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Electrocoagulation treatment of municipal wastewater in Torreon Mexico

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ABSTRACT

One of the biggest challenges of the twenty-first century is to provide water, primarily potable, to the majority of the world population. This is a critical issue, especially in developing countries. One way to augment that effort is to reuse water. Currently, some emerging wastewater treatment technologies based on electrochemistry (e.g. electrocoagulation (EC), electroflotation, electrodecantation, and electrooxidation) are available that are competitive and advantageous over conventional technologies. Although EC has been known for more than a century, it has not been comprehensively studied. EC has been commercialized for the removal of specific contaminants and wastewater treatment. This work demonstrates EC as an alternative method for the treatment of municipal wastewater in Torreon; Coah, Mexico. COD value for wastewater in Torreon is considered as medium level (245 ppm). Experiments were conducted to determine the optimum operational conditions. Results show a remarkable removal efficiency for: Chemical Oxygen Demand (COD) 77–94%; and also for coliforms 80%, and colony forming units (CFU) 99.98% within 30 s of residence time.

Keywords: Electrocoagulation; COD; CFU; Municipal wastewater

1. Introduction

Wastewater is the liquid end-product (by-product), of municipal, agricultural, and industrial activity. The chemical composition of wastewater reflects its origin. The term wastewater implies that it is a waste product to be discarded. However, the water we use (e.g. drinking, washing, bathing, etc.) ultimately ends up back in the stream, river, lake, or groundwater to be withdrawn, treated, and used again. So wastewater must be treated and/or reused in an environmentally sound manner before it is discarded [1].

Water pollutants consist of floating materials, settable solids, organic and inorganic suspended solids, organic and inorganic dissolved solids and liquids, oil, fats and grease, dissolved gases, and micro-organisms. All water pollutants have different characteristics, imposing separate threats for human health and their removal. Wastewater treatment plants are devoted to remove most of the pollutants while delivering water

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Table 1
Typical contaminant concentration of untreated domestic wastewater (after: Metcalf and Eddy (1991)

Parameter	Unit	Concentration		
		Weak	Medium	Strong
BOD_5 at 20°C	$mg\,L^{-1}$	110	220	400
Chemical Oxygen Demand (COD)	$mg\,L^{-1}$	250	500	1000
Grease	$mg\,L^{-1}$	50	100	150
Total coliform	$CFU\,100\,mL^{-1}$	10^6–10^7	10^7–10^8	10^8–10^9

with little or no risk back in the stream, river, lake, groundwater, or industrial reuse.

Conventional wastewater treatment include: screening, coagulation flocculation, sedimentation, biological treatment, filtration, and disinfection. Table 1 shows typical concentrations of some key contaminants in domestic wastewater, and the classification according to the concentration of contaminants. These parameters are often used to determine the efficiency of wastewater treatment. Removal of fecal coliforms and colony-forming unit (CFU) are especially important because communicable diseases can be transmitted by the pathogenic organisms in wastewater.

In this paper, we will show our work on the electrocoagulation treatment of municipal wastewater of Terreon, Mexico. COD values for wastewater of Torreon have been ranked as medium concentration of 245 ppm.

Biological Oxygen Demand (BOD) is defined as the amount of oxygen required by bacteria to decompose organic matter for a specified period of time (usually five days), under aerobic conditions. The amount of oxygen reported with this method represents only the carbonaceous oxygen demand (CBOD) or the easily decomposed organic matter.

BOD is commonly used to measure natural organic pollution. Its analysis technique uses a titrimetric determination of O_2 consumption due to the biological oxidation of sample.

An alternative to the BOD test for determining the oxygen consuming potential of a wastewater sample is the Chemical Oxygen Demand (COD) test.

Chemical Oxygen Demand (COD) is the amount of oxygen required to oxidize organic and oxidizable inorganic matters present in water or wastewater. Its analysis technique determines oxidant consumed during a chromic acid digestion of the sample in standard conditions. Analysis of COD is much more practical than analyses for specific pollutants. Like BOD, the units for COD are in milligrams of oxygen per liter ($mg\,L^{-1}$).

