

Original Research Paper

Remediation of Feedlot Nutrients Runoff by Electrocoagulation Process

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Abstract: Nutrient runoff from Concentrated Animal Feeding Operations (CAFOs) may cause ground and surface water pollution. Scientists and researchers are continually searching for appropriate technologies to mitigate feedlot nutrient runoff pollution. In this study, suitability of electrocoagulation treatment process was examined under laboratory conditions to mitigate nutrient pollutants from the feedlot runoff. Feedlot runoff was treated with three different type of electrodes such as aluminum (Al-Al), iron (Fe-Fe) and hybrid (Al-Fe) at different electrical voltage potentials (5, 10 and 15 Volts) for a designated time step (up to 30 min). The electrocoagulation cell consisted of two parallel rectangular plate electrodes, immersed in a beaker with 500 mL feedlot runoff and powered by a Direct Current (DC) supply. This study was conducted in batches at room temperature. Results indicated that Electrical Conductivity (EC), Total Phosphorus (TP), Total Nitrogen (TN) and Chemical Oxygen Demand (COD) concentration reduced significantly irrespective of electrode types. Overall, TP concentration reduction was higher (100%) followed by COD (50-75%) and TN (25-60%) concentration. Nutrient removal and specific electrical energy consumption increased with increasing voltage level. Aluminum electrodes were more effective than the other two electrodes for TP reduction at all applied potentials and COD reduction was better at lower applied potential. Hybrid electrodes (Al-Fe) reduced TN better than the other two electrodes.

Keywords: Feedlot Runoff, Nutrients, Electrocoagulation, Electrode Potential and Specific Electrical Energy Consumption

Introduction

Feedlot is a concentrated animal feeding operation where beef cattle are finished to slaughter weight and have little or no access to pasture land (Spellman and Whiting, 2010). Feedlot is a potential source of nutrient runoff, if manure is not managed properly. The runoff generated from the feedlot pen surfaces has a considerable amount of nutrients such as nitrogen, phosphorus, potassium along with organic matter, pathogen, hormone and antibiotics (Crane *et al.*, 1983; Dillaha *et al.*, 1989). This runoff may contaminate surface and groundwater, can cause eutrophication and reduce the oxygen level in surface water which may suppress the biodiversity of lagoons and estuaries (Ansari *et al.*, 2011; Hribar and Schultz, 2010; Prophet and Edwards, 1973). To mitigate this problem, researchers are trying to adopt different technologies such as membrane filtration, advanced oxidation

process, air flotation, distillation, evapotranspiration, nitrification, precipitation, ammonia stripping and electro-dialysis (Bensadok *et al.*, 2011; Ilhan *et al.*, 2008). Though some of these methods are effective, sometimes these methods may become complex, expensive and sophisticated which may require specialized technical knowledge (Crites *et al.*, 2014). Moreover, some of the methods may not be economically viable for livestock growers (Kim *et al.*, 2013). Therefore, electrocoagulation can be used for treatment of feedlot runoff to cope with this issue.

Electrocoagulation technology is a treatment process where electrical current is applied to treat and flocculate contaminants (Butler *et al.*, 2011; Mollah *et al.*, 2001). The electrocoagulation process works on the principle of oxidative or reductive chemistry and it needs relatively simple equipment called electrodes at ambient temperature and pressure. Electrocoagulation is

generated in-situ by electrolytic oxidation of an appropriate anode material (Mollah *et al.*, 2001). They also mentioned that in the coagulation process, charged ionic are removed from wastewater by allowing it to react (i) with an ion having opposite charge, or (ii) with floc of metallic hydroxides generated within the effluent. Electrocoagulation is environmentally compatible, low area demanded, small volume of sludge produced and short treatment time required (Chaturvedi, 2013; Inan and Alaydin, 2014). However, electrocoagulation process also possesses some challenges such as an expensive process due to current uses, an impermeable oxide film may be formed on the cathode and high conductivity of the wastewater suspension is required (Mollah *et al.*, 2001). Typically, runoff water has high conductivity, which overcome of these issues and the sludge produced during electrocoagulation can be used as a fertilizer or used for extracting different valuable elements (Bridle and Skrypski-Mantele, 2000; Gaber *et al.*, 2011; Sano *et al.*, 2012; Sethu *et al.*, 2008). The effluent produced from the electrocoagulation can be used for irrigation. In the past, limited studies on the use of electrocoagulation were performed on livestock wastewater including swine (Bejan *et al.*, 2007; Cho *et al.*, 2010; Laridi *et al.*, 2005; Rahman and Borhan, 2014), dairy (Bensadok *et al.*, 2011; Şengil, 2006; Tchamango *et al.*, 2010; Yavuz *et al.*, 2011), slaughter house wastewater (Bazrafshan *et al.*, 2012), industrial effluents (Ali and Yaakob, 2012; Basha *et al.*, 2008), pharmaceutical wastewater (Yi-zhong *et al.*, 2002), agroindustry (Kim *et al.*, 2013) and textile dye wastewater (Merzouk *et al.*, 2009). However, until today, electrocoagulation was not use to treat feedlot runoff. Therefore, this article investigated the electrocoagulation treatment of feedlot runoff in a batch under laboratory conditions

using different electrodes at varying applied electrical potential level. The specific objectives were to compare TP, COD and TN removal efficiencies and energy consumptions of three metal electrodes (iron-iron, aluminum-aluminum and iron aluminum combination) in treating feedlot runoff.

Materials and Methods

Description of Electrocoagulation Operation Systems

Parallel plates with identical dimensions of aluminum (Al-Al), iron (Fe-Fe) and hybrid (Al-Fe) electrodes pair were used in electrocoagulation process. Electrical power was applied through the single anode and cathode using a DC power source equipped with digital ammeter and voltmeter (BK precision 1621A DC regulated power supply equipment) and maintained at 5, 10 or 15 V electrical potential (Fig. 1). The submerged portion of an electrode was $90 \times 25 \times 1.5$ mm ($h \times b \times t$) though its actual dimension was $280 \times 25 \times 1.5$ mm ($h \times b \times t$). The space between the electrodes was kept constant at 8 mm and the effective submerged area was 4807.5 mm². Corresponding currents against applying voltage potentials were measured to determine electrical energy consumption. During electrocoagulation, the polarity of electrodes was altered manually to minimize passivation on electrodes and the runoff water in the beaker was mixed continuously with a 30 mm magnetic stirrer at 200 to 300 rpm. After electrocoagulation, the sludge was collected and filtered using 0.45 micron mixed cellulose ester filter (EZ-Pak membrane Filter, Cat# EZHAWG474) and dried in an oven at 105°C for the elemental analysis. Electrodes were rinsed with diluted hydrochloric acid (5% v/v) followed by De-Ionized (DI) water rinse to avoid the electrode passivation due to oxidation and contamination of products.

