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# Investigation of the electro-coagulation treatment process for the removal of total suspended solids and turbidity from municipal wastewater

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#### Abstract

In this work, raw municipal wastewater was electro-coagulated for the removal of total suspended solids (TSS), turbidity, and particulate BOD using stainless steel electrodes. The removal efficiency of TSS and turbidity is shown to depend on the amount of iron generated from the anode of the reactive electrode used in this study, when applying the lower currents of 0.05 A and 0.1 A. For such lower currents, the results suggested that the removal is consistent with charge neutralization coagulation mechanism. When applying higher currents of 0.2 A, 0.4 A, and 0.8 A, the results suggested that the dominant removal mechanism is sweep-floc coagulation as the generated soluble ferrous ions are converted to insoluble ferric ions due to oxidation with chlorine generated during the electrochemical process at the higher currents. The highest TSS removal efficiency of 95.4% occurred at a current of 0.8 A and contact time of 5 min. The effect of electro-coagulation on the removal of particulate BOD was shown to depend on the TSS removal efficiency.

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# 1. Introduction

Wastewater treatment serves two main objectives, protecting the environment and conserving fresh water resources. Treated municipal wastewater is considered, in many major cities of the world, as a supplementary water source for several uses such as agricultural irrigation, landscaping, industrial activities (cooling and process needs), groundwater recharge, recreational and other uses (Tchobanoglous et al., 2003). The continued dependence on treated municipal wastewater for the previously mentioned uses coupled with emerging stricter policies/guidelines for such uses prompted scientists and researchers to concentrate in improving current treatment technologies and to come up with innovative ideas to treat municipal wastewater in an efficient and cost effective manner.

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Biological processes are considered the main method for any treatment scheme employed for the treatment of municipal wastewater. The biological process depends, among other factors, on the concentration of BOD. As the BOD concentration increases, the aeration volume and the oxygen requirements increases resulting in higher capital and energy costs. Therefore, reduction in the concentration of BOD entering the aeration tank would result in substantial reduction in the energy requirements. BOD exists in municipal wastewater in two forms, particulate and soluble. The particulate form of BOD constitutes the major fraction ranging from 60% to 80% of total BOD (Stearns and WheleI', 2005; Nieuwenhuijzen et al., 2001; HydroQual, Inc., 2005). In conventional treatment, primary sedimentation is used to reduce the BOD loading entering the aeration tank primarily through settling of particulate (suspended solids) BOD. The efficiency of a well designed and operated primary sedimentation ranges between 50% and 70% (Tchobanoglous et al., 2003). To enhance the removal

efficiency of suspended solids, chemical coagulants such as alum, ferric chloride, ferric sulfate, and lime are usually used resulting in suspended solids removal efficiencies ranging between 80% and 90% (Tchobanoglous et al., 2003). The major disadvantages of the chemical precipitation process for the removal of suspended solids are that it involves the addition of chemicals which can be costly and results in the increase of total dissolved solids (TDS). Therefore, these is a need to look for an alternative process that could efficiently remove suspended solids at relatively low capital and operating costs. Research, in the past few decades, have shown that the electro-coagulation process can be developed as a potential one for the treatment of different wastewaters including textile (Vlyssides et al., 2000; Gurses et al., 2002; Kobya et al., 2003; Zaroual et al., 2006), food (Beck et al., 1974), restaurant (Chen et al., 2000b,a), urban (Pouet and Grasmik, 1995), gray water (Lin et al., 2002), and for the removal of heavy metals (Adhoun et al., 2004; Pogrebnaya et al., 1995; Balasubramanian and Madhavan, 2001; Kumar et al., 2004) and suspended particles (Donini et al., 1994; Abuzaid et al., 1998; Mattenson et al., 1995).

Electro-coagulation is a complex process involving chemical and physical mechanisms operating simultaneously to remove pollutants from wastewaters. When reactive electrodes are used such as iron or aluminum, metal ions are produced (in situ) resulting in the removal of suspended solids. The process takes place in three successive stages: (i) formation of coagulants by the dissolution of metal ions from the anode of the reactive electrode, (ii) destabilization of particulate\colloidal suspension, (iii) aggregation of the destabilized suspension and formation of flocs (Mollah et al., 2004). For iron electrodes, ferrous ions (Fe2+) are released into solution by electrolytic oxidation of the anode material. The produced ferrous ions hydrolyze to form monomeric hydroxide ions and polymeric hydroxide complexes depending on the pH of the solution. These polymeric hydroxides, which are highly charged cations, destabilize the negatively charged colloidal particles allowing for their aggregation and formation of flocs. Also, depending on the solubility of the metal hydroxide, when the amount of iron added to water exceeds the solubility of the metal hydroxide, formation of the amorphous metal hydroxide precipitates takes place causing sweep-floc coagulation (Benefield et al., 1982).

