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Removal of *Escherichia coli* from Domestic Wastewater Using Electrocoagulation

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ABSTRACT

The objective of this study was to evaluate the efficiency of electrocoagulation in the removal of Escherichia coli from domestic and urban wastewaters and to determine the effects of the main operational parameters on the process. An electrocoagulation reactor with aluminum and iron electrodes was built for this purpose. A factorial design was applied, where amperage, treatment time, and pH were considered as the factors and E. coli percent removal was the response variable. After 20 min of treatment, >97% removal efficiency was achieved. The highest E. coli removal efficiency achieved was 99.9% at a neutral pH of 7, amperage of 3 A, and treatment time of 60 min. However, the removal efficiency of close to 99% was also achieved at natural wastewater pH of 8.5. The statistical analyses showed that the three tested factors significantly affected the E. coli removal percentage ($p < 0.05$). These results indicate that electrocoagulation has a high disinfection power in a primary reactor in removing water contaminants as well as simultaneously removing pathogenic microorganisms when compared to biological treatment processes. This represents an additional benefit, because it will considerably reduce the use of chlorine during the final disinfection stage.

Keywords: domestic wastewater, electrocoagulation, *escherichia coli*, electrochemical disinfection

INTRODUCTION

The primary objective of most traditional drinking and wastewater treatment plants in Peru is to remove organic matter and different contaminants. However, they are not very efficient in the disinfection process, that is, in the removal of pathogenic microorganisms. An additional disinfection process is therefore required to remove pathogens. The addition of chlorine is the most commonly used method worldwide, employed in approximately 90% of water treatment plants [Silva, 2015], owing to its simplicity, low cost, and high efficiency. Chlorine is an important biocidal agent; however, it is not properly used and, in some cases, it is overused. When added to water, chlorine, in its different forms, reacts with the naturally occurring organic substances, generating by-products, such as trihalomethanes (THMs). These compounds reportedly cause various diseases, including cancer [Silva, 2015]. The use of chlorine is prohibited in such countries as Germany, and there are various governing regulations in Spain [Guillemes, 2015]; however, it is widely used in Peru. Moreover, such chemicals become less effective over time; that is, they have a limited storage time and, as such, cannot be stored in large volumes, thereby increasing the logistics costs [Guillemes, 2015].

Novel disinfection methods that do not require chlorine, such as UV irradiation, ozonation, and electrocoagulation or electrochemical disinfection, have been studied over the last few years. Electrocoagulation generates a wide range of oxidizers, such as hydrogen peroxide and ozone, in the presence of oxygen molecules as well as free chlorine and chlorine dioxide in the presence of chloride ions [Ricordel, 2014].

Electrocoagulation involves the generation of coagulants in the sacrificial anode, usually made of aluminum or iron [Holt et al., 2005], as a result of the continuous electrical current supply [Can O.T., 2014], which enables the agglomeration of colloidal particles that are removed by flotation or precipitation [Piña et al., 2011]. Metallic cations such as Al^{3+} and Fe^{2+} are produced *in situ* in

the anode and therefore do not need to be externally supplied (Figure 1). Simultaneously, gases, mainly hydrogen, which make the coagulated contaminant float to the surface, are produced in the cathode [Holt et al., 2005]. Numerous electrochemical reactions occur during electrocoagulation (equation 1) [Mouedhen et al., 2008; Chen 2004].

In the anode:

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

In the cathode:

$$3H_2O + 3e \rightarrow \frac{3}{2}H_2(g) + 3OH^- \quad (2)$$

In the solution:

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

During the electrocoagulation process an electric field is generated around different contaminants and microorganisms present in water. Several authors have theoretically explained the effects of an electric field on bacteria in terms of membrane permeability and stability, and this electric field could rupture the cellular membrane [Zimmermann, 1973; Boudjema, 2014]. Some studies report electrochemical inactivation of bacterial and yeast cells. In the case of fecal coliforms such as *Escherichia coli*, an increase in the distance between the inner and outer membranes is seen during polarization and rupture. The microorganism inactivation rate increases along with amperage, while effectiveness increases with treatment time [Zimmermann, 1973; Boudjema, 2014].