The advantage of this test is that it is quick and reproducible.

However, COD test measures nonbiodegradable organic matter, that can be oxidized by potassium dichromate, and may not completely oxidize low-molecular weight fatty acids and aromatic hydrocarbons, but may oxidize inorganic ions (e.g. chloride and nitrite for instance). These items may lead to some incompatibilities. The addition of certain catalysts during the test can eliminate most of these interferences [2].

1.1. Electrocoagulation

EC can be considered an accelerated corrosion process with green rust (GR) recognized as an important intermediate phase in corrosion of Fe(0).

Green rust (GR). GRs are layered Fe(II)–Fe(III) hydroxides having a pyroaurite-type structure consisting of alternating positively charged hydroxide and hydrated anion layers. Some Fe(II) of the octahedral sheets of $Fe(OH)_2$ is replaced by Fe(III). This results in positive layers of charge that are balanced by the inclusion of anions between the layers. Its exact nature depends on the interlayer anion. Various forms have been synthesized and studied. Results of many studies show GR conforming to a general chemical composition and stoichiometry, that can be represented with the general formula: $[Fe^{II}_{(6-x)}Fe^{III}_x(OH)_{12}]^{x+}[(A)_{x/n}\cdot yH_2O]^{x-}$, where x ranges from 0.9 to 4.2, A is an n-valent anion (typically CO_3^{2-}, Cl^- or SO_4^{2-}, and y denotes the varying amounts of interlayer water (typically y ranges from 2 to 4 for most GRs) [3].

GRs, unlike most iron oxides and their form, have great specific internal surface areas and consequently great reactivities. They are reactive ion exchangers and sorbents (Hansen et al., 2001). The amphoteric surface hydroxyl groups lead to both sorption of heavy metals (cations) and organic anions, e.g. linear alkyl benzene sulfonates (LAS), the major synthetic surfactant in laundry detergents and cleaning products, and inorganic anions, e.g. silicate, arsenate, and selenate. Polar noncharged compounds are sorbed into the interlayer. Cations (e.g. Cu(II), Ni(II), Zn(II), Cd(II), Co(II), and Mg(II)) may isomorphically substitute for Fe(II) during GR formation (co precipitation). For instance, nickelous-ferric GR, end products obtained by substitution of Fe(II) ions by Ni(II) ions in GR (Refait and Génin (1993, 1997) and Refait et al. (1994, 1998)). This incorporation of cationic inorganic contaminants into the structures of GR may provide an effective means of sequestering contaminants in the subsurface.

The reactivity of GR is responsible of the removal of cationic and anionic pollutants, by substituting atoms of Fe(II) or Fe(III) or OH^- ions [4].

Current theory of EC involves several successive stages:

(1) Generation of metal ions.

Anode:

$$Fe \rightarrow Fe^{+2} + 2e^{-1} \quad (1)$$

$$Fe \rightarrow Fe^{+3} + 3e^{-1} \quad (2)$$

(2) Hydrolysis of metal ions and generation of metal hydroxides.

$$Fe + 6H_2O \rightarrow Fe(H_2O)_4(OH)_{2(aq)} + 2H^{+1} + 2e^{-1} \quad (3)$$

$$Fe + 6H_2O \rightarrow Fe(H_2O)_3(OH)_{3(aq)} + 3H^{+1} + 3e^{-1} \quad (4)$$

(3) With iron electrodes, hydrogen evolution at the cathode.

$$2H^+ + 2e^- \rightarrow H_{2(g)} \uparrow \quad (5)$$

$$2H_2O_{(l)} + 2e^- = H_{2(g)} + 2OH^- \quad (6)$$

Electrochemistry depends on thermodynamics and kinetics. The rate of reaction depends on the dissolution of iron and the removal of $[H^+]$ via H_2 evolution. This reaction occurs fast for low pH values (strong acids) and for a weak acid the rate will depend on the p*K*a of the acid.