The aim of this study is to investigate the potential of the electro-coagulation process using iron electrodes in the removal of suspended solids, turbidity and particulate BOD from municipal wastewater. The effects of applied current and the amount of iron generated on the removal efficiency of the investigated parameters are discussed.

#### 2. Methods

Raw municipal wastewater samples were collected from the Northern Dhahran Sewage Treatment Plant located in the Eastern Province of Saudi Arabia. The electrochemical set-up shown in Fig. 1 was composed mainly of two stainless steel electrodes, a DC power source (Hampden, USA), a rheostat (Engield-Middlesex, UK) to keep the current invariant, and a 1.51 capacity test beaker used as a reactor. Two Pyrex glass strips attached to the inside of the glass beaker were used as baffles to create turbulence and facilitate proper mixing of the sample using a magnetic bar stirrer. Each electrode had an active surface area of 44 cm<sup>2</sup> and a spacing of 3 cm between electrodes. The electrodes are inserted in the test beaker. An ammeter (Hampden, USA) was used to measure the current. An electrical switch was used to change the polarity of electrodes in order to reduce the passivation phenomenon that usually occurs during the process and impedes the oxidation/reduction reactions.

The experiments were carried out in a batch mode. For each experiment, a wastewater sample of 1.2 l was collected in the electrochemical cell with the stainless steel electrodes dipped into the sample solution up to an active surface area of 88 cm<sup>2</sup>. Five different currents were applied (0.05, 0.1, 0.2, 0.4 and 0.8 A). For each current, contact time of 5, 10, 20, 30, and 50 min were used. Therefore, a total of 25 batch experiments were carried out. After the pre-specified contact time in the electrochemical cell had passed (i.e., after each batch experiment), the sample was transferred into another beaker, and measured for pH. The measured sample was then taken to a standard jar test apparatus, where it was rapidly mixed at 100 rpm for 1 min, and moderately mixed (flocculated) at 30 rpm for 20 min. The

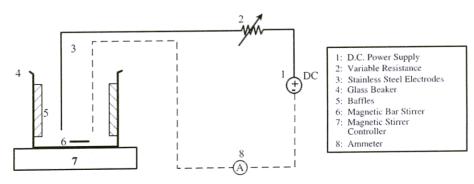


Fig. 1. Schematic diagram of experimental set-up.

flocculated sample was then kept undisturbed for 20 min in order to allow the flocs to settle. Subsequently, after settling, a 250 ml sample of supernatant was collected to perform the analysis of total suspended solids (TSS), turbidity, and BOD.

TSS measurement was carried out by filtering the samples influent (raw wastewater) and effluent (the supernatant of the treated sample after settling) using 0.45 µm filter papers. The BOD of the suspended solids was determined by measuring the total BOD of the sample before filtering through 0.45 µm filter papers (particulate plus soluble BOD) and after filtration (soluble BOD). The difference between the two measurements gives the particulate BOD. The BOD tests were carried out in accordance with the method described in the Standard Methods for the Examination of Wastewater (APHA, 1995).

#### 3. Results and discussion

## 3.1. Influent raw wastewater characteristics

Table 1 shows the characteristics of the influent raw municipal wastewater samples used in the experiments. As observed, the average BOD concentration is in the lower side which is typical of the characteristics of the wastewater of the Eastern Province of Saudi Arabia. Such low concentration may be due to the fact that a large portion of the domestic water supply is brackish well water. It can also be observed that most of the BOD, nearly 68%, is in the particulate form. The high raw wastewater conductivity is advantageous to the electrochemical treatment since it will eliminate the need to add an electrolyte that is necessary to facilitate the passage of current in the wastewater solution. Also, the presence of chloride at relatively high concentration helps in the production of chlorine as a result of the electrochemical process. Chlorine is an oxidizing agent that can participate in oxidizing soluble ferrous ions into insoluble ferric ions as will be discussed later on.