In 2008, Ghernaout applied this process for the disinfection of artificial wastewaters contaminated with strains of *E. coli*. The results indicate that aluminum electrodes are the most efficient in destroying *E. coli* cells in comparison with stainless steel and carbon steel electrodes. In 2013, Ricordel investigated the effect of aluminum electrodes on model solutions with *E. coli* cultures, achieving 97% reduction after 35 min at an amperage of 0.22 A. In 2014, Boudjema

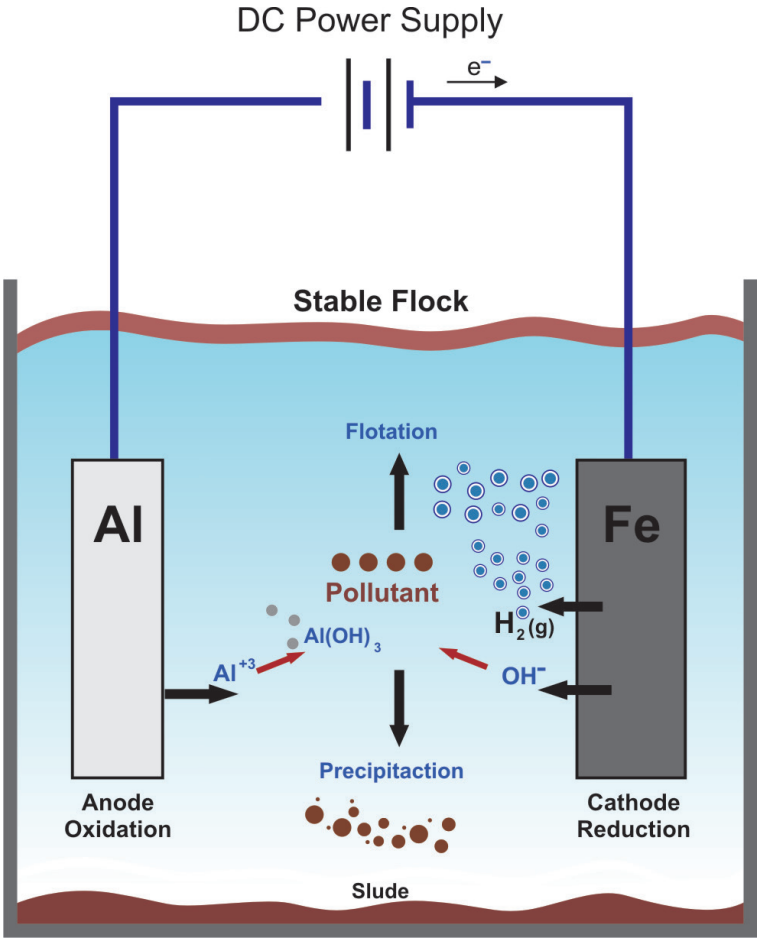

Figure 1. Reactions from the electrocoagulation process

applied electrocoagulation using aluminum electrodes to remove fecal coliforms from river water and achieved 99% reduction of these parameters. Taking into consideration the problem presented, it is of utmost importance to search for alternatives for disinfection, and in this particular case, to eliminate pathogenic microorganisms such as *E. coli*. Because only few studies have applied the electrocoagulation process to domestic and urban wastewaters, this study aimed at providing results using real conditions for the removal of *E. coli* using electrocoagulation.

To this end, the main objective of this study was to evaluate the efficiency of electrocoagulation on the removal of *E. coli* from domestic and urban wastewaters and to determine how the main parameters of the process (amperage, treatment time, and pH) affect it.

MATERIALS AND METHODS

Wastewater used

Domestic wastewater was obtained from a treatment plant at the Wastewater and Hazardous Residues Research Center of the National Engineering University (Citrar-UNI), which receives the domestic effluents from an urban zone of the city of Lima.

Electrocoagulation reactor

A batch-type laboratory-scale electrocoagulation reactor with a capacity of 10 L of water was fabricated using transparent acrylic. Four aluminum plates were used as sacrificial electrodes in the anode, and four 0.01-cm thick, 10-cm wide, and 10-cm long iron plates were used in the cathode, resulting in an area of 100 cm^2 (Fig. 2). A power source capable of supplying 0–15 A and adjustable voltage of 0–32 V was used to supply electrical current.

Experimental trials

The experimental design included three types of trials, maintaining the natural pH of the water and then modifying it towards a neutral value. In all of these trials, amperage was manipulated, and samples were collected at 10-min intervals until 60 min of treatment. The measurements of pH, conductivity, and temperature were taken in the field with using Oakton PCS 35 multiparameter device. The samples to determine the *E. coli* concentration were stored, labeled, and sent to the laboratory for analysis. The analysis method in "SMEWW-APHA-AWWA-WEF, Part9221B, C, E, G.2012; 22nd Ed. Other Escherichia coli Procedures" was used to determine the *E. coli* concentration in the raw and treated samples. Equation 4 was used to calculate the percent removal.