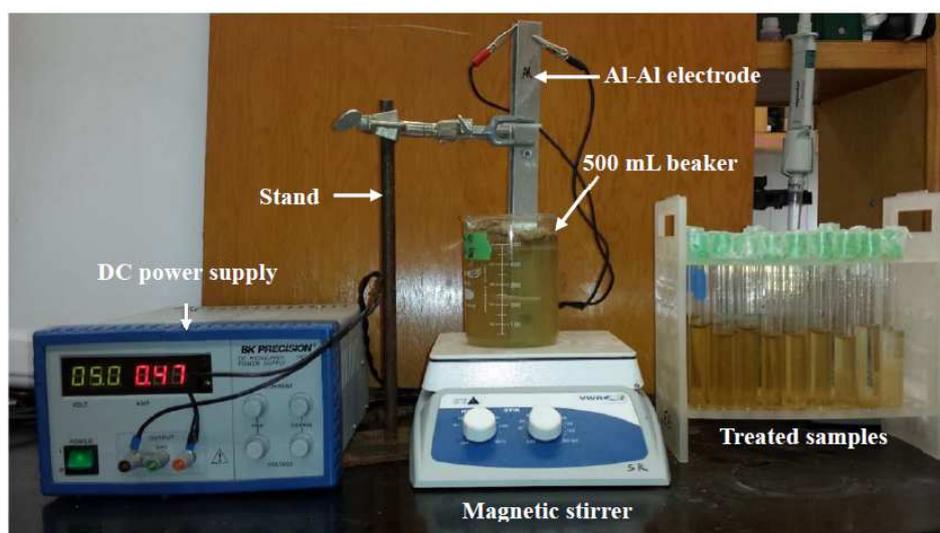


Fig. 1. Photographic views of the electrocoagulation setup used in this study

Feedlot Runoff Collection, Storage and Sample Collection

Feedlot runoff samples were collected from the Beef Research Centre at North Dakota State University, Fargo, North Dakota, USA. Collected sample was stored in a 20 L bucket at 4°C and analyzed at room temperature (25±2°C). During electrocoagulation, a 500 mL sample was placed into a 550 mL beaker. Initial pH and EC of runoff wastewater were measured with a handheld pH and EC meter (YSI Pro Plus, YSI Inc., Ohio, US). Total Solids (TS) contents were measured before starting electrocoagulation treatment. At predetermined times (0, 1, 2, 3, 5, 8, 10, 20 and 30 min of electrocoagulation) 10 mL of treated samples were pipetted in test tubes. These samples were left overnight (8-12 h) at room temperature for settlement and nutrient analysis was done later on using treated waste from the supernatant. In this experiment, three potentials such as 5, 10 and 15 VDC were applied for each electrode with three replicates. A total of 243 (3×3×3×9) samples were collected during the electrocoagulation study.

Sample and Data Analysis

Total Solids (TS) contents were measured following the standard procedure (method 2540B, APHA, 2005). The Hach Method 10127 (Molybdovanadate Method with Acid Persulfate Digested, 1-100 mgL⁻¹) was used for TP analysis. The Hach Method 10072 (Persulfate digestion method 2-250 mgL⁻¹) was used for TN analysis and Hach Method 8000 (Reactor digestion method 20-1500 mgL⁻¹) was used for COD analysis. Mineral concentration in the dried sludge was measured with Inductively Coupled Plasma Spectroscopy (ICP) using a 2010-11-15 Standard Method in the Wet Ecosystem Lab at North Dakota State University. The mean concentrations of pH and EC were compared before and after an electrocoagulation event. TN, COD and TP concentrations at each time step while treating the runoff were compared with initial concentration. The mean concentration of the pollutants (EC, pH, TP, COD and TN) and estimated removal efficiencies in each voltage potential and electrode type were compared using ANOVA. The null hypothesis tested in the experiment was that there is no significant difference in pollutant concentration across electrode types and applied electrode potentials. All statistical analyses were performed with SAS software (version 9.3) using the PROC means procedure at the 5% level of significance.

Calculation of Removal Efficiency and Specific Energy Uses

The removal efficiencies for TN, TP and COD were calculated using Equation 1. Similarly, the

electrical energy consumption per unit mass of the individual parameters and per unit volume of runoff processed was calculated using the Equations 2 and 3, respectively:

$$\% \text{Removal efficiency of parameter} = \frac{\text{Initial concentration}(C_i) - \text{final concentration}(C_f)}{\text{Initial concentration}(C_i)} \quad (1)$$

$$\text{Electrical energy consumption}(kWh) \text{ per unit mass of parameter} = \frac{V \times I \times t(kWh)}{\text{mass of parameter reduced}(kg)} \quad (2)$$

$$\text{Electrical energy consumption}(kWh) \text{ per unit volume} = \frac{V \times I \times t(kWh)}{\text{volume of runoff used}(m^3)} \quad (3)$$

Where:

V = Applied potential difference in electrocoagulation process, voltage

I = Current generated in electrocoagulation process, amperes

t = Time of electrocoagulation process, hours

Results and Discussion

Initial Concentration of Feedlot Runoff Nutrients

The average initial nutrient concentrations of 27 samples of feedlot runoff used in this study are listed in Table 1. Following electrocoagulation experiment, subsequent changes are reported and discussed in the following sections.

pH and Electrical Conductivity (EC) Change in Feedlot Runoff

In most of the cases, after electrocoagulation, the pHs of the electrocoagulation solution were increased except for 10 V Al-Fe electrodes and 5 V Al-Al electrodes (Fig. 2). The increased pH in electrocoagulation solution was likely due to the excess hydroxyl ions produced at the cathode and liberation of free OH⁻ (Dalvand *et al.*, 2011; Feng *et al.*, 2007). In this study, following electrocoagulation, the pHs of the electrocoagulation solution increased except for 10 V Al-Fe electrodes and 5 V Al-Al electrodes. Iron electrodes (Fe-Fe) treated runoff had the highest pH, whereas Fe-Al treated runoff had the lowest pH. Aluminum electrodes (Al-Al) treated runoff resulted pH in between these two types of electrodes. It is also evident from Fig. 2 that the electrocoagulation had effects on pH change and almost all cases pH levels were increased due to electrocoagulation.

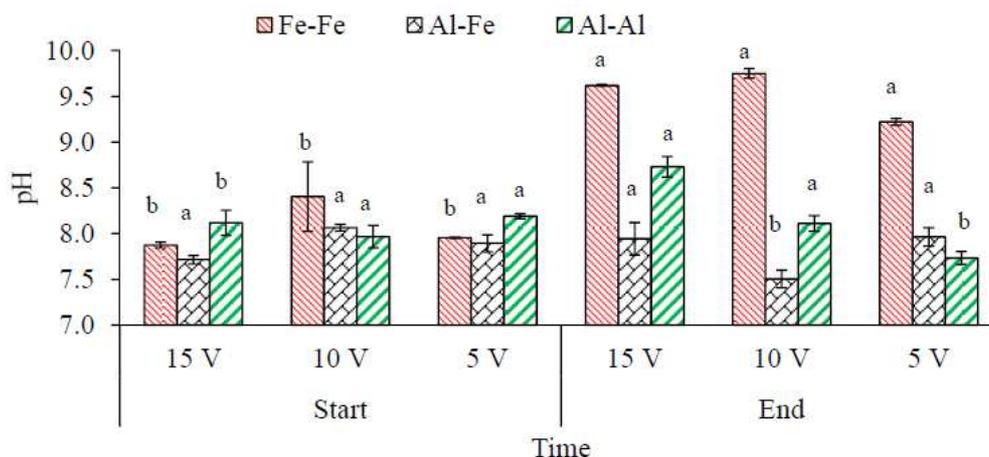


Fig. 2. pH of the feedlot runoff at start and end of electrocoagulation by different electrodes. The bars with the same letter between start (0 min) and end time (30 min) for an electrode at an applied electrode potential are not significantly different at $p \leq 0.05$

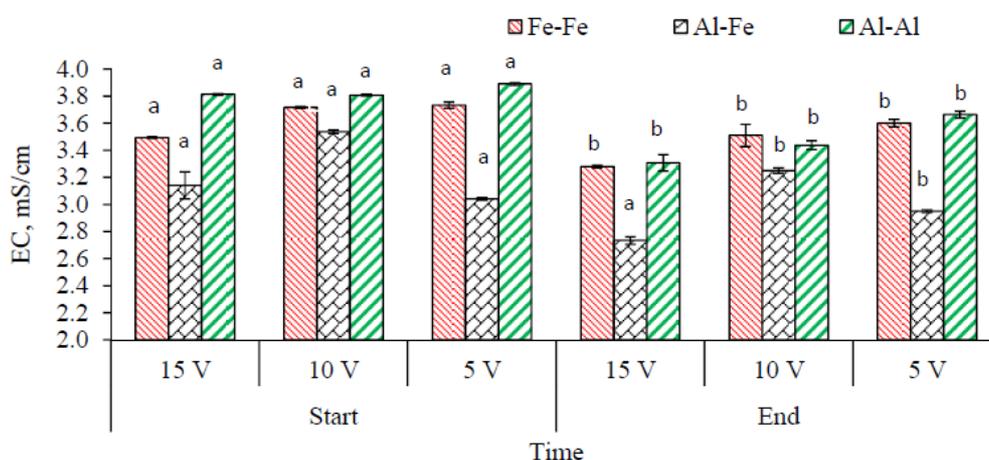


Fig. 3. Effect of electrode types and voltage potentials on Electrical Conductivity (EC) values while electro-coagulating feedlot runoff. The bars with the same letter between start (0 min) and end time (30 min) for an electrode at an applied electrode potential are not significantly different at $p \leq 0.05$

This study demonstrated that the EC of the wastewater samples were decreased significantly following electrocoagulation (Fig. 3). The highest EC reductions were observed at an applied voltage of 15 V (3.49 to 3.28 mS cm^{-1} for Fe-Fe, 3.81 to 3.31 for Al-Al electrodes and 3.14 to 2.73 mS cm^{-1} for Al-Fe electrodes), followed by 10 V (3.72 to 3.51 for Fe-Fe, 3.54 to 3.25 mS cm^{-1} for Al-Fe, 3.81 to 3.44 mS cm^{-1} for Al-Al electrodes) and 5 V (3.73 to 3.60 mS cm^{-1} for Fe-Fe, 3.04 to 2.95 mS cm^{-1} for Al-Fe and 3.89 to 3.66 mS cm^{-1} for Al-Al electrodes).