### 3.2. Total suspended solids (TSS) removal

The relationship between the TSS removal efficiency and contact time for the range of currents used is depicted in

Table 1
Physicochemical characteristics of the municipal wastewater used in the study

Influent parameters	Concentration range	Average concentration
TSS (mg/l)	126–160	143
Turbidity (NTU)	49-53	51
Total BOD (mg/l)	84-112	98
Soluble BOD (mg/l)	26-32	29
Particulate BOD (mg/l)	51-84	68
pH	6.9-7.1	7
Chloride (mg/l)	1238	1238
Conductivity ( $\mu\Omega^{-1}/cm$ )	4000	4000

Fig. 2. The figure shows that as the contact time increased from 5 to 50 min, removal efficiencies increased from 42.9% to 78.6% and from 56.3% to 87.5% for the currents of 0.05 A and 0.1 A as a function of time, respectively. For the 0.2 A, the removal efficiency increased up to 30 min of contact time and beyond that no further increase is observed. The highest removal efficiency of 92.5% for the 0.2 A current occurred at a contact time of 30 min. For the currents of 0.4 A and 0.8 A, the highest removal efficiencies of 92.1% and 95.4%, respectively, occurred at a contact time of 5 min. Further increase in contact time up to 10 min resulted in decrease of the removal efficiency. However, beyond the contact time of 10 min, removal efficiency continued to increase as a function of time.

The amount of iron released into solution by electrolytic oxidation of the anode material is a function of current and time and can be calculated using the following form of Faraday's law:

$$w = \frac{itM}{ZF},\tag{1}$$

where,

w is the iron dissolved (g);

*i* is the current (A);

t is the contact time (s);

M is the molecular weight of Fe (M = 55.85);

Z is the number of electrons involved in the redox reaction (Z = 2); and

F is the Faraday's constant (96500 C/mol of electrons).

Fig. 3 shows the TSS removal efficiency as a function of the amount of iron generated for the different currents used in this study. It can be seen that the removal efficiency for the lower currents of 0.05 A and 0.1 A is dependant of the

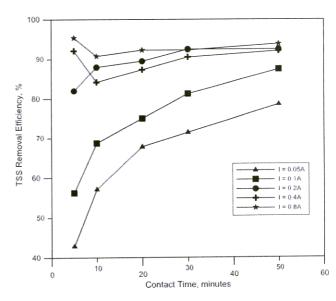


Fig. 2. The relationship between TSS removal efficiency and contact time for the range of currents used in this study.

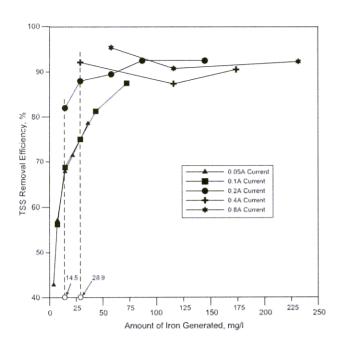


Fig. 3. TSS removal efficiency as a function of the amount of iron generated for the different currents used in this study.

amount of iron generated. In other words, as the amount of iron generated increases, removal efficiency increases proportionally. However, as the current increased to 0.2 A and higher, the relationship between removal efficiency and amount of iron released is no longer valid. For example, the amount of iron generated at a current of 0.05 A and contact time of 20 min is 14.5 mg/l which is equivalent to the amounts generated at a current of 0.1 A with 10 min of contact time and 0.2 A with 5 min of contact time. For the same amount of iron generated (14.5 mg/l), the removal efficiencies for the 0.05 A and the 0.1 A were 67.85% and 68.75%, respectively, which were almost identical. However, for the 0.2 A with the same amount of iron generated of 14.5 mg/l, the removal efficiency was much higher reaching about 82%. Again, the removal efficiency at a current of 0.1 A and 20 min which generated 28.9 mg/l of iron was 75%, while the removal efficiency for the same amount of iron generated at the currents of 0.2 A and 0.4 A were about 88% and 92%, respectively.