For *E. coli:*

$$\%R = \left(\frac{Ecoli_i - Ecoli_f}{Ecoli_i}\right)x100 \quad (4)$$

where:

%R – E.coli percent removal,
$E.coli_i$ – Initial *Escherichia coli* concentration,
$E.coli_f$ – Final *Escherichia coli* concentration.

Experimental design

A fully randomized three-factor factorial design with varying values was used. The factors considered in the design were amperage (x_1), treatment time (x_2), and pH (x_3), while the *E. coli* percent removal was the response variable (y_1)

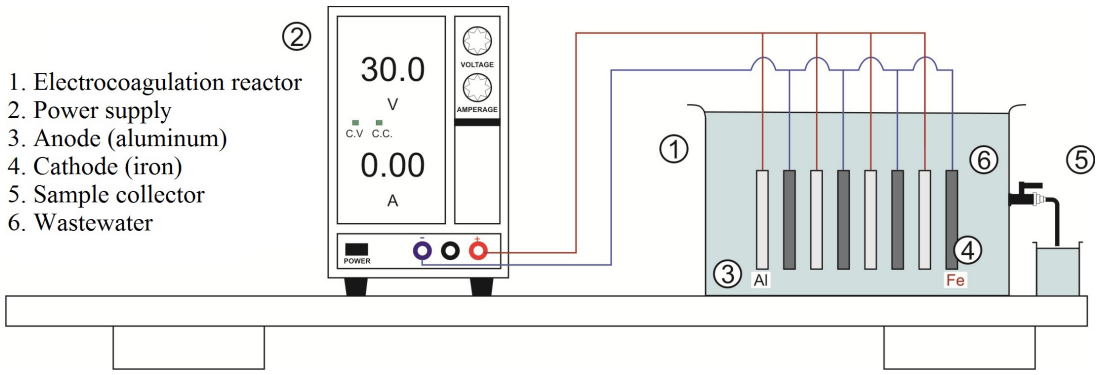

Figure 2. Diagram of the electrocoagulation reactor

(Table 1). This enabled us to determine the individual and interaction effects of the factors on the response variable and to identify how significant they were in the process. The statistical analyses were performed using Statgraphics Centurion XVI software and included an analysis of variance (ANOVA table) with a confidence level of 95% ($p < 0.05$), regression coefficients of a generalized second-order polynomial, and response surface figures. The model used for the analysis is detailed in equation 5.

$$y_i = b_0 + \sum_{i=1}^{n} b_i x_i + \sum_{i=1}^{n} b_{ii} {x_i}^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} b_{ij} x_i x_j \quad (5)$$

Where b_0, b_i, b_{ii}, and b_{ij} are the coefficients for linear, quadratic, and second-order interaction, respectively. Moreover, x_i and x_j represent the values for the independent variable, and y_i represents the *E. coli* percent removal. The fit of the polynomial model was expressed using the regression coefficient R^2.

Table 1. Experimental design factors and levels for *Escherichia coli*

Parameter	Value
Total coliforms (MPN/100 ml)	92000000
Fecal coliforms (MPN/100 ml)	16000000
Escherichia coli (MPN/100 ml)	2400000
pH	8.5
Conductivity (μ S/cm)	1200
COD (mg/L)	390.9
BOD_5 (mg/L)	137.0
Total solids (mg/L)	113
Oils and fats (NTU)	27.1
Total nitrogen (mg/L)	5.29
Total phosphorous (mg/L)	4.820

RESULTS AND DISCUSSION

Table 2 presents the results of this study, which shows the concentration of *E. coli* as 2400000 (MPN/100 ml). On the other hand, the values of BOD and COD were 137 mg/L and 390.9 mg/L, respectively, while conductivity and pH had acceptable values to use the electrocoagulation process.

