig.1(f) shows 99% phosphate removal at 3 cm distance whereas 98% phosphate was removed at 5 cm and 98% at 7 cm. Omwene *et al.*[19] reported 99.99% phosphate removal using hybrid aluminium (Al)-iron (Fe) anodes and titanium cathode at initial pH of 4, current density of 20 A/m^2 and EC time of 80 min from domestic wastewater.

Effect of current:

Current plays an important role which affect the EC efficiency. This is responsible for release of metal ion from the anode into the solution which work as coagulants. Current is also important for bubble production and floc growth. The amount of oxidized metal increases with increasing current, thus generates higher amount of hydroxide flocs and reactive species which aggravates dissolved pollutants removal. It was observed that the increase of current also accelerated the rate of decontamination and generates higher quantity of sludge flotation during experiments, thus it leads to best removal of contaminants. It is also reported that H_2 bubble production increases during EC process and their size decreases when the applied current increases, this enhances pollutant removal efficiency and subsequently big quantity of floated sludge[20]. As shown in Fig. 2(a) pH at different current, it is observed that there were almost negligible changes at 1 A, though increasing trend can be observed at 0.5 and 2 A. Lewis acidity of aluminium ions is mainly responsible for this low increase of pH, which can be counter-balanced by the constant generation of OH^- at the cathode[21]. Thus, in electrocoagulation the hydrogen evolution at cathode and also H_2O reduction at cathode leads to increase in pH, as shown in eq. (4), whereas the oxidation of water at anode and formation of the different aluminium/iron species by the combination of the electro-dissolved ions with hydroxyl ions, results into decrease in pH. Due to increasing current there is drop in the conductivity (Fig. 2b). Fig. 2(c) shows that current of 2 A is optimum for the turbidity removal in comparison to other current values tested. It is known that when the current is higher the treatment time, is shorter[20]. This can be related that at higher current, more amount of precipitate is formed due to the enhanced anodic dissolution of aluminium resulting into higher pollutants removal. However, bubble size decreases and rate of bubble generation increases with increasing current. These effects are useful because a lot of pollutant can be removed due to H_2 flotation. The decrease in turbidity removal with increasing current confirms that here adsorption phenomenon for oxygen bubble takes place which is produced at the anode. Merzouk *et al.*[22] also found 89.54% turbidity removal at 1 cm electrode distance and initial pH

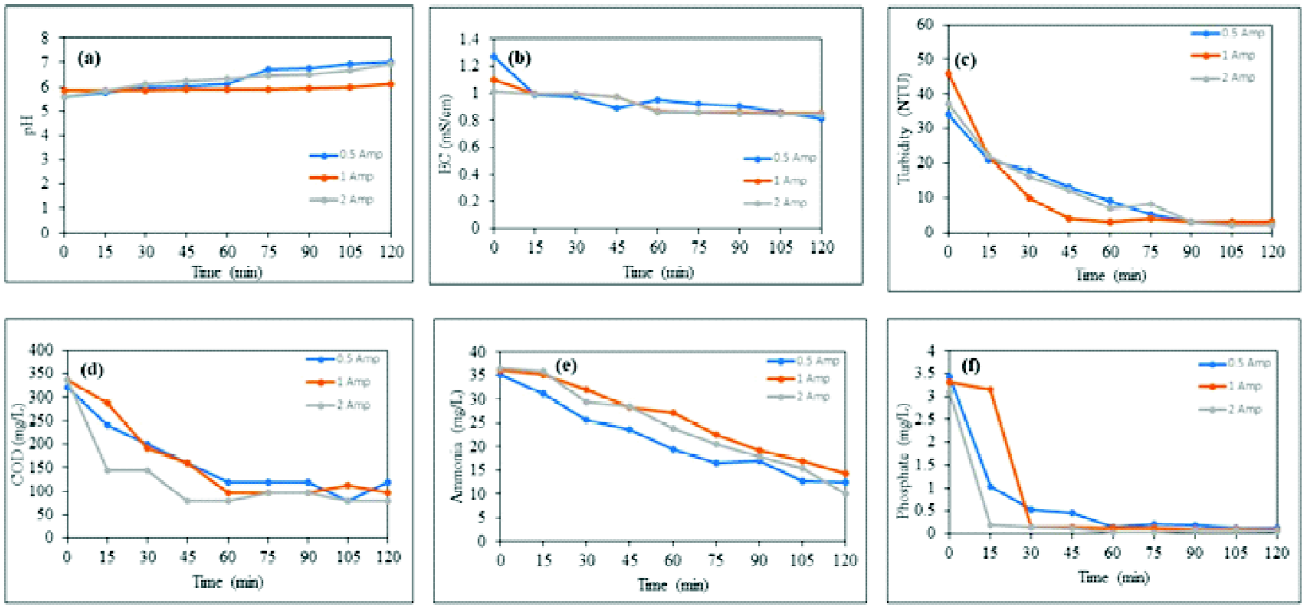

Fig. 2. Effect of current on (a) pH, (b) EC, (c) turbidity, (d) COD, (e) ammonia and (f) phosphate removal with time.

7.6 and current density 11.56 mA/cm^2. Fig. 2(d) depicts that 2 A current optimum for COD removal as at this current maximum COD removal can be observed. Ammonia removal at different current applied showed that maximum removal 72% occurred at current of 2 A (Fig. 2(e)). Zailani *et al.*[23] also found 37% ammonia removal using Al anode at current density of 200 A/m^2 and pH 4 from leachate. Bukhari *et al.*[24] reported 47% ammonia removal using Al electrode and 36% ammonia removal using copper electrode at 15 V and 1.48–1.50 A current, which was lower than that recorded during this study. Phosphate removal increased with increasing electrolysis time (Fig. 2f). 2A current was optimum for phosphate removal from municipal wastewater, 99% phosphate removal was achieved at this current compared to 0.5 and 1 A.

Operating cost:

Operating cost was calculated using eqs. (6), (7) and (8) to 0.12, 0.24 and 0.48 USD/m^3 for 0.5 A, 1 A and 2 A, respectively. This indicates that when current increases operating cost also increases. Thakur *et al.*[25] calculated operating cost for continuous electrocoagulation process for arsenic and fluoride removal and found that operating cost decreases when increased in flow rate and for all flow rates of 0.48, 0.88 and 1.40 L/h operating cost calculated was 0.728, 0.358 and 0.216 USD/m^3, respectively.

Conclusions

This study conclude that electrocoagulation process can be applied for the municipal wastewater treatment. In this study removal of turbidity, COD, ammonia and phosphate from municipal wastewater was assessed. It was found that 3 cm electrode spacing was optimum for COD removal, at this distance COD removal efficiency of 89% was achieved, which was maximum in this study compared to other two interelectrode distance. Ammonia reductionat 3 cm, 5 cm and 7 cm interelectrode distance was 55%, 38% and 32% respectively. Phosphate removal was 99% at 3 cm and 98% both at 5 cm and 7 cm. Turbidity removal also followed the same trend, at 3 cm maximum removal of 95.5% was recorded and at 5 cm and 7 cm respective 94% and 93% removal occurred. This study showed that electrocoagulation can be applied for turbidity, COD, phosphate and ammonia removal from municipal wastewater with aluminium and stainless-steel electrode. Operating cost determination showed that increase in current increases operating cost. Further studies on continuous mode operation process will be helpful in taking the technology forward.

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