Original Research Article

ELECTROCOAGULATION AND ELECTROOXIDATION OF AUTOMOBILE INDUSTRY WASTEWATER

ABSTRACT

Electrocoagulation and electrooxidation processes with aluminium and steel electrodes respectively have been used to improve the physicochemical properties of wastewater from an automobile industry. The physicochemical analyses of the wastewater done by standard APHA, AWWA, WEF recommended methods showed that chromium (0.714mg/l), lead (2.44mg/l) and turbidity (120 NTU) did not meet the requirements for discharge by FEPA (Federal Environmental Protection Agency of Nigeria) and cadmium was marginally met. The optimum time for electrocoagulation, 50mins, was determined by measuring the conductivity of wastewater over a 90-minute period. Since the cost of electrocoagulation depends on the current density, the effect of current density on the concentration of pollutants (metals), COD (Chemical oxygen demand) and BOD_5 (Five-day Biochemical oxygen demand) has been verified. On the average, the current density of 25.86mA/cm^2 was effective in substantially reducing the concentration of lead by 96.36% (0.089mg/l), chromium by 88.66% (0.081mg/l) thereby making the wastewater meet the FEPA standard for discharge, which are 0.12mg/l and 0.1mg/l respectively. At this current density, cadmium concentration was reduced by 87.64% (0.011mg/l) which clearly met the limit of discharge of 0.1mg/l, but the turbidity which reduced by 87.50% (15NTU) did not meet the requirement of 10NTU for discharge. Though the COD and $BOD_5$of the wastewater were within acceptable limits, electrooxidation of the wastewater at the optimal time of 80 minutes with an average current density of 18.85mA/cm^2 reduced the COD by 73.26% and the BOD_5 by 57.14%. These values were lower than what was achieved generally by electrocoagulation; COD was reduced by >90% and BOD_5 by >86%. By applying the simple process of electrocoagulation, the automobile industry should meet the regulatory requirements for discharge into water bodies or on land.

Keywords: Electrocoagulation, electrooxidation, automobile industry, wastewater, heavy metals, current density

1. INTRODUCTION

Automotive industries such as service stations and automotive manufacturing facilities use a large amount of water during manufacturing of auto spare parts, assembling, maintenance works and washing purposes. This water comes out as a sludge containing pollutants (oil and paints particles, hydrocarbons, heavy metals, grease, dye, detergents, bio-refractory organic compounds, etc) which are difficult to remove using conventional treatment methods [1]. These contaminants are potentially hazardous to the environment if the effluent water is discharged to the environment without adequate treatment.

Excessive exposure to heavy metal pollution of soil and water bodies could lead to higher levels of accumulation in plant, human and animal tissues, leading to toxic and detrimental health risks. Lead, zinc, and chromium are among the priority toxic pollutants present in automotive wastewater. Chromium is a toxic element that negatively affects plant metabolic activities, hampering crop growth and yield, and reduced vegetable and grain quality [2-3]. It can also be harmful for humans, causing skin allergies and increasing the risk of lung cancer, among other health effects reported [4]. Lead has been found to contribute to many diseases and allergies such as encephalopathy seizures, mental retardation, anemia, dermatitis, severe harm to kidneys and reproductive system in human [5-6].

Globally, the discharge of wastewater effluent to the environment has attracted more attention due to the adverse effects of contaminants in this effluent on the eco-system and various useful and practical remediation technologies such as electrocoagulation (EC), electrooxidation (EO) and their hybrid have

been employed by researchers (7-15] in the past to treat industrial and municipal wastewater effluents. The electrocoagulation process involves an electrochemical cell unit of metal electrodes that are arranged in pairs of two—anodes and cathodes and connected to a stable power supply. The metal ions produced during EC process from the anode combines with hydroxide from the electrolysis of water to generate metal hydroxide and unbiased metal hydroxide, a coagulant which agglomerates pollutants to form flocs, the hydrogen gas generated at the cathode brings the flocs to the water surface by providing further buoyancy [16]. Electro-oxidation process utilizes the oxidation reactions occurring at the anode to degrade and eliminate pollutants in wastewater. Both electrocoagulation and electrooxidation have become a rapidly expanding area of wastewater treatment because of their low energy consumption, lack of chemical use, potential recovery and reuse of treated water [17-19].