Table 2. Physicochemical and microbiological analyses of domestic wastewater

Factors	Values					
x_1: Amperage (A)	1		2		3	
x_2: Time (min)	10	20	30	40	50	60
x_3: pH	7		8.5			

Effect of amperage

Amperage, also expressed as current density, which is defined as the relation between the current and the electrode surface, is a key parameter in the electrocoagulation process. Amperage determines the coagulant dosing at the anode and the formation of hydrogen gas in the cathode, as a consequence of Faraday's Law [Holt, 2005; Hakizimana, 2017]. The bubble density affects the system hydrodynamics, which in turn influences the mass transfer among contaminants, coagulant, and gas microbubbles, and finally dictates the collision rate of coagulated particles, which results in the formation of flocs [Hakizimana, 2017]. An increase in amperage accelerates the electrocoagulation process, particularly at the beginning [Attour et al., 2014]. (Fig. 3 and 5) illustrate an increase in the removal of microorganism with increasing amperage, although it is not substantial. After 30 min of treatment, at all three amperage values, 99% removal efficiency was achieved. Figures 4 and 6 show that the best results were obtained at the amperage values of 2 A and 3A, following 60 min of treatment, wherein the *E. coli* concentration decreased from 2400000 (MPN/100 ml) to 9200 (MPN/100 ml), thereby achieving up to 99.9% removal efficiency. These results match those reported by Gusmao in 2010, who indicated that a treatment time of 60 min could eliminate 98.8% of *E. coli*. Moreover, in 2015, Castro indicated that a 1-log *E. coli* removal from lab-prepared synthetic water can be achieved after 40 min of treatment. This removal is attributed to the adsorption of *E. coli* on the flocs formed by the electrocoagulation process, which rise to the surface or precipitate to the bottom of the reactor due to flotation [Ghernaout et al., 2008]. The high removal efficiencies found are also extremely similar to those obtained by Ricordel, who reported 97% removal efficiency, where the removal of bacteria using electrocoagulation could be attributed to strong bacterial adhesion on the surface of aluminum-electrogenerated particles, followed by the separation of the precipitated solids. Conversely, in 2014, Chopra

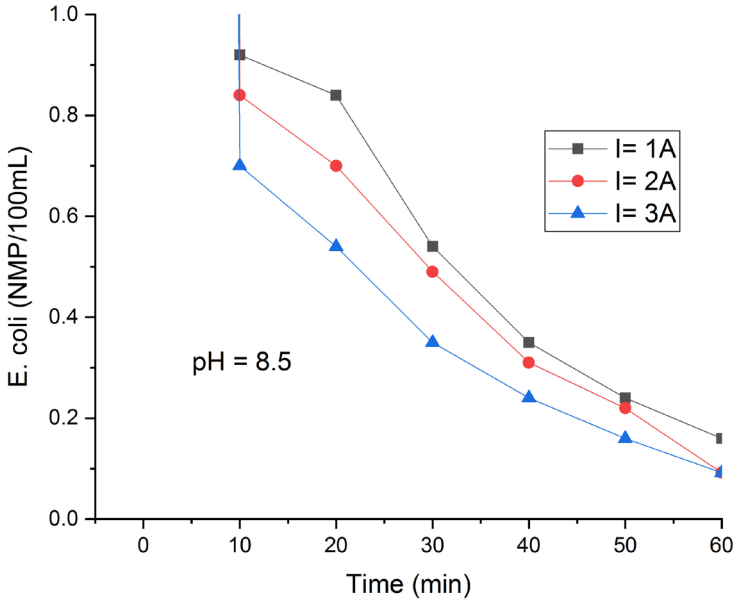

Figure 3. *Escherichia coli* variation as a function of time at different amperage values (*E. coli* = 2400000 MPN/100 ml; pH = 8.5)