(4) Destabilization of the contaminants, particulate suspension, breaking of emulsions, and aggregation of the destabilized phases to form floc.

(5) Theory of EC consider that chemical reactions and precipitation can occur during the EC process or that other cation and/or the hydroxyl ion (OH^-) form a precipitate with the pollutant.

Overall reaction. In general, hydrogen gas and GR are formed at the cathode as shown in Eq. (7) [5,6]:

$$6Fe + (12 + x)H_2O \rightarrow 1/2(12 + x)H_{2(g)} \uparrow + xFe(OH)_3(6 - X)Fe(OH)_{2(s)} \quad (7)$$

(6) Formation of rust (dehydrated hydroxides) occurs as shown in the following:

$$2Fe(OH))_3 \leftrightarrow Fe_2O_3 + 3H_2O \quad \text{(hematite, maghemite)} \quad (8)$$

$$Fe(OH)_2 \leftrightarrow FeO + H_2O \quad (9)$$

$$2Fe(OH)_3 + Fe(OH)_2 \leftrightarrow Fe_3O_4 + 4H_2O \quad \text{(magnetite)} \quad (10)$$

$$Fe(OH)_3 \leftrightarrow FeO(OH) + H_2O \quad \text{(goethite, lepidocrocite)} \quad (11)$$

Hematite, maghemite, rust, magnetite, lepidocrocite, and goethite have been identified as EC by-products [7,8].

1.2. EC removal efficiency

There is an extremely high variability in organic compound removal efficiencies for organic compounds, BOD, and COD using EC. This is one reason why EC is not universally accepted as a conventional technology. Removal efficiency for COD might show negative, medium and surprising high values [9,10] with the variability depending on the compounds that contribute to COD and their reactivity with iron and/or aluminum.

For example, suspended solids in small quantities are not a problem for EC. Organic compounds reacting with iron(II) and iron(III) to form insoluble products are also removed. Organic compounds reacting with iron(II) to form insoluble compounds and with iron(III) to form soluble compounds or inversely

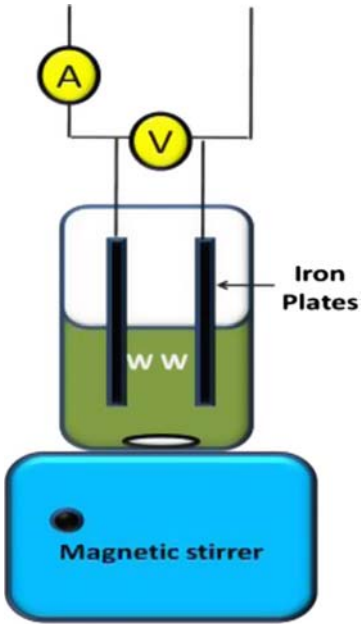

Fig. 1. Experimental setup.

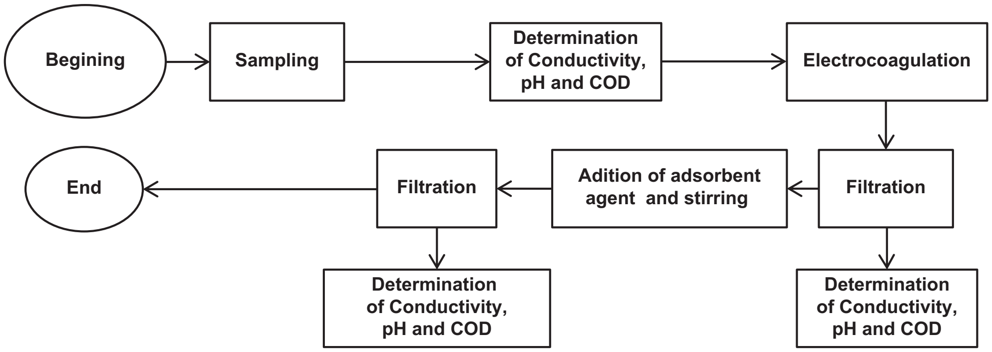

Fig. 2. Process flow diagram of the experimental tests.

will be partially removed. Organic compounds that do not react with iron(II) or iron(III) will not be affected by the EC process [4].