Studies have shown that Electrocoagulation and electrooxidation have been successful in removing heavy metal, COD, BOD pollutants, turbidity and other emerging pollutants from wastewater especially at higher concentrations. Chakchouk, et al [15] investigated the removal of COD, turbidity and colour from dairy wastewater using EC, EO, and hybrid of EC and EO. The results showed that EC was very effective and quick (6 min) to remove colloidal and suspended particles, but ineffective to remove COD. Similarly, EO alone was able to reduce COD by about 40% at 30min. The authors suggested that to increase the removal of COD, a hybrid of EC and EO was implemented which reduced the COD level by about 60% in 21mins. Linares-Hernandez et al [13] determined that 99% COD, 100% color, and 100% turbidity were removed by a two-step process—electrocoagulation with iron electrode, and electrooxidation with a boron-dipped diamond electrode. Wang and Chou [20] concluded that COD concentration could be reduced to values greater than 90% by electrocoagulation, below the Taiwan discharge standard of 100 mg/l, provided that the concentration of chemical mechanical polishing wastewater was below 200 mg/l NaCl, electrical potential of 20 V, and temperature of 25 degrees Celsius. Merzouk et al [21] determined that 85.5% SS, 76.2% turbidity, 88.9% BOD, 79.7% COD, and 93% colour could be removed by the combination of electrocoagulation-electroflotation after ensuring optimum conditions for 300 mg/L silica, current density of 11.55 mA/cm^2, pH of 7.6, conductivity of 2.1 mS/cm, treatment time of 10 minutes, and electrode gap of 1 cm. Heidmann and Calmano [12] treated galvanized wastewater by successfully reducing heavy metals of Cr and Cu by over 99% and 90% of Ni, as long as optimum conditions of a PH were greater than 5, 0.2 A for Fe electrodes, 1.5 A for Al electrodes, and a power consumption of 9.0 kWh/m^3. Deniel, et al [22] found that by using iron and hybrid Al/Fe electrodes for electrocoagulation, the electrodes were capable of reducing the arsenic concentration by 99%, as the current density was increased from 0.0082 to 0.0816 mA/cm^2. Petsriprasitet al [14] determined that Cu, Cr, Pb, and Zn from billet industry wastewater was reduced by 99%, at current density of 98 A/m^2, pH of 5, and 30 minutes electrolysis time. It was noticed that within 120 minutes, pH of 3, and flow rate of 55 ml/min similar results could be obtained.

One of the most significant operational parameters in electrocoagulation process is current density i.e. current per area of the electrode. The amount of electrode dissolution is directly proportional to the amount of current passed through the electrolytic solution according to Faraday's law of electrolysis [16]. To achieve the maximum removal efficiency using an electrocoagulation process at a minimal electrolytic time and operational cost, it is essential to understand the effect of current density on EC. The current density determines the coagulant dosage at the anode and the formation of hydrogen gas at the cathode. Unnecessarily high current values may negatively affect the EC efficiency as coagulant overdose can reverse the charge of the colloids and redistribute them, reducing coagulation efficiency and also reducing electrode lifetime [23][17]. Current density is also one of the most important parameters used to study EO since it affects the rate of reactions in the EO process [24]. It should be noted that increase in current density will not necessarily result in an increase in oxidation efficiency or oxidation rate. The use of higher current densities usually leads to higher operating costs due to the increase in energy consumption [24].

Innoson vehicle manufacturing industry (commonly shortened to IVM) is an indigenous automobile manufacturer headquartered in Nnewi, Anambra State, Nigeria. The industry fabricates auto-spare parts and assembles trucks, mini and luxury buses; and cars. Due to the importance of water in car manufacturing, the industry discharges enormous amount of waste water per annum to the environment. The adequate treatment of automotive wastewater is essential for safeguarding the environment and maintaining a sound public health. Since these metals are highly toxic even at low concentrations, the methods employed here to reduce their concentrations to acceptable limits should be explored.

2. MATERIALS AND METHOD

2.1 Sample collection

50 litres of wastewater sample was obtained from IVM at the discharge point after passing through all available treatment units.