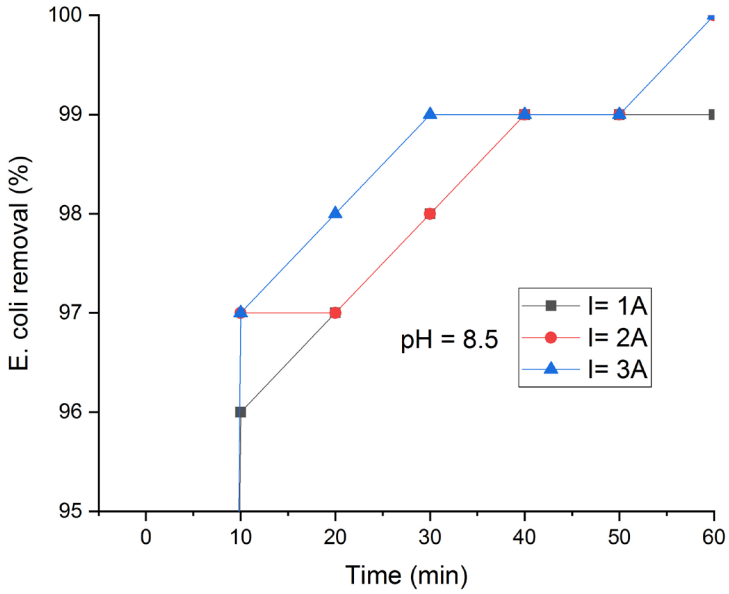

Figure 4. *Escherichia coli* percent removal as a function of time (*E. coli* = 2400000 MPN/100 ml; pH = 8.5)

reported high pathogenic microorganism removal considering total coliforms, achieving 99.9% removal efficiency after 40 min of treatment at a current density of 2.65 A/m^2.

Effect of treatment time

With increasing treatment time, the concentration of metallic ions and their hydroxide flocs grows as well [Chopra, 2014], allowing for better contaminant coagulation and flocculation. As a result, the contaminant removal efficiency increases along with retention time, up to a point where this efficiency becomes constant [Khandegar, 2013]. (Fig. 3, 4, 5, and 6) illustrate a high pathogenic microorganism removal within the first 10 min of treatment, reaching 97% owing to the removal of a large quantity of organic matter and flocs. After 30 min of treatment, 99% removal efficiency was achieved both at natural as well as neutral pH values, and it became almost constant up to 60 min of treatment. Treatment time is faster with higher current density; these two parameters are closely connected [Attour et al., 2014].

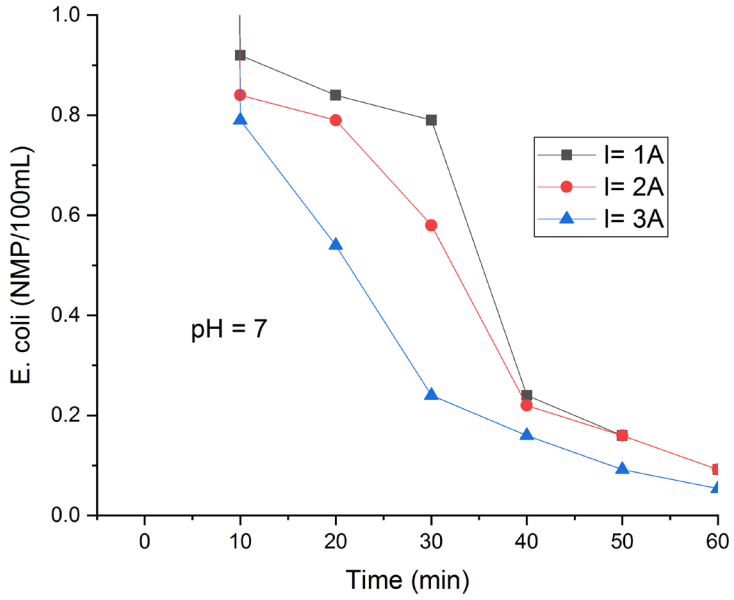

Figure 5. *Escherichia coli* variation as a function of amperage values (*E. coli* = 2400000 MPN/100 ml; pH = 7)

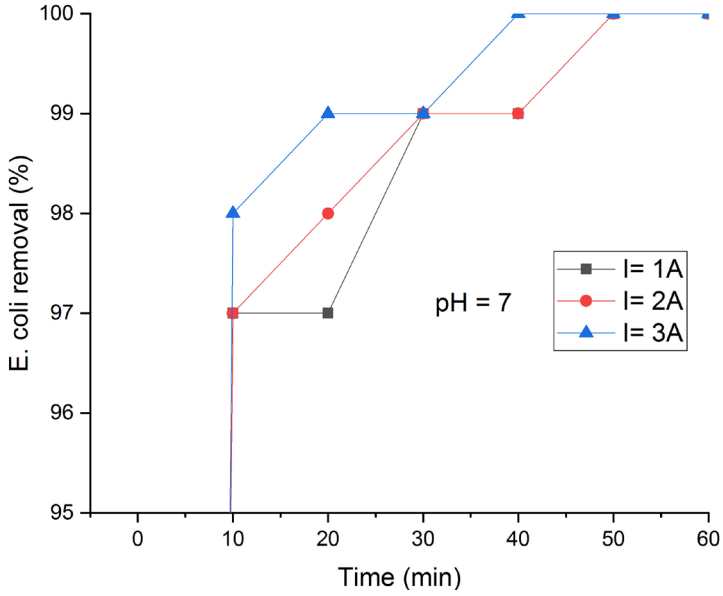

Figure 6. *Escherichia coli* percent removal as a function of time (*E. coli* = 2400000 MPN/100 ml; pH = 7)