2. Experimental

The corresponding experiments used the following: lab materials, beakers of different capacities, activated carbon (AC) as adsorbent agent, iron electrodes of 59 × 32 × 2 mm placed 6 mm apart, Steren MUL-285 multimeters, a model ELI-055 Steren voltage converter 110 ac V to 15 dc V, an Arrow-Hart switch 82608, 0.56 kW four poles interruptor to reverse current, a LabTech Hot plate stirrer; Model LMS-1003, for filtration, 150 mm Whatman # 1 filter papers, an HI/839800 Hanna Digestor, COD Hanna vials, and an HI83099 Hanna Photometer for COD determination.

A schematic diagram of the experimental setup is shown in Fig. 1. The wastewater sample is in the beaker size rector; as electrodes, iron plates are used, and current flow is reversed every minute if required with a four pole interrupter to avoid electrode polarization and uneven plate consumption. Plates are connected to an ammeter and a voltmeter to measure current and voltage, respectively.

All samples were taken from the “Las Fuentes” collector, next to the El Campestre wastewater treatment plant in Torreon; Coahuila, Mexico, and were run as depicted in the process flow diagram as shown in Fig. 2. Samples were also treated with a water adsorption agent (WAA, 1 g/L). Three replicates were made with the average value reported.

For all samples, pH ranked from 9.07 to 10.04 and conductivity from 804 to 1057 μS.

MPN (Most Probable Number) of coliforms and CFU were determined using EPA standard methods in the Biology Faculty of the Coahuila Autonomus University, Campus Torreon.

Table 2
Parameters and results for experiment 1 with 5 min of residence time

Adsorbent used	V	A	Initial COD (mg/L)	Final COD (mg/L)	% COD removal
None	12.58	0.54	245	122	50.24
AC	12.96	0.49		81	66.93

Table 3
Parameters and results for experiment 2 with 10 min of residence time

Adsorbent used	V	A	Initial COD (mg/L)	Final COD (mg/L)	COD removal (%)
None	13.4	0.47	245	123	49.79
AC				74	69.79

3. Results and discussion

Experiment 1: This experiment was run to assess the quality of the method, on sample 1, for 5 min. Parameters and results are shown in Table 2.

Experiment 2: Five minutes residence time (experiment 1) showed good results. To test the effect of residence time, experiment 2 was run with the residence time of 10 min. Parameters and results for this test are presented in Table 3.

Results are similar to those for experiment 1, so it is advisable to run EC for 5 min, but stopping the process every minute and determining COD removal efficiency.

Experiment 3: This set of experiments was run to determine the effect of residence time on COD removal efficiency. Results can be seen in Table 4 and Fig. 3.

Table 4
Determination of the effect of residence time on COD removal (experiment 3)

Residence time t_r (min)	V	A	Adsorbent used	Initial COD (mg/L)	Final COD (mg/L)	COD removal (%)
1	13.01	0.53	None	245	144	41.22
			AC		82	66.53
2	15.03	0.46	None		141	42.44
			AC		84	65.71
3	14	0.46	None		121	50.61
			AC		98	60.00
4	13.24	0.48	None		136	44.49
			AC		104	57.55
5	13.23	0.47	None		127	48.16
			AC		86	64.89