2.2 Physicochemical analysis of wastewater sample

The total dissolved solids, pH, conductivity, turbidity, COD, BOD_5, heavy metals (zinc, chromium, lead and cadmium) were determined by APHA, AWWA, WEF [25] standard methods.

2.3 Determination of optimal time

The optimal time for electrocoagulation was obtained by measuring the conductivity of the wastewater while that for electrooxidation was from the COD. After subjecting the wastewater to the required process, samples were taken at intervals of 10min over a 90-minute period, and the conductivity and COD measured. The time where these parameters were lowest was chosen as the optimal time for the respective process.

2.4 The experimental procedure/set up

The electrochemical reactors consist of a cylindrical one-litre (1L) pyrex glass beaker containing 800ml of the industrial wastewater (Fig. 1). The electrocoagulation process (EC) had a pair of aluminium electrodes, each having a surface area of 11.6cm^2 (fig. 2). All the experiments were conducted at a constant temperature of 25 °C. The electrodes were set 50mm apart and connected to a current generator (TTi EL302R digital bench power supply) which supplied a constant voltage of 20V.

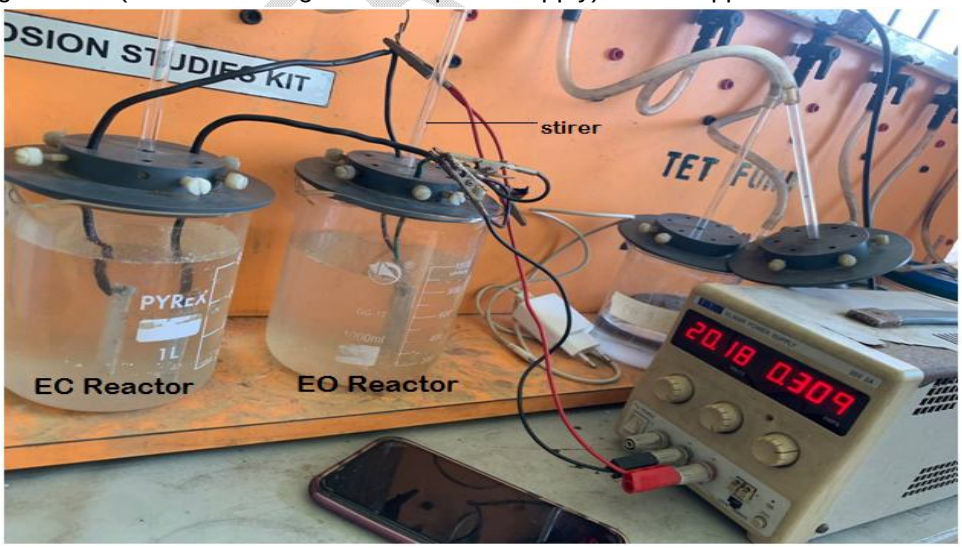

Fig.1 The electrochemical process set up.

The effect of current density on the EC process was obtained by passing a specific amount of current into the raw wastewater sample equivalent to current densities of 8.62, 17.24, 25.86, 34.48, 43.10, 51.72, 60.34, 68.97 and 77.59mA/cm^2 at the optimal time of 50min. Samples of the wastewater exposed to these current densities were tested for BOD_5, COD, TDS (total dissolved solids), heavy metals, pH, turbidity, and conductivity.

The mechanism of EC is based on generating polyvalent metal cations (Al^{3+}) directly into the wastewater by anodic dissolution of the aluminium electrodes as a result of the current imposed on the electrodes [15].