In electro-coagulation using iron electrodes, two mechanisms for the production of metal hydroxides have been proposed in the literature as the following:

#### Mechanism I:

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

$$2Fe^{2+} + 5H_2O + \frac{1}{2}O_2 \rightarrow 2Fe(OH)_3(s) + 4H^+$$
 (3)

Mechanism II:

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

$$Fe^{2+} + 2OH^{-} \rightarrow Fe(OH)_{2}(s) \tag{5}$$

From the above mechanisms, we can see that iron ions can either exist in the ferrous state, where iron carries two positive charges as in Eq. (5) or in the ferric state, where iron carries three positive charges as in Eq. (3). The ferric state which is commonly referred to as the oxidized state is much less soluble in water than the ferrous state. If iron is present in the soluble ferrous state, then we can postulate that the dominant mechanism of particulate removal is charge neutralization. From this work, we can see that the trend of suspended solids removal at the lower currents of 0.05 A and 0.1 A are consistent with charge neutralization mechanism, where removal increases as the coagulant dose increases. The soluble ferrous form of iron can be converted to the insoluble ferric from by oxidation. As mentioned previously, the wastewater used in this study contains relatively high concentration of chloride ions. The electrochemical process through anodic oxidation converts chloride ions into chlorine gas (Bukhari et al., 1999). Chlorine is used as an oxidant to convert ferrous iron to insoluble ferric iron. The results obtained at higher currents (0.2 A, 0.4 A, and 0.8 A) suggest that the dominant mechanism of particulate removal is sweep-floc coagulation. At such higher currents (higher potential) more of chlorine gas is produced than the lower current resulting in converting ferrous iron to insoluble ferric iron, hence, effecting sweep-floc coagulation. In support of our findings, it has been shown by Murugananthan et al. (2004) that the electro-coagulation process using iron electrodes at higher potential favors the formation of Fe(III) ions. Precipitation of Fe(III) hydroxides have a better coagulating character than Fe(II) hydroxides (Zaroual et al., 2006).

# 3.3. Turbidity removal

Fig. 4 shows the residual turbidity observed as a function of contact time for the range of currents used in this study. At a current of 0.05 A, the residual turbidity is observed to decrease as the contact time increased. The lowest residual turbidity of 14.3 NTU for the 0.05 A current occurred at a contact time of 50 min. When the current increased to 0.1 A, the residual turbidity is observed to decrease as a function of time at a higher rate than the 0.05 A current. The lowest residual turbidity of 9.3 NTU for the 0.1 A current occurred at a contact time of 30 min. Further increase in current to 0.2 A, resulted in a sharp decrease of residual turbidity from the initial value of 49.0 NTU down to 13.4 NTU at the first 5 min of contact time. Further increase in contact time resulted in gradual decrease of residual turbidity down to 8.6 NTU at a contact time of 20 min. However, beyond the 20 min of contact time, the residual turbidity is observed to increase up to 21.5 NTU at a contact time of 50 min.

For the higher current of 0.4 A, the residual turbidity decreased from the initial value of 53.5 NTU down to 5.1 NTU in the first 5 min of contact time. However, further increase in contact time to 10 min resulted in a marked

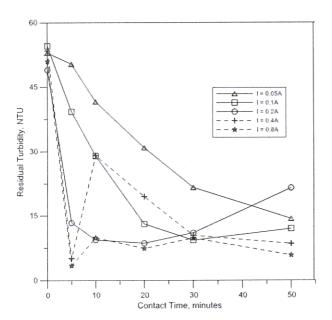


Fig. 4. Residual turbidity observed as a function of contact time for the range of currents used.

increase in residual turbidity up to 29.1 NTU, beyond which a gradual decrease in residual turbidity is observed down to 8.5 NTU at a contact time of 50 min. Similar trend of sharp decrease of residual turbidity in the first 5 min of contact time observed for the 0.2 A and 0.4 A currents is also observed for the 0.8A current. The residual turbidity for the 0.8 A current decreased from the initial value of 51 NTU down to 3.4 NTU in the first 5 min of contact time. However, further increase in contact time resulted in a slight increase in residual turbidity.

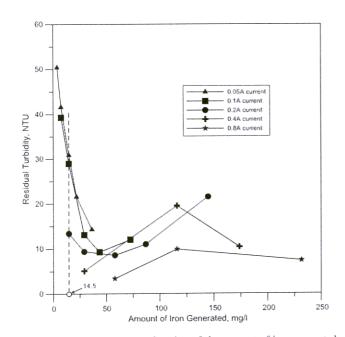


Fig. 5. Residual turbidity as a function of the amount of iron generated for the different currents used.