This study demonstrated that the EC of the wastewater samples was decreased significantly following electrocoagulation. The Al-Fe electrodes at 15 V resulted in the highest EC reduction than the Fe-Fe and Al-Al electrodes during 30 min of

electrocoagulation time. On the contrary, with 10 V and 5 V applied electrical potentials and 30 min treatment time; Al-Al electrodes reduced more EC than Fe-Fe and Al-Fe electrodes. The changes in EC during electrocoagulation were likely to occur by the free ions present in the solution.

In practice, after electrocoagulation, the electrostatic charge of dispersed particles present in the solution are neutralized and thus the EC of the solution is reduced (Kılıç and Hoşten, 2010). Tchamango *et al.* (2010) also mention that by means of electrocoagulation process EC could be decreased due to the consumption of protons by transformation of phosphoric acid into solid metal phosphate. This indicates that the unwanted ions were settled down by the formation of insoluble product or neutralized by charged metal ions during electrocoagulation process,

which helps to purify the wastewater during the wastewater treatment process.

Total Phosphorus (TP) Reduction

In this experiment, TP reduction was 100% within 30 min of treatment time by all electrodes combinations and at all three applied electrical potential levels. At 15, 10 and 5 V applied voltage potentials, approximately 100% TP reduction was achieved within 3 to 5 min, 3 to 10 min and 8 to 10 min, respectively, irrespective of electrode types and combination (Fig. 4a to c). The Al-Al and Fe-Fe electrodes reduced TP concentration significantly within 3 min of treatment initiation than Al-Fe electrodes for an applied potential of 15 V (Fig. 4a) and 10 V (Fig. 4b). However, Al-Fe electrode took 8 min for an applied potential of 5 V (Fig. 4c).

Overall, Al-Al electrodes had shown better TP removal than other electrodes under test conditions (Fig. 4). According to others (Dinh-Duc *et al.*, 2014; Ilhan *et al.*, 2008; Inan and Alaydin, 2014; Laridi *et al.*, 2005), the TP reductions were mainly due to the production of Al or Fe ions in an anode. The OH⁻ produced in the cathode is immediately react with metal ions in the runoff and produce metallic hydroxides. Subsequently, this process initiates polymerization reactions when metallic hydroxide particles reached a sufficient concentration and react with phosphate ions present in the solution and formed either aluminum or iron phosphate and sedimentated in the solution and helps to reduce total phosphorus from the wastewater (Dinh-Duc *et al.*, 2014; Ilhan *et al.*, 2008; Inan and Alaydin, 2014; Laridi *et al.*, 2005).

Total Nitrogen (TN) Reduction

Total Nitrogen (TN) concentration at different treatment times during an electrocoagulation process for an applied voltage is presented in Fig. 5a to c. For all electrodes tested in this study, the highest TN reduction occurred at 15 V electrical potentials (Fig. 7) when compared with 10 V (Fig. 5b) and 5 V (Fig. 5c) applied electrode potentials. At 15 V applied electrical potential and 30 min treatment time, TN reduction was approximately 63, 56 and 41% for Al-Fe, Al-Al and Fe-Fe electrodes, respectively (Fig. 5a). Similarly, at 10 V potential and 30 min treatment, the TN reductions were approximately 47, 42 and 38% for Al-Al, Al-Fe and Fe-Fe electrodes, respectively (Fig. 5b). However, at 5 V for the same treatment time, Fe-Fe electrodes resulted in the lowest TN reduction (Fig. 5c). The TN reduction at 5 V and 30 min treatment time by the Al-Al, Al-Fe and Fe-Fe electrodes were about 45, 38 and 27% by, respectively (Fig. 5c).

Similarly, the highest TN reduction occurred at 15 V electrical potentials for all electrodes as compared to another electrode potential. At 15 V applied electrode potential, Al-Fe electrodes reduced significantly greater amount of TN than the Al-Al and Fe-Fe electrodes (Fig. 5a). Similarly, with 10V potential, Al-Al electrodes reduced significant amount of TN than the Al-Fe and Fe-Fe electrodes (Fig. 5b). For 5V electrode potential, Al-Fe and Al-Al electrodes reduced significant amount of TN than the Fe-Fe electro (Fig. 5c). Therefore, any of the electrode combinations may be used in reducing TN, but Al-Al and Al-Fe electrodes combination performed the best at greater applied potential (15 V). The main reason for TN reduction are denitrification, ammonia stripping, hydrogen electroflotation, electron oxidation and electrocoagulation (Kabuk *et al.*, 2014).

Chemical Oxygen Demand (COD) Change

During the electrocoagulation process, three levels of voltage potentials were applied. Among them, 15 V applied electrical potential reduced the highest percentage of COD (Fig. 6a) than those 10 V (Fig. 6b) and 5 V (Fig. 6c). With higher electrode potential, higher nutrient reductions were likely due to the release of higher amount of metal ions to the runoff wastewater, thus reducing more COD by electrocoagulation. At 15 V applied potential and at 30 min of treatment time, the COD reduction was about 78% by all the electrodes and the differences in COD reduction among the electrodes were not significant (Fig. 6a). Similarly, for the same treatment time, an applied potential of 10 V reduced COD concentration by approximately 73, 68 and 67% for Al-Al, Al-Fe and Fe-Fe electrodes, respectively, but no significant differences among electrode types in terms of COD reduction was achieved (Fig. 6b).

The highest COD reduction was also obtained at 15 V applied electrical potential than those observed at 10 V and 5 V electrical potential. The COD reduction was much lower with 5 V than those of 10 and 15 V electrical potential. However, at each level of electrical potentials, COD reduction increased with increasing treatment time. Therefore, any of the electrode combinations at 15 V electrical potential may be used to reduce COD significantly. This research showed that the removal efficiency of COD increased irrespective of electrode types with increasing applied voltage potentials (5, 10 and 15 V) and treatment times. Other researchers (Bensadok *et al.*, 2011; Inan *et al.*, 2004; Laridi *et al.*, 2005; Merzouk *et al.*, 2011; Rivera *et al.*, 2009) also reported that at a particular voltage, removal of COD increased with increased process/treatment time.