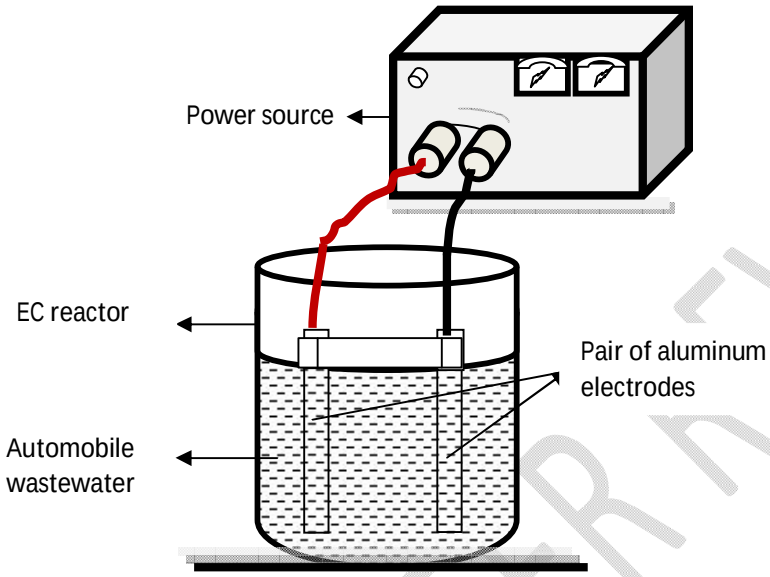

Fig. 2 schematic diagram of Electrocoagulation process

The electrochemical reactions taking place at the anode and cathode are represented by equations 1 and 2:

At the cathode:

$$3H_2O + 3e^- = {}^3/_2\ H_{2(g)} + 3OH^- \quad (1)$$

Anode:

$$AI = AI^{3+} + 3\,e^- \quad (2)$$

In the solution:

$$AI^{3+}(aq) + 3H_2O = AI(OH)_3 + 3H^+(aq) \quad (3)$$

The H_2 produced as a result of the redox reaction may remove dissolved organics or any suspended materials by flotation; this phenomenon is one of the advantages of the EC process [26].

2.5 Electrooxidation Process

The electrooxidation was carried out in batches in 1L Pyrex glass beaker containing 800ml of wastewater and a pair of cylindrical stainless steel electrodes each of surface area 10.61cm^{2}spaced 5 cm apart and connected to a current connector (TTi EL302R digital-bench power supply) supplying constant voltage of 20V (fig. 1 and 3). The effect of current density on the EO process was obtained by passing a specific amount of current equivalent to current densities of 9.43, 18.85, 28.29, 37.70, 47.13, 56.55, 65.98, 75.40 and 84.83mA/cm² at the optimal time of 80 minutes and measuring the BOD_5, COD and TDS.

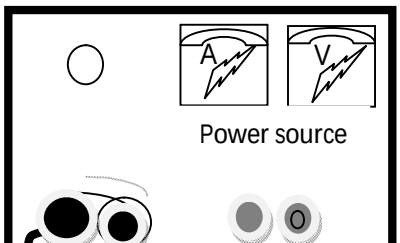

Fig. 3 Schematic diagram of electrooxidation process

3. RESULTS AND DISCUSSION

3.1 Physicochemical properties of the wastewater

Table 1 shows the characteristics of raw wastewater sample in comparison with the discharge standards. Parameters such as pH, conductivity, TDS, BOD_5, COD, cadmium and zinc metal concentrations of the wastewater met the discharge standards while the turbidity (120 NTU), lead (2.447mg/l) and chromium (0.714mg/l) concentrations in the wastewater were above the permissible limits of 10NTU, 0.2mg/l and 0.1mg/l respectively.

Table 1.The physicochemical properties of the raw wastewater effluent

Parameters	Initial concentrations in wastewater samples	National effluent discharge standards [27]
pH	6.6	6-9
Turbidity(NTU)	120	10
Conductivity (µS/cm)	303	2000
Total dissolved solids (TDS) (mg/L)	150	1500
BOD_5 (mg/l)	1.5	30
COD (mg/l)	48.8	250
Dissolved oxygen (mg/l)	3.5	4
Zinc (mg/l)	4.123	2
Cadmium (mg/l)	0.089	0.1
Lead (mg/l)	2.447	0.2
Chromium (mg/l)	0.714	0.1

3.2 Electrocoagulation

Optimal Time for Electrocoagulation

Fig. 4 shows that 50min is the optimal time for the electrocoagulation process which corresponds to the lowest conductivity value of 149µS/cm.