Fig. 5 shows the residual turbidity as a function of the amount of iron released for the different currents used in this study. It can be seen that the residual turbidity for the lower currents of 0.05 A and 0.1 A is dependant of the amount of iron released in the same manner as in the TSS case. As the amount of iron released increases, residual turbidity decreases proportionally. As discussed previously in the TSS case, this trend is consistent with charge neutralization mechanism, where residual turbidity decreases as the coagulant dose increases. However, as the current increased to 0.2 A and higher, the residual turbidity is no longer dependant on the amount of iron released. From Fig. 5, we can see that for the same amount of iron released (14.5 mg/l), the residual turbidity for the

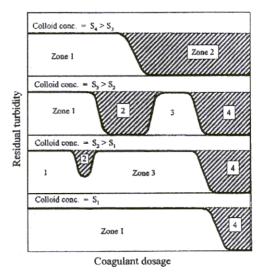


Fig. 6. Schematic diagram of coagulation curves. The shaded areas represent regions in which coagulation occurs (after Stumm and O'Melia, 1968).

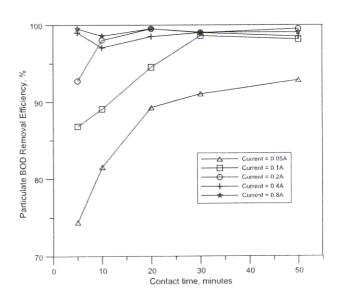


Fig. 7. The relationship between particulate BOD removal efficiency and contact time.

0.05 A and the 0.1 A were 41.6 NTU and 39.3 NTU, respectively, which were almost identical. Whereas, for the 0.2 A with the same amount of iron released of 14.5 mg/l, the residual turbidity was much lower down to 13.4 NTU.

The increase in turbidity observed at different contact times (coagulant doses) beyond the contact time of 5 min for the currents of 0.2 A, 0.4 A, and 0.8 A maybe explained based on Fig. 6 adapted from Stumm and O'Melia (1968). Fig. 6 shows a schematic presentation of the residual turbidity as a function of coagulant dose. From the figure, we can see that if insufficient coagulant dose is applied, destabilization does not occur (zone 1). If coagulant dose is increased, destabilization takes place (zone 2) effecting turbidity removal. However, further increase in coagulant dose can lead to particle restabilization (zone 3). At zone 4, sweep-floc coagulation occurs.

#### 3.4. Particulate BOD removal

As mentioned in the introduction, the BOD of municipal wastewater results from particulate and soluble matter with the particulate source being the major contributor to municipal BOD. The effect of electro-coagulation on the removal of particulate BOD associated with TSS removal is discussed in this section.

The relationship between the particulate BOD removal efficiency and contact time is shown in Fig. 7. It can be seen that as the contact time increased from 5 to 50 min, the removal efficiency for the 0.05 A current increased from 72.4% to 92.9%, respectively. For the 0.1 A current, as the contact time increased from 5 to 30 min the removal efficiency increased from 86.8% to 98.6%, respectively, and beyond the 30 min no further increase in removal efficiency is observed. For the 0.2 A current the removal

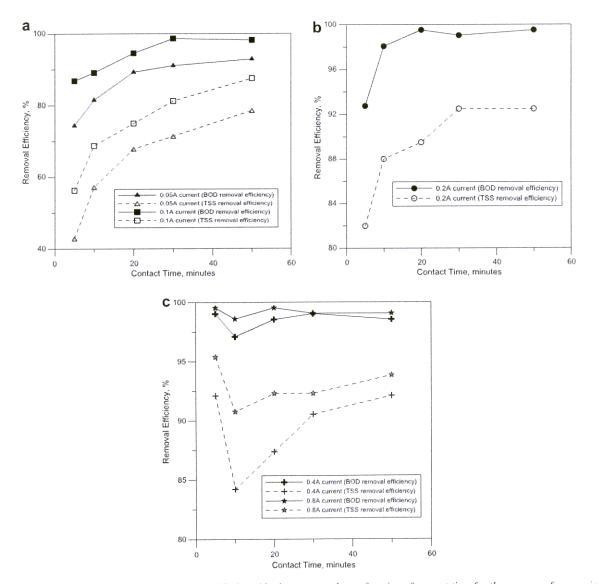


Fig. 8. The removal efficiencies of particulate BOD and TSS plotted in the same graph as a function of contact time for the purpose of comparing trends (a) Removal efficiencies for 0.05 A and 0.1 A currents. (b) Removal efficiencies for 0.2 A current. (c) Removal efficiencies for 0.4 A and 0.8 A currents.