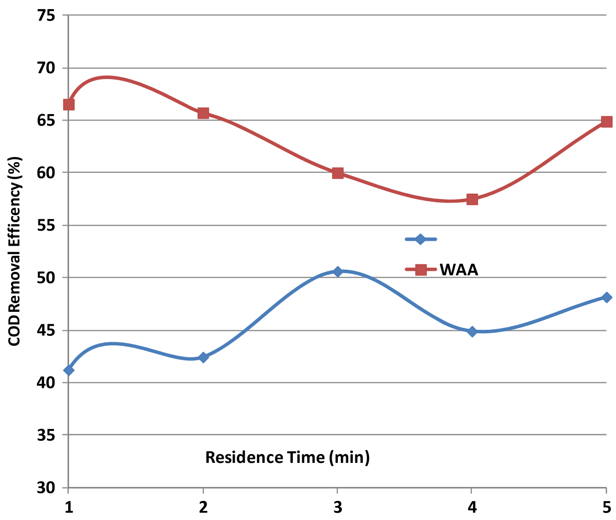

Fig. 3. Graphic of COD removal efficiency vs. residence time for experiment 3.

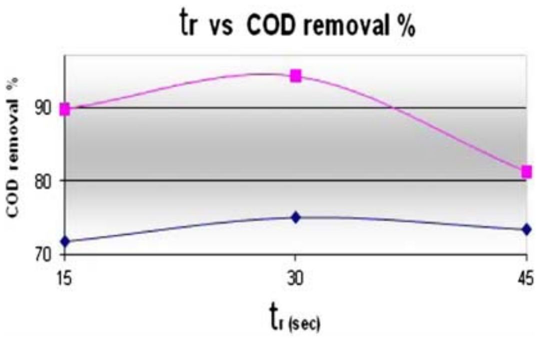

Fig. 4. COD removal efficiency (%), at different residence time, t_r (experiment 4). The square marker indicates the COD removal efficiency with adsorbent, and the diamond marker without adsorbent.

The best result with the adsorbent agent added was attained within 1 min of residence time, with little difference between trials.

Table 5
Determination of the effect of residence time on COD removal (exp. 4)

Residence time t_r (s)	V	A	Watts	Adsorbent used	Initial COD (mg/L)	Final COD (mg/L)	COD removal (%)
15	11.22	0.62	6.95	None	245	69	71.83
				AC		25	89.79
30	12.08	0.57	6.88	None		61	75.10
				AC		14	94.28
45	11.02	0.6	6.61	None		65	73.46
				AC		46	81.22

Table 6
COD removal with 30 s of residence time (experiment 5)

Experiment no.	Residence time t_r (s)	V	A	Adsorbent used	Initial COD (mg/L)	Final COD (mg/L)	COD removal (%)
5.1	30	11.18	0.58	None	352	187	46.87
				AC		80	77.27
5.2	30	10.2	0.59	None	282	122	56.73
				AC		65	76.95
5.3	30	10.2	0.59	None	342	92	73.09
				AC		51	85.08
		Average		None			58.89
				AC			79.77

Some samples turned yellowish red, which indicating the presence of excess iron and its oxidative products, which are indicators of a long residence time.

Experiment 4: To continue studying the effect of residence time on COD removal efficiency, samples were run at 15, 30, and 45 s. Results are shown in Table 5 and Fig. 4. After electrocoagulation, the reaction mixture clearly shows the formation of green rust. When it is filtered, the EC-filtrate exhibits a little color and odor. The addition of adsorbent makes the filtrate transparent indicating completion of treatment.

Less than a minute was needed to completely treat wastewater. A COD removal efficiency of 94.28% was achieved within 30 s using the adsorbent agent. With these parameters, the energy consumption is just 7 W.

A decrease in COD removal efficiency (81.22%) was achieved with an increase in residence time (45 s). This increase of COD may cause due to the increased formation of oxidizable species (such as, Fe(II)) during longer electrocoagulation run.

Experiment 5: Three additional samples were run at a residence time of 30 s and the results are presented in Table 6. Results are consistent with those obtained for biological wastewater treatment plants.