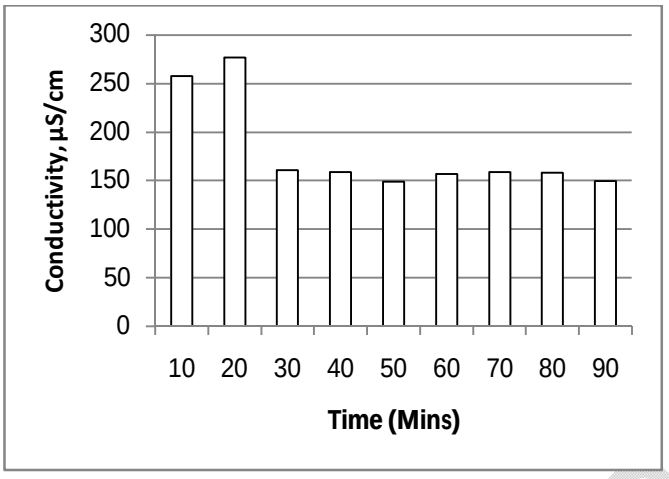

Fig. 4 Optimum Time for Electrocoagulation Measured with Conductivity

3.3 Effect of Current Density on Heavy Metals Removal

Figure 5 and table 2 shows the effect of current density on heavy metals lead, zinc, chromium and cadmium at the optimal time of 50 minutes for EC. Since increased current density increases the cost of the EC process, there is need to balance the cost of increasing the current density with the economic benefit, that is, the purification achieved. Hence 25.86mA/cm² is the recommended current density for removal of the heavy metals from the wastewater. At this condition lead is reduced by 96.36%, chromium by 88.66% and cadmium by 87.64%, with the wastewater meeting the requirements for discharge by FEPA. Even zinc which was below the permissible limit for discharge was reduced further by 65.59%.

Table 2Effect of current density on heavy metals removal

Current Density (mA/cm^2)	Zinc		Lead		Cadmium		Chromium	
	conc. (mg/l)	% removal	conc. (mg/l)	% removal	conc. (mg/l)	% removal	conc. (mg/l)	% removal
8.62	1.311	68.20	1.116	54.39	0.083	6.74	0.101	85.85
17.24	0.089	97.84	0.072	97.06	0.064	28.09	0.290	59.38
25.86	1.501	63.59	0.089	96.36	0.011	87.64	0.081	88.66
34.48	1.104	73.22	2.344	4.21	0.034	61.79	0.090	87.39
43.10	0.066	98.39	0.095	96.12	0.009	89.89	0.088	87.66
51.72	0.921	77.66	0.651	73.39	0.062	30.34	0.514	28.01
60.34	1.932	55.14	1.110	54.64	0.082	7.87	0.334	53.22
68.97	0.055	98.67	0.851	65.22	0.045	49.44	0.142	80.11
77.59	0.224	94.57	0.333	86.39	0.022	75.28	0.223	68.77

Studies by other researchers [28-31] also recorded similar percentage efficiency for heavy metals removal using an EC process.

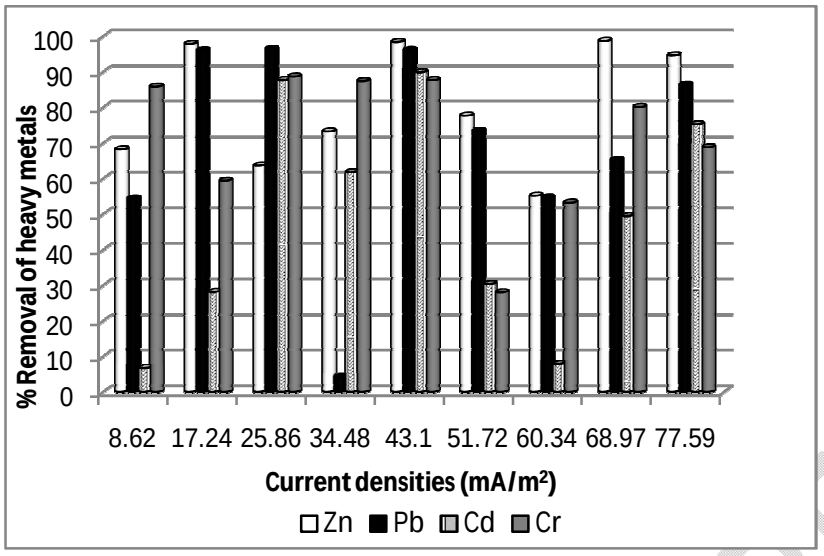

Fig. 5 Effect of current densities on heavy metal removal

3.4 Effect of current density on pH, conductivity, COD, BOD, TDS and turbidity

The concentration of organic compounds in the wastewater was assessed by the COD and BOD_5. The influence of current density on conductivity, COD, BOD_5, TDS and turbidity of the wastewater at the optimal time of 50 minutes for the EC process are shown in figure 6 and table 3. The target parameter is the turbidity, which did not meet the requirement for discharge. Wastewater turbidity is caused by the presence of colloidal species and suspended solids. At the current density of 25.86mA/cm² the turbidity was reduced by 87.50% to 15NTU which was above the limit for discharge. Since the experiment is a single-pass, it's advisable that a two-stage EC process be used to ensure complete compliance.