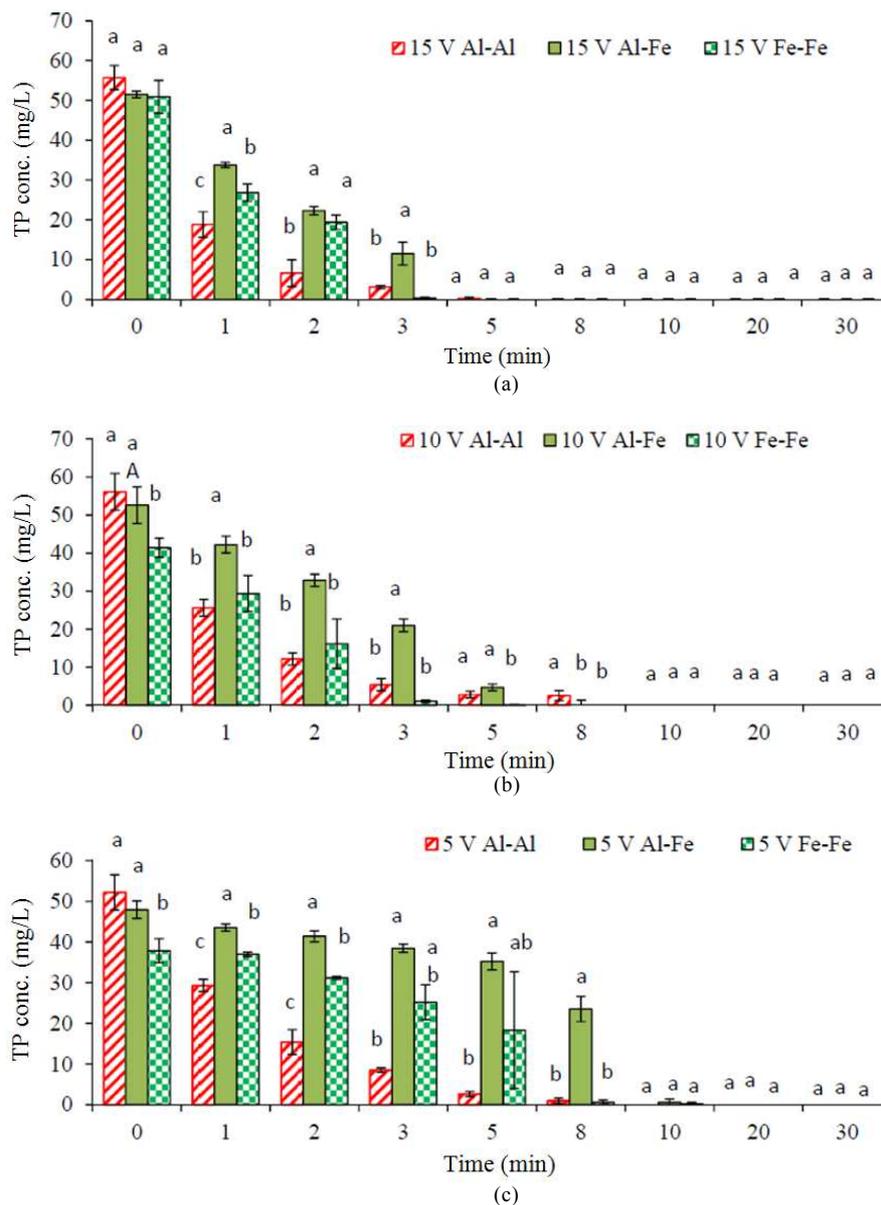
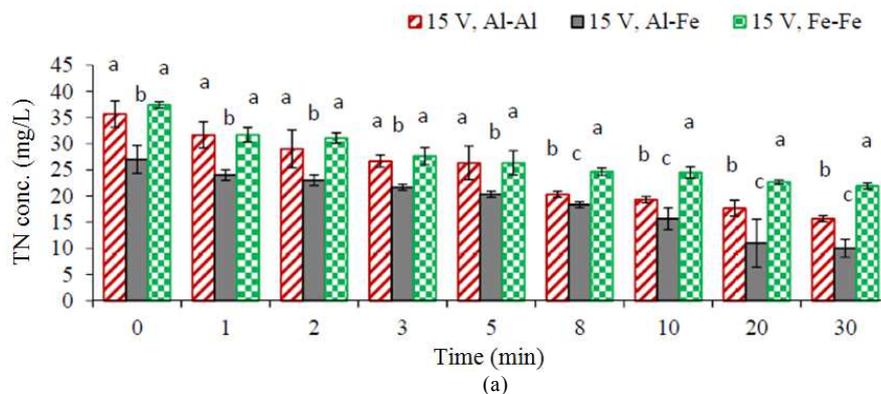


Fig. 4. Total Phosphorous (TP) values at (a) 15 V (b) 10 V and (c) 5 V applied potential by different electrodes. The bars with the same letter at each sampling time with different electrodes are not significantly different at $p \leq 0.05$



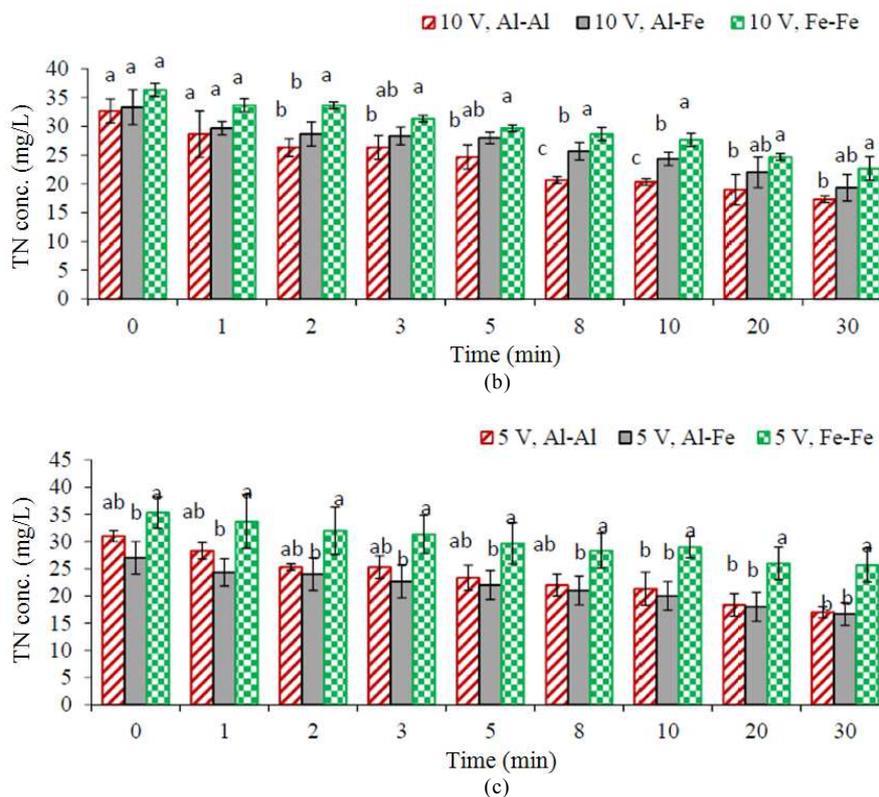
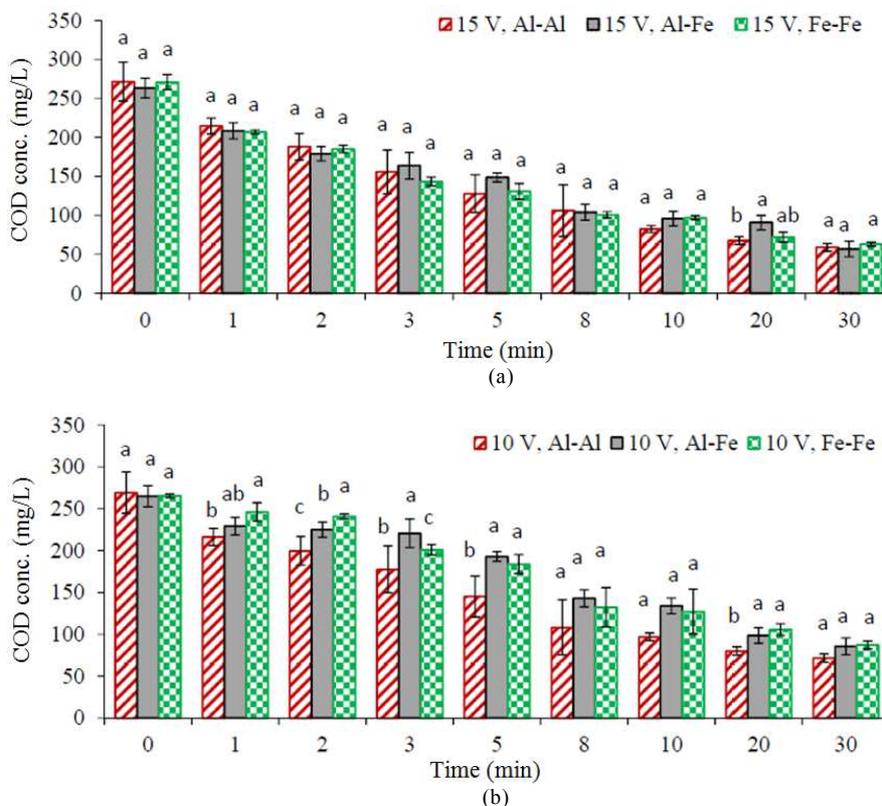