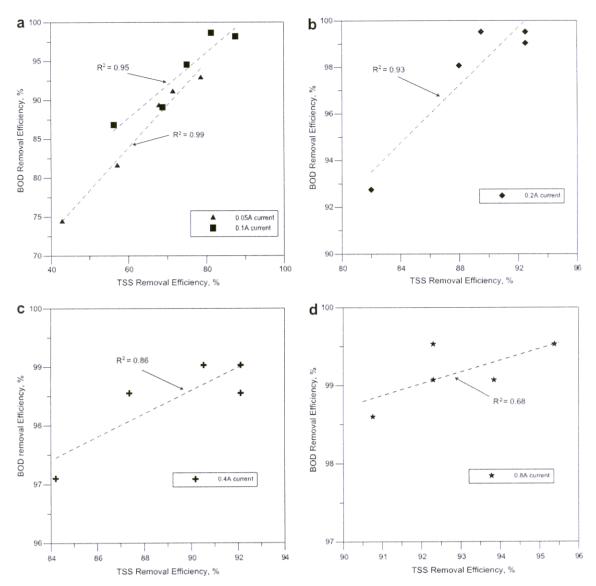


Fig. 9. The relationship between BOD and TSS removal efficiencies. (a) Linear curve fit for 0.05 A and 0.1 A currents. (b) Linear curve fit for 0.2 A current. (c) Linear curve fit for 0.4 A current. (d) Linear curve fit for 0.8 A current.

efficiency increased from 92.8% to 99.5% as the contact time increased from 5 min to 20 min and beyond the 20 min contact time no further increase is observed. However, for the higher currents of 0.4 and 0.8 A, the removal efficiency is observed to reach its maximum removal (around 99%) in the first five min of contact time beyond which the removal starts to oscillate between 98% and 99% efficiencies.

Fig. 8 shows the removal efficiencies of particulate BOD and TSS plotted in the same graph as a function of contact time for the different currents used in this study. The figure is separated into three graphs (a, b, and c) for better illustration. The graphs clearly show that BOD removal is dependant on the TSS removal as both trends are similar. The relationship between TSS removal and BOD removal is shown to be linear as depicted in Fig. 9. The figure shows that for the currents of 0.05 A, 0.1 A and 0.2 A the curve fit

has high correlation coefficients ( $R^2$ ) ranging from 0.99, 0.95, and 0.93, respectively. However, for the higher currents of 0.4 A and 0.8 A, the correlation coefficients are low down to 0.86 and 0.68, respectively. The low correlation coefficients for the 0.4 A and 0.8 A currents are probably due to the fact that the removal efficiencies of BOD and TSS are in the higher range and any slight change in the measurement produces higher errors.

### 4. Conclusions

The effect of electro-coagulation on the removal efficiency of the TSS is shown to be dependent on the amount of iron generated for the lower currents of 0.05 A and 0.1 A. In other words, as the amount of iron generated increased, removal efficiency increased regardless of the current used to generate such amount of iron as long as

the current is in the range of 0.05–0.1 A. The dominant removal mechanism of suspended/colloidal matter, when using the lower currents of 0.05 A and 0.1 A was shown to be consistent with charge neutralization coagulation mechanism. For the higher currents of 0.2 A, 0.4 A, and 0.8 A, the results suggest that the dominant removal mechanism of suspended/colloidal matter is consistent with sweep-floc coagulation mechanism as more of the soluble ferrous ions are converted to insoluble ferric ions due probably to oxidation of ferrous ions to ferric ions with chlorine produced in the electrochemical process. The highest removal efficiency of 95.4% occurred at a current of 0.8 A and contact time of 5 min.

The residual turbidity for the lower currents of 0.05 A and 0.1 A is shown to depend on the amount of iron released in the same manner as in the TSS case. As the amount of iron released increases, residual turbidity decreases proportionally. This trend is consistent with charge neutralization mechanism, where residual turbidity decreases as the coagulant dose increases. For the higher currents of 0.2 A, 0.4 A, and 0.8 A, the results suggest, in the same manner as in the TSS case, that the dominant removal mechanism of turbidity removal is consistent with sweep-floc coagulation mechanism.

The effect of electro-coagulation on the removal of particulate BOD, a major constituent of municipal wastewater BOD, was shown to depend on the TSS removal efficiency. The relationship between TSS and BOD removal is shown to be linear. The highest removal efficiency of particulate BOD was around 99% and occurred at a current of 0.8 A and a contact time of 5 min.

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