Table 3 Results of the effect of current density on some properties of the wastewater sample

Current Density (mA/cm^2)	pH	values after EC (mg/L)					Percentage Removal (%)				
		Cond. (µS/cm)	COD mg/l	BOD mg/l	TDS mg/l	Turb. (NTU)	Cond.	COD	BOD	TDS	Turb.
8.62	6.53	205	4.92	1.0	78	17	32.34	89.92	33.33	48	85.83
17.24	6.22	149	5.16	0.7	80	18	50.83	89.43	5333	46.67	85.00
25.86	6.36	199	4.84	0.8	83	15	34.32	90.08	46.67	44.64	87.50
34.48	6.55	185	4.76	0.5	79	17	38.94	90.25	66.67	47.33	85.83
43.10	6.69	211	4.70	0.4	80	16	30.36	90.37	73.33	46.67	86.67
51.72	6.82	213	4.60	0.5	81	14	29.70	90.57	66.67	46	88.33
60.34	6.66	201	4.75	0.7	75	17	33.66	90.27	53.33	50	85.83
68.97	6.54	216	4.80	0.3	83	14	28.71	90.16	80.00	44.67	88.33

77.59	6.88	218	4.91	0.2	85	13	28.05	89.93	86.67	43.33	89.16

*Cond. = conductivity, *turb. = turbidity

Table 3 shows that current density did not substantially affect the pH of the wastewater. It remained acidic and within the acceptable range for discharge. pH has a considerable effect on the performance of an EC process [32]. A study by Adhoum and Monser [33] showed that the highest COD and colour removal efficiencies were obtained in acidic medium, at pH values in the limits of 4.0–6.0. However, very poor removals were found either at low (<2.0) or high pH (>10). This behaviour was attributed to the amphoteric character of $Al(OH)_3$ that does not precipitate at pH less than 2.0 [13]. However, high pH value will increase Al $(OH)_3$ solubility and lead to the formation of soluble $Al(OH)_4$, which is not needed in waste water treatment.

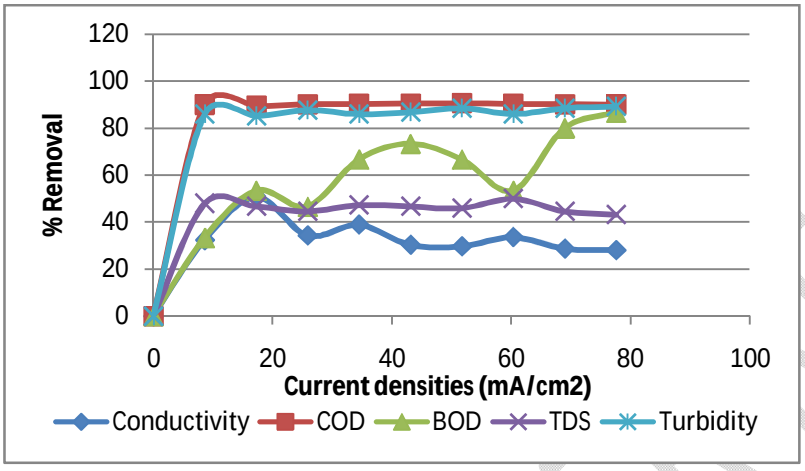

Fig 6 Effect of current density on conductivity, COD, BOD, TDS and turbidity reduction by EC

3.5 Electrooxidation Results

Optimal Time for Electrooxidation

Electrooxidationwas used to monitor the COD, BOD_5 and TDS of the wastewater. Figure 7 shows that the optimal time for the EO process was 80 minutes which corresponded to the time when the least COD of the wastewater was obtained (0.9mg/l). This result shows that reaction equilibrium for EO requires a longer time than for EC.