Fig. 5. Total Nitrogen (TN) values at (a) 15 V, (b) 10 V and (c) 5 V applied potential by different electrodes. The bars with the same letter at each sampling time with different electrodes are not significantly different at $p \leq 0.05$



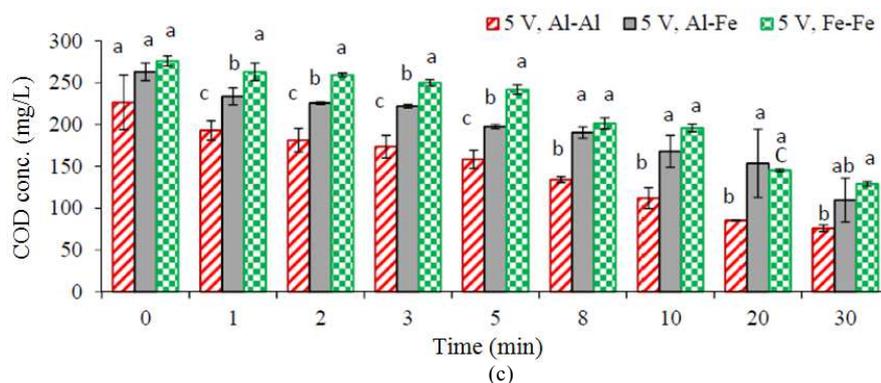


Fig. 6. Chemical Oxygen Demand (COD) values at (a) 15, (b) 10 V and (c) 5 V applied potential by different electrodes. The bars the same letter at each sampling time with different electrodes are not significantly different at $p \leq 0.05$

Table 1. Initial characteristics of feedlot runoff used in the electrocoagulation experiment

Parameter	Initial concentration
pH	8.02±0.229
Conductivity (mScm ⁻¹)	3.60±0.30
TN (mgL ⁻¹)	32.86±4.17
TP (mgL ⁻¹)	49.59±6.52
COD (mgL ⁻¹)	263.52±19.50
TS (mgL ⁻¹)	3.06±0.28

Specific Electrical Energy Consumption (SEEC)

For the same treatment time, the SEEC (Energy required per unit TP, TN or COD reduction, or per unit volume of feedlot runoff) was higher for 15 V applied potential than the 10 and 5 V applied potential (Table 2). It was also observed that treatment time decreased with increased applied voltage potential for the same amount of TP, COD and TN reduction. Therefore, at higher applied electrical potential, the treatment time of electrocoagulation can be reduced, which is preferable for designing continuous or higher capacity batch reactor for treating feedlot runoff under field condition.

All electrodes removed 100% TP at similar SEEC, except Al-Al electrodes at 15V. Overall, Fe-Fe electrodes outperformed other electrodes. The lowest SEEC per kg TP removed was estimated 7.98 for Al-AL electrodes. Similarly, the lowest SEEC per kg COD and TN removed were estimated as 4.77 and 70.89 kWh/pollutants (Table 2).

Characteristics of Sludge Generated by Different Electrode in Electrocoagulation

Elemental analysis of sludge generated by the electrocoagulation for 30 min at 15V is presented in Table 3. The aluminum residue produced by Al-Fe electrodes were 55.4, 51.1 and 37.6% less than the aluminum residue produced by Al-Al electrodes at 15, 10 and 5 V potential, respectively (10 and 5 V data are not shown). Similarly, iron residue produced by Al-Fe electrodes were 43.9, 48.5

and 63.2% lower than the iron residue produced by the Fe-Fe electrodes at 15, 10 and 5 V potentials, respectively ($p < 0.05$), which is significantly lower than that of Fe-Fe electrodes (10 and 5 V data are not shown). Aluminum electrodes (Al-Al) produced significantly higher Ca, Cu, K, Li, Mg, Pb, S, Ti and V residue than the Al-Fe and Fe-Fe electrodes. Similarly, Fe-Fe electrodes produced more Ag, As, Cd, Ce, Co, Mn, Ni and Tl residue than the Al-Fe and Al-Al electrodes.

Comparison of Percentage Reduction of TP, TN and COD during Electrocoagulation

For 30 min treatment times, the TP reduction was the highest for all electrode types followed by the COD and TN reduction for each level of applied voltage potential (Table 2). The TP reduction was about 100% in all three voltage potentials (5, 10 and 15 V) within 30 min of treatment times. Though the percentage reduction of TN and COD increased with the increasing applied voltage potential levels, it did not reach to 100% under test conditions (30 min).

The higher TP reduction was likely due to the formation of an abundant amount of insoluble metal phosphate when the OH⁻ released from the cathode react with the soluble phosphate ions already contained in the feedlot runoff during electrocoagulation process according to equations 8-12 (Dinh-Duc *et al.*, 2014; Inan and Alaydin, 2014). Though the reduction of COD was greater than TN, it was lower than TP. The average COD reduction was >60% and the main reason of COD reduction was the electrolytic oxidation of organic and inorganic carbon present in the feedlot runoff. The higher percentage of COD reduction than the TN could be due to the presence of simple oxidizable carbon compound in the form of suspended solids and liquids and oxidizing these compounds during the electrocoagulation process (Moreno-Casillas *et al.*, 2007; Yun *et al.*, 2014). TN reduction was lower (<60%) and lower TN reduction rate than COD could be due to lower

denitrification and ammonia stripping process occurred during the electrocoagulation process (Emamjomeh and Sivakumar, 2009; Ilhan *et al.*, 2008; Yun *et al.*, 2014).

Higher electric potential performed better, but it also required higher SEEC (Energy required per unit TP, TN or COD reduction, or per unit volume of feedlot runoff). In general, TN showed the highest SEEC per kg removed, followed by COD and TP (Table 2). Both Al-Al and Al-Fe electrodes performed equally, removed 100% TP at similar SEEC and outperformed Fe-Fe electrodes. As mentioned before, the lowest SEEC per

kg TP removed was estimated 7.98 for Al-Al electrodes. Similarly, the lowest SEEC per kg COD and TN removed were estimated at 4.77 and 70.89 kWh/pollutants for Al-Fe electrode (Table 2). In this research, aluminum based electrodes (Al-Al and Al-Fe) removed more TP and COD compared to Fe-Fe electrodes. This was likely due to excess active ionization when aluminum ion combined with the hydroxyl ion (OH-) and contributed to the generation of higher amounts of aluminum hydroxide (Al(OH)₃) (Hong *et al.*, 2013; Lindsay *et al.*, 1996; Rahman and Borhan, 2014).