Experiment 6: It is designed to determine the removal efficiency of MPN (Most Probable Number) of coliforms in wastewater with EC. A sample was run using the residence time determined in previous experiments. Results are presented in Table 7.

Experiment 7: This examines the EC process for removal of CFU (Colony-Forming Unit), again run for

Table 7
EC parameters, initial, and final MPN (experiment 6)

Residence time t_r (s)	V	A	Adsorbent used	Coliforms MPN		Coliforms removal (%)
				Initial	Final	
30	11.64	0.56	None	>2400	1100	54.16
			WAA		480	80

Table 8
EC parameters for the removal of CFU (experiment 7)

Residence time t_r (s)	Adsorbent used	pH	CFU/ml (Colony-Forming Unit)		CFU removal (%)
			Initial	Final	
30	None	7.30	2,130,000	21,000	99.01
	WAA	7.27		400	99.98

Table 9
Comparison of COD removal for various residence times (experiment sets of 1–3 and 4–5)

Experiment	Residence time t_r (min)	COD removal (%) Adsorbent used		Experiment	Residence time t_r (s)	COD removal (%) Adsorbent agent	
		None	AC			None	AC
1	5	50.24	66.93	4	15	71.83	89.79
2	10	49.79	68.79		30	75.1	94.28
3	1	41.22	66.53		45	73.46	81.22
	2	42.44	65.71	5.1	30	73.09	85.08
	3	50.61	60.0	5.2	30	46.87	77.27
	4	44.49	57.55	5.3	30	56.73	76.95
	5	48.16	64.89				
Mean (μ)		46.71	64.06	Mean (μ)		66.18	84.10
Variance (σ^2)		15.43	14.01	Variance (σ^2)		134.88	48.61

Table 10
Brief description of NOM-003-ECOL-1997

Type of reuse	Monthly average				
	Fecal coliforms (MPN/100 ml)	Helminto eggs (e/l)	Oil and grease (mg/L)	BOD^5	TSS (mg/L)
Service to the public with direct contact	240	1	15	20	20
Service to the public with occasional or indirect contact	1000	5	15	30	30

30 s with results shown in Table 8. The attained results are excellent as the removal efficiency for CFU is almost 100%.

According to the data seen in Table 9, for a residence times between 1 and 10 min, COD removal is almost constant (64% with AC). With residence times of 15–45 s the mean value for COD removal is 84% with a variance of 48 which makes the results consistent. A longer EC-run generates more reductants (e.g. Fe(II)) making the COD values of the end solution higher. Consequently, COD removal efficiency is higher at EC time in seconds than in minutes.

4. Conclusions

It is demonstrated that using EC in municipal wastewater treatment is efficient and effective for removing COD, coliforms, and CFU. Residence times less than only a minute are required to complete the wastewater treatment, with 30 s established as an optimal residence time. COD removal is good and comparable to oxidation pond biological treatment where residence times range from three to six days. Conventional methods (e.g. biological) also generate odors and a lot of sludges. EC generates just the exact required amount of coagulant. EC wastewater treatment might substitute primary and secondary treatment in conventional treatment plants due versatility and substantially lower residence times.

For all residence times, COD removal is higher with AC.

The maximum permissible values of pollutants allowed for the official Mexican norm NOM-003-ECOL-1997, for residual treated wastewaters, for reuse in services to the public are presented in Table 10.

Comparison with an original simple coliform removal efficiency of 80% (e.g. >2400 MPN of coliformes), a minimum 480 MPN of coliforms is attained while the norm indicates a maximum value of 1000 for services with indirect or occasional contact to the public. Results comply with the regulations at less than half the maximum value allowed, and treated wastewater may be used for reuse in green areas and industrial process as well.

In brief, objectives are accomplished, and it is established that EC combined with the use of an adsorbent agent for wastewater treatment can be applied to determine parameters for optimum operation, and foundation for industrial application in a continuous process.

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