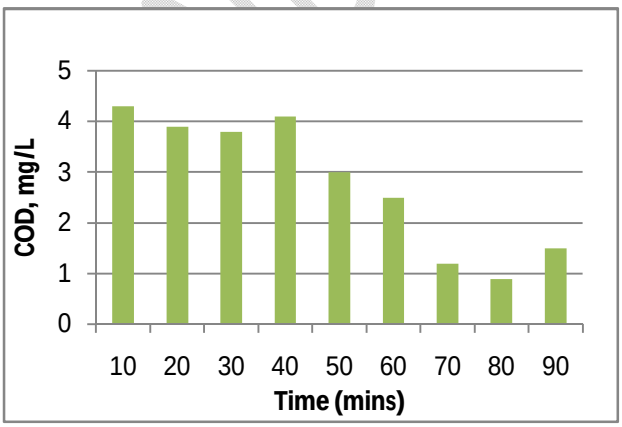

Fig. 7 Optimal time for Electrooxidation

3.6 Effect of current density on pollutants during Electrooxidation

Table 1 which has the physicochemical properties of the wastewater shows that prior to treatment the COD, BOD_5 and TDS met the requirements for discharge. From table 4, considering the values of COD and BOD the most economic current density may be 18.85mA/cm². Though electrooxidation reduced the organic content of the wastewater as shown by the reduced values of COD, BOD_5 and TDS, table 3 shows that generally EC is a more efficient and capable process for removing organics from the wastewater. Without considering the constraint of current density which is indicative of cost, EC achieved a maximum COD reduction of 90.57% and 86.67% for BOD which was higher than values from EO. Hence, EC is the preferred treatment process. TDS remediates well during EO process (68.67% maximum removal) than in EC (50% maximum removal). The result corroborates with previous studies [22][15] that EO is more efficient in removing dissolved organic pollutants in wastewater. A study by Linares Hernández, et al.[13] have also found that electrocoagulation was very effective in removing colloids and suspended particles after only 30 minutes of operation. However, it was less effective at removing COD, removing only about half from the wastewater. The author emphasized that the use of electrooxidation has a relatively good removal effect on organic matters, COD and BOD but it takes a long time to work and is not convenient. They concluded that EC is a fast but incomplete process and EO is a slow process and can improve the efficiency of treatment. This may somehow be said to agree with our findings in that the optimal time for EO is much higher than EC but the only improvement in the quality of wastewater is in the reduced TDS. This is not enough to implement EO in the automobile industry since the TDS was already within the FEPA limits.

Table 4 Effect of current densities on COD, BOD and TDS with EO

Current Density (mA/cm^2)	COD of wastewater after EO (mg/l)	BOD of wastewater after EO (mg/l)	TDS of wastewater after EO (mg/l)	Percentage removal (%)		
				COD	BOD	**TDS**
9.43	3.41	0.4	42	30.69	60.0	46.15
18.85	1.38	0.30	53	73.26	57.14	33.75
28.29	1.55	0.34	44	67.98	57.5	46.99
37.70	2.26	0.12	60	52.52	76.0	24.05
47.13	2.10	0.25	55	55.31	37.5	31.25
56.55	1.99	0.33	43	56.74	34.0	46.91
65.98	2.21	0.42	49	53.46	40.0	34.63
75.40	1.50	0.11	26	68.75	63.33	68.67
84.83	2.11	0.10	36	57.03	50.0	57.65

4. CONCLUSION

Electrocoagulation and electrooxidation of automobile industry wastewater has been done at the optimal times of 50min and 80min respectively. The electrocoagulation process was able to reduce the heavy metal pollutants lead, chromium and cadmium to permissible limits for discharge on land or into water bodies. Though electooxidation improved marginally the TDS, electrocoagulation was a more efficient and capable process for removing heavy metals lead, chromium and cadmium, as well as reducing substantially the COD and the BOD_5. Adding an electrocoagulation unit that works at the recommended process parameters will be enough to achieve environmental compliance.

Ethical approval Not applicable

Consent to participate Not applicable

Consent for publication All authors approved the final manuscript to be published.

Data availability The authors confirm that the data supporting the findings of this study are available within the article

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