Table 2. Comparison of removal efficiencies, Specific Electrical Energy Consumptions (SEEC) at three applied electrical potentials for 30 min electrocoagulation time

Electrical potential	Current density (A/cm ²)	Percent change		Percentage reduction			Energy (kWh/m ³)	SEEC (KWh /kg pollutant removed)		
		EC	pH	TP	COD	TN		TP	COD	TN
Al-Al electrode										
15 V	0.036	13.28	-7.55*	100	78.28	56.07	26.02	62.26	61.19	650.60
10 V	0.020	9.72	-1.79*	100	73.39	46.93	9.85	29.24	24.92	321.20
5 V	0.010	5.82	5.57	100	66.47	45.16	2.50	7.98	8.30	89.28
Fe-Al electrode										
15 V	0.023	18.97	-2.98*	100	78.48	62.96	16.39	26.53	39.66	482.20
10 V	0.017	8.10	6.94	100	67.67	42.00	8.27	20.96	23.06	295.40
5 V	0.006	2.96	-0.88*	100	58.30	38.27	1.46	10.18	4.77	70.89
Fe-Fe electrode										
15 V	0.032	6.10	-22.18*	100	76.75	41.44	22.99	37.65	55.28	741.80
10 V	0.022	5.56	-16.02*	100	67.12	37.61	10.37	33.39	43.61	379.40
5 V	0.010	3.57	-15.92*	100	53.26	27.35	2.30	20.25	23.47	118.90

*- sign indicate the increase in value than the initial

Table 3. Concentration of the elements (mgL⁻¹) in sludge following the electrocoagulation of runoff wastewater

Elements	Al-Fe 15 V	Fe-Fe 15 V	Al-Al 15 V
Ag	1.14a*±0.09	2.30a±0.12	2.34a±0
Al	87313b±6199	822c±379	195703a±11462
As	64.47a±2.57	120.97a±2.72	129.87a±0
B	10.53c±2.81	17.96b±0.99	23.28a±1.45
Ba	14.65a±1.77	18.05a±2.91	7.13b±2.02
Ca	10597b±1113	13537a±832	14698a±113
Cd	5.35b±0.31	8.06a±1.28	2.69c±0.10
Ce	12.09a±0.37	23.47a±0.85	21.01a±0
Co	7.41b±0.03	12.89a±0.43	2.14c±0.10
Cu	47.02ab±11.44	33.98b±8.22	62.49a±11.20
Fe	107299a±64014	191328b±545	1214c±237
K	4439b±51	4195b±633	6532a±368
Li	28.41a±8.14	2.85b±0.40	28.59a±1.07
Mg	20228a±599	11622b±833	20197a±656
Mn	327b±13	517a±11	149c±9
Na	1237b±55	1231b±239	1869a±110
Ni	44.13b±1.08	71.92a±5.23	13.29c±1.35
P	4829a±362	3583b±310	3316b±260
Pb	76.47b±6.56	-22.03c±0.36	169.85a±9.59
S	13707a±1157	5585b±939	15048a±632
Si	72.98a±11.10	71.57a±24.37	83.84a±10.38
Sn	3.27a±0.73	3.70a±0.22	4.08a±0.08
Ti	9.66a±2.01	7.52a±1.59	11.24a±1.43
Tl	6.35b±2.33	10.62a±0.83	4.63b±1.27
V	15.57b±2.02	13.20b±0.73	19.55a±1.61
Zn	536.48a±564.37	406.15a±554.16	329.46a±217.09

*Averages within a row followed by different letters are significantly different at p<0.05 according to Duncan multiple range tests

Elemental analysis of sludge revealed that except iron residue produced by the Fe-Fe electrodes and aluminum residue produced by Al-Al electrodes and both residue presented for hybrid electrode, rest of elemental residue presented in sludge was due to the sedimentation of metal residue in a sludge during an electrocoagulation process.

From this batch experiment, it can be concluded that electrocoagulation system may be used with greater implication than other methods such as membrane process or advance oxidation process. For the practical implication, the electrocoagulation process should be used in the continuous mode, which is viable for the livestock's grower because it consumed little amount of electrical energy and requires easily available Fe-Fe or Al-Al electrodes for the feedlot runoff treatment than the membrane treatment process or other advance wastewater treatment equipment. Also, precipitation of phosphorus from runoff or wastewater stream through the electrocoagulation process may be used as bio-fertilizer. This process, also removed the metal ions due to the sedimentation of soluble metal ion from the feedlot runoff, thus minimizes surface water quality concerns and soil quality concerns. Therefore, this phenomenon clearly indicated electrocoagulation process may be implemented in runoff containing pond or manure storage pond to reduce pollutants from feedlot runoff.

Conclusion

The comparative performances of three electrodes (Al-Al, Fe-Fe and Al-Fe) in treating feedlot runoff at varying voltage levels and treatment times were evaluated. The following conclusions can be drawn from this experiment:

- Overall, irrespective of electrode types, the percentage 358 TP reduction was the highest, followed by COD and TN
- The reduction efficiencies of nutrients (TP, COD and TN) were positively correlated with the increasing treatment times and the applied electrical potential and the energy consumption for each electrode material
- Aluminum electrodes were more effective than the other two electrodes (Al-Fe and Fe-Fe) for reducing high TP (100%) and they also consumed lowest specific energy
- Hybrid (Al-Fe) electrodes outperformed Al-Al and Fe-Fe electrodes in terms of specific energy consumptions per kg of COD removed
- Electrocoagulation process demonstrates significant amounts of metal elements in the sludge that indicates the soluble metal ion are settled down from the feedlot runoff after

electrocoagulation and it helps to improve wastewater quality after treatment process

- For the practical implication, the electrocoagulation process may be used at the runoff collection pond or settling basin in a continuous mode, since it consumed little electrical energy per kg of pollutant removed. Also, electrocoagulation process requires low maintenance

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Author's Contributions

Arjun Thapa: Former graduate student, who did his M.S.c under Dr. Shafiqur Rahman's supervision. He conducted the experiment, collected and analyzed data. Wrote the manuscript.

Shafiqur Rahman: Supervised the first author. Designed the experiment and helped student in interpreting the data. Reviewed and corrected the manuscript before submitting for publication.

Md Saidul Borhan: Assisted the graduate student in collecting and analyzing data. Reviewed the manuscript.

Ethics

This manuscript has neither been published, nor under consideration for publication or in press elsewhere concurrently.

References

- Ali, E. and Z. Yaakob, 2012. Electrocoagulation for treatment of industrial effluents and hydrogen production. INTECH Open Sci. Open Minds.
- Ansari, A.A., S.S. Gill and F.A. Khan, 2011. Eutrophication: Threat to Aquatic Ecosystems. In: Eutrophication: Causes, Consequences and Control, Ansari, A.A., S.S. Gill, G.R. Lanza and W. Rast (Eds.), Springer, pp: 143-170.
- APHA, 2005. Standard Methods for Examination of Water Wastewater. 2nd Edn., American Public Health Association, Washington, D.C.
- Basha, C.A., S.J. Selvi, E. Ramasamy and S. Chellammal, 2008. Removal of arsenic and sulphate from the copper smelting industrial effluent. Chem. Eng. J., 141: 89-98.
DOI: 10.1016/j.cej.2007.10.027

- Bazrafshan, E., F.K. Mostafapour, M. Farzadkia, K.A. Ownagh and A.H. Mahvi, 2012. Slaughterhouse wastewater treatment by combined chemical coagulation and electrocoagulation process. *Plos One*, 7: e40108-e40108.
DOI: 10.1371/journal.pone.0040108
- Bejan, D., L.M. Rabson and N.J. Bunce, 2007. Electrochemical deodorization and disinfection of hog manure. *Canad. J. Chem. Eng.*, 85: 929-935.
DOI: 10.1002/cjce.5450850615
- Bensadok, K., N. El Hanafi and F. Lapique, 2011. Electrochemical treatment of dairy effluent using combined Al and Ti/Pt electrodes system. *Desalination*, 280: 244-251.
DOI: 10.1016/j.desal.2011.07.006
- Bridle, T. and S. Skrypski-Mantele, 2000. Assessment of sludge reuse options: A life-cycle approach. *Water Sci. Technol.*, 41: 131-135.
- Butler, E., Y. Hung, R.Y. Yeh and M.S. Al Ahmad, 2011. Electrocoagulation in wastewater treatment. *Water*, 3: 495-525. DOI: 10.3390/w3020495
- Chaturvedi, S., 2013. Electrocoagulation: A novel wastewater treatment method. *Int. J. Modern Eng. Res.*, 3: 99-100.
- Cho, J., J. Lee and C. Ra, 2010. Effects of electric voltage and sodium chloride level on electrolysis of swine wastewater. *J. Hazardous Mater.*, 180: 535-541.
DOI: 10.1016/j.jhazmat.2010.04.067
- Crane, S.R., J.A. Moore, M.E. Grismer and J.R. Miner, 1983. Bacterial pollution from agricultural sources: A review. *Trans. ASABE*, 26: 858-866.
DOI: 10.13031/2013.34036
- Crites, R.W., E.J. Middlebrooks and R.K. Bastian, 2014. *Natural Wastewater Treatment Systems*. 2nd Edn., CRC Press, ISBN-10: 1466583266, pp: 549.
- Dalvand, A., M. Gholami, A. Joneidi and N.M. Mahmoodi, 2011. Dye removal, energy consumption and operating cost of electrocoagulation of textile wastewater as a clean process. *CLEAN-Soil Air Water*, 39: 665-672.
DOI: 10.1002/clen.201000233
- Dillaha, T.A., R.B. Reneau, S. Mostaghimi and D. Lee, 1989. Vegetative filter strips for agricultural nonpoint source pollution control. *Trans. Asae*, 32: 513-519. DOI: 10.13031/2013.31033
- Dinh-Duc, N., S.D. Kim and Y.S. Yoon, 2014. Enhanced phosphorus and COD removals for retrofit of existing sewage treatment by electrocoagulation process with cylindrical aluminum electrodes. *Desalination. Water Treatment*, 52: 2388-2399.
DOI: 10.1080/19443994.2013.794707
- El-Shazly, A.H., A.A. Al-Zahrani and Y.A. Alhamed, 2013. Kinetics and performance analysis of batch electrocoagulation unit used for the removal of a mixture of phosphate and nitrate ions from industrial effluents. *Int. J. Electrochem. Sci.*, 8: 3176-3185.
- Emamjomeh, M.M. and M. Sivakumar, 2009. Denitrification using a monopolar Electrocoagulation/Flotation (ECF) process. *J. Environ. Manage.*, 91: 516-522.
DOI: 10.1016/j.jenvman.2009.09.020
- Feng, J.W., Y.B. Sun, Z. Zheng, J.b. Zhang and S. Li *et al.*, 2007. Treatment of tannery wastewater by electrocoagulation. *J. Environ. Sci.*, 19: 1409-1415.
DOI: 10.1016/S1001-0742(07)60230-7
- Gaber, S.E., M.S. Rizk and M.M. Yehia, 2011. Extraction of certain heavy metals from sewage sludge using different types of acids. *Biokemistri*, 23: 41-48.
- Hong, K.H., D. Chang, H.S. Bae, Y. Sunwoo and J.H. Kim *et al.*, 2013. Electrolytic removal of phosphorus in wastewater with noble electrode under the conditions of low current and constant voltage. *Int. J. Electrochem. Sci.*, 8: 8557-8571.
- Hribar, C. and M. Schultz, 2010. Understanding concentrated animal feeding operations and their impact on communities. National Association of Local Boards of Health.
- Ilhan, F., U. Kurt, O. Apaydin and M.T. Gonullu, 2008. Treatment of leachate by electrocoagulation using aluminum and iron electrodes. *J. Hazardous Mater.*, 154: 381-389. DOI: 10.1016/j.jhazmat.2007.10.035
- Inan, H. and E. Alaydin, 2014. Phosphate and nitrogen removal by iron produced in electrocoagulation reactor. *Desalination. Water Treatment*, 52: 1396-1403.
DOI: 10.1080/19443994.2013.787950
- Inan, H., A. Dimoglo, H. Simsek and A. Karpuzcu, 2004. Olive oil mill wastewater treatment by means of electro-coagulation. *Separat. Purificat. Technol.*, 36: 23-31. DOI: 10.1016/S1383-5866(03)00148-5
- Kabuk, H.A., F. Ilhan, Y. Avsar, U. Kurt and O. Apaydin *et al.*, 2014. Investigation of leachate treatment with electrocoagulation and optimization by response surface methodology. *CLEAN-Soil Air Water*, 42: 571-577. DOI: 10.1002/clen.201300086
- Kılıç, M.G. and Ç. Hoşten, 2010. A comparative study of electrocoagulation and coagulation of aqueous suspensions of kaolinite powders. *J. Hazardous Mater.*, 176: 735-740. DOI: 10.1016/j.jhazmat.2009.11.097
- Kim, D., W. Kim, C. Yun, D. Son and D. Chang *et al.*, 2013. Agro-industrial wastewater treatment by electrolysis technology. *Int. J. Electrochem. Sci.*, 8: 9835-9850.
- Laridi, R., P. Drogui, H. Benmoussa, J.F. Blais and J.C. Auclair, 2005. Removal of refractory organic compounds in liquid swine manure obtained from a biofiltration process using an electrochemical treatment. *J. Environ. Eng.*, 131: 1302-1310.
DOI: 10.1061/(ASCE)0733-9372(2005)131:9(1302)
- Lindsay, W., P. Walthall and G. Sposito, 1996. The environmental chemistry of aluminum.

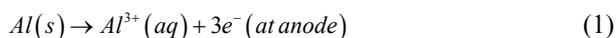
- Lucas, M.S. and J.A. Peres, 2009. Removal of COD from olive mill wastewater by Fenton's reagent: Kinetic study. *J. Hazardous Mater.*, 168: 1253-1259. DOI: 10.1016/j.jhazmat.2009.03.002
- Merzouk, B., B. Gourich, K. Madani, C. Vial and A. Sekki, 2011. Removal of a disperse red dye from synthetic wastewater by chemical coagulation and continuous electrocoagulation. A comparative study. *Desalination*, 272: 246-253. DOI: 10.1016/j.desal.2011.01.029
- Merzouk, B., B. Gourich, A. Sekki, K. Madani and C. Vial *et al.*, 2009. Studies on the decolorization of textile dye wastewater by continuous electrocoagulation process. *Chem. Eng. J.*, 149: 207-214. DOI: 10.1016/j.cej.2008.10.018
- Mollah, M.Y.A., R. Schennach, J.R. Parga and D.L. Cocke, 2001. Electrocoagulation (EC)-science and applications. *J. Hazardous Mater.*, 84: 29-41. DOI: 10.1016/S0304-3894(01)00176-5
- Moreno-Casillas, H.A., D.L. Cocke, J.A.G. Gomes, P. Morkovsky and J.R. Parga *et al.*, 2007. Electrocoagulation mechanism for COD removal. *Separat. Purificat. Technol.*, 56: 204-211. DOI: 10.1016/j.seppur.2007.01.031
- Prophet, C.W. and N.L. Edwards, 1973. Benthic macroinvertebrate community structure in a great plains stream receiving feedlot runoff. *J. Am. Water Resources Assoc.*, 9: 583-589. DOI: 10.1111/j.1752-1688.1973.tb01772.x
- Rahman, S. and M.S. Borhan, 2014. Electrolysis of swine manure effluents using three different electrodes Fe-Fe, Al-Al and Fe-Al. *Am. J. Agric. Biol. Sci.*, 9: 490-502. DOI: 10.3844/ajabssp.2014.490.502
- Rivera, M., M. Pazos and M. Sanroman, 2009. Improvement of dye electrochemical treatment by combination with ultrasound technique. *J. Chem. Technol. Biotechnol.*, 84: 1118-1124. DOI: 10.1002/jctb.2141
- Rodriguez, J., S. Stopic, G. Krause and B. Friedrich, 2007. Feasibility assessment of electrocoagulation towards a new sustainable wastewater treatment. *Environ. Sci. Pollut. Res.*, 14: 477-482. DOI: 10.1065/espr2007.05.424
- Sangal, V.K., I.M. Mishra and J.P. Kushwaha, 2013. Electrocoagulation of soluble oil wastewater: Parametric and kinetic study. *Separat. Sci. Technol.*, 48: 1062-1072. DOI: 10.1080/01496395.2012.719985
- Sano, A., M. Kanomata, H. Inoue, N. Sugiura and K.Q. Xu *et al.*, 2012. Extraction of raw sewage sludge containing iron phosphate for phosphorus recovery. *Chemosphere*, 89: 1243-1247. DOI: 10.1016/j.chemosphere.2012.07.043
- Şengil, İ.A., 2006. Treatment of dairy wastewaters by electrocoagulation using mild steel electrodes. *J. Hazardous Mater.*, 137: 1197-1205. DOI: 10.1016/j.jhazmat.2006.04.009
- Sethu, V.S., A.R. Aziz and M.K. Aroua, 2008. Recovery and reutilisation of copper from metal hydroxide sludges. *Clean Technol. Environ. Policy*, 10: 131-136. DOI: 10.1007/s10098-007-0133-4
- Spellman, F.R. and N.E. Whiting, 2010. *Environmental Management of Concentrated Animal Feeding Operations (CAFOs)*. 1st Edn., CRC Press.
- Tchamango, S., C.P. Nanseu-Njiki, E. Ngameni, D. Hadjiev and A. Darchen, 2010. Treatment of dairy effluents by electrocoagulation using aluminium electrodes. *Sci. Total Environ.*, 408: 947-952. DOI: 10.1016/j.scitotenv.2009.10.026
- Yavuz, Y., E. Ocal, A.S. Koparal and U.B. Ogutveren, 2011. Treatment of dairy industry wastewater by EC and EF processes using hybrid Fe-Al plate electrodes. *J. Chem. Technol. Biotechnol.*, 86: 964-969. DOI: 10.1002/jctb.2607
- Yi-Zhong, J., Z. Yue-Feng and L. Wei, 2002. Experimental study on micro-electrolysis technology for pharmaceutical wastewater treatment. *J. Zhej. Univ. Sci.*, 3: 401-404. DOI: 10.1631/jzus.2002.0401
- Yun, C., D. Kim, W. Kim, D. Son and D. Chang *et al.*, 2014. Application and assessment of enhanced electrolytic process for laundry wastewater treatment. *Int. J. Electrochem. Sci.*, 9: 1522-1536.

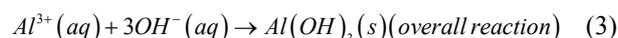
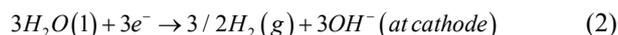
Supplementary Information

Electrocoagulation Mechanism

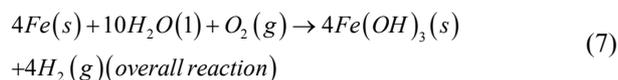
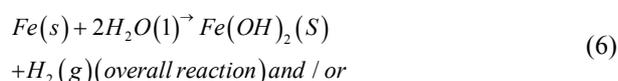
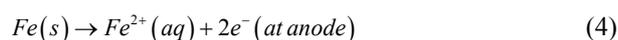
Anode generates the coagulant in situ by electrolytic oxidation during electrocoagulation. Charged ionic species are removed from wastewater by allowing it to react with either (i) ions having opposite charge, or (ii) flocs of metallic hydroxides generated within the effluent (Mollah *et al.*, 2001). Electro-flotation, electro-oxidation and electro-coagulation occur simultaneously and particles aggregate by following at least one or a combination of any of the above-mentioned mechanisms. Flocculation can enhance by continuous mixing. In this study, three different types of electrodes were used. Some of the reaction mechanisms that take place in the anode and cathode were presented below (El-Shazly *et al.*, 2013; Lucas and Peres, 2009; Sangal *et al.*, 2013).

At the Aluminum (Al-Al) electrode:





The aluminum hydroxide flocs have large surface area, are adsorbed, trapped or polymerized colloidal particles and can be removed from the aqueous solution. Aluminum hydroxide is also an important adsorbent of organic and inorganic ions, molecules and colloidal particles (Rodriguez *et al.*, 2007). At the Fe-Fe electrode:

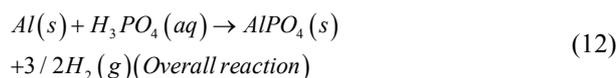
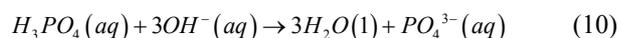
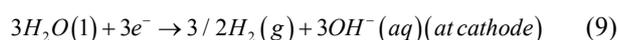
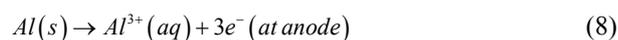


The liberated Fe^{2+} and OH^- react with various monomeric or polymeric iron hydrolyzed species and adsorb pollutants present in the wastewater to form bigger size flocs that settled down.

Pollutant Reduction Mechanism Due to E Electrocoagulation Process

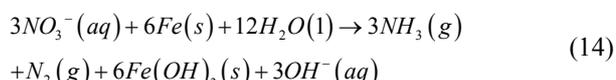
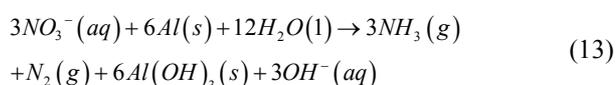
Total Phosphorus (TP) Reduction Mechanism

During electrocoagulation, OH^- liberated from the cathode reacts with soluble phosphate-containing material and liberate PO_4^{3-} . The PO_4^{3-} reacts with the metal ions and produced metal phosphates such as $AlPO_4$ or $FePO_4$ by covalent bonds. The $AlPO_4$ or $FePO_4$ are insoluble in water and settle to the bottom. During the process of settlement, these ions thereafter the micro colloidal particles absorb micro-colloidal particles. Thus, the absorbed micro-colloidal particles form flocs, which is settling down to the bottom and decreasing the amount of TP from the effluent or runoff:

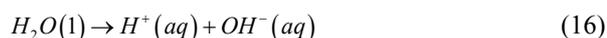
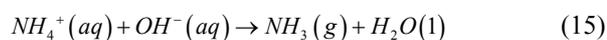


Total Nitrogen (TN) Reduction Mechanism

According to the EPA, TN is the sum of organic nitrogen, NH_4-N and NO_2-N+NO_3-N . Therefore, a TN reduction during electrocoagulation is the reduction of organic nitrogen, NH_4-N and NO_2-N+NO_3-N . The NO_2-N+NO_3-N present in the runoff is reduced by a chemical denitrification process with the help of metal electrodes such as aluminum or iron during the electrocoagulation process. The overall denitrification process during an electrocoagulation process is given below (Emamjomeh and Sivakumar, 2009):

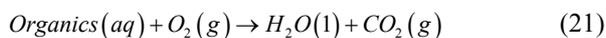
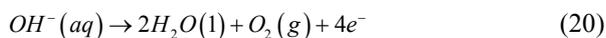
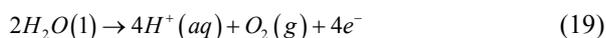


The NO_2-N+NO_3-N present in the runoff or wastewater is removed in the form of nitrogen and ammonia from the wastewater during electrocoagulation by denitrification (Equation 13 and 14). Similarly, hydrogen electro-flotation process occurs at the cathode and NH_4-N present in the wastewater or runoff removes by an ammonia stripping method. Ammonia is also removed by electro-oxidation at the anode (Kabuk *et al.*, 2014). The ammonia stripping method is enhanced by high pH (>8), high temperature (>50°C) and, high airflow rate (Emamjomeh and Sivakumar, 2009; Ilhan *et al.*, 2008). The overall reaction during the ammonia stripping method is as equation 15. According to Yun *et al.* (2014) the organic nitrogen present in the solution is also removed during the electrocoagulation process is as Equation 16 to 18. Therefore, in electrocoagulation, a different process reduces all types of nitrogen and its effect is reflected significantly in TN reduction:



Chemical Oxygen Demand (COD) Reduction Mechanism

Chemical Oxygen Demand (COD) is the amount of the oxygen needed for the chemical oxidation of inorganic and organic matter present in wastewater. Compounds that contribute to COD are biodegradable organic compounds, non-biodegradable compounds and inorganic oxidizable compounds (Moreno-Casillas *et al.*, 2007). According to Yun *et al.* (2014), the reduction of organic compounds occurred by the electrolytic oxidation and electrocoagulation process. During electrolytic oxidation, organic compounds convert into carbon dioxide gas by complete oxidation processes (Equation 21):



Organic or inorganic compounds present in feedlot runoff react with the metal hydroxide and produce an insoluble compound (Equation 16 to 18). The COD reduction is also promoted by the upward flow of hydrogen gas produced during the electrocoagulation process at the cathode. The soluble COD compound formed during electrocoagulation does not help with COD reduction (Ali and Yaakob, 2012; Moreno-Casillas *et al.*, 2007).