Effect of pH

The initial pH of wastewater is one of the most important factors affecting the efficiency of electrocoagulation [Can, 2014; Bouamra et al., 2012]. The solubility of metallic hydroxides [Mollah, 2004], which act as coagulants, enabling the agglomeration of colloidal particles, also affects the efficiency of electrocoagulation [Piña et al., 2011]. For aluminum, the predominant forms are Al^{3+} cations at low pH, $Al(OH)_4^-$ aluminates at pH ≥ 10, and non-soluble hydroxides $Al(OH)_3$ at an intermediate or neutral pH [Yehya et al., 2014]. The highest *E. coli* percent removal reached 99.9% at a neutral pH of 7, amperage of 3 A, and treatment time of 60 min (Fig. 5 and 6). However, a similar efficiency of 99% was obtained at the natural pH of water (pH 8.5). These results are consistent with those reported by Chopra in 2014, who indicated achieving the best efficiency at a close to neutral pH of 7.5. In 2013, Ricordel analyzed the effect of pH on the *E. coli* removal efficiency and pointed out that when the initial pH increased from 7.5 to 9.5, *E. coli* removal efficiency

significantly decreased from 96% to 72% in an average time of 35 min and an amperage of 0.22 A. The best removal efficiency was achieved at the pH values close to 7.

Experimental design results

According to the proposed experimental design, Table 3 illustrates the 36 experiments conducted in this study. Moreover, equation 5 presents the quadratic regression model for *E. coli* percent removal (y_1), which enables us to investigate the effects of the independent variables (x_1, x_2, and x_3) and their effect on the response variable (y_1).

$$Y_{(1)} = 98.452 + 0.35x_1 + 0.117024x_2 - \\ - 0.474074x_3 + 0.0833333x_1^2 - 0.01x_1x_2 + \\ + 0.0x_1x_3 - 0.00077381x_2^2 + 0.00190476x_2x_3 \quad (6)$$

ANOVA showed a regression coefficient (R^2) of 0.9287 for *E. coli*, which means that the proposed statistical model explains 92.87% of the variation in *E. coli* percent removal and indicates a good model fit (Table 4).

Figure 7 illustrates the main effects on the response variable, with the effect of time, amperage, and pH being significant at a significance level of $\alpha = 0.05$.

Table 3. Experimental design with *Escherichia coli* percent removal as response variable

Exp. N°	Factors				Exp. N°	Factors			
	Amperage (A)	Time (min)	pH	*E. coli* percent removal		Amperage (A)	Time (min)	pH	*E. coli* percent removal
	x_1	x_2	x_3	y		x_1	x_2	x_3	y
1	1	10	7	97	19	2	10	8.5	97
2	1	20	7	97	20	2	20	8.5	97
3	1	30	7	99	21	2	30	8.5	98
4	1	40	7	99	22	2	40	8.5	99
5	1	50	7	100	23	2	50	8.5	99
6	1	60	7	100	24	2	60	8.5	100
7	1	10	8.5	96	25	3	10	7	98
8	1	20	8.5	97	26	3	20	7	99
9	1	30	8.5	98	27	3	30	7	99
10	1	40	8.5	99	28	3	40	7	100
11	1	50	8.5	99	29	3	50	7	100
12	1	60	8.5	99	30	3	60	7	100
13	2	10	7	97	31	3	10	8.5	97
14	2	20	7	98	32	3	20	8.5	98
15	2	30	7	99	33	3	30	8.5	99
16	2	40	7	99	34	3	40	8.5	99
17	2	50	7	100	35	3	50	8.5	99
18	2	60	7	100	36	3	60	8.5	100

Table 4. Chart of *Escherichia coli* ANOVA

Source of variation	Sum of squares	DF	MS	F-value	P-value
x_1: Amperage (A)	2.66667	1	2.66667	21.82	0.0001
x_2: Time (min)	34.8595	1	34.8595	285.21	0.0000
x_3: pH	3.36111	1	3.36111	27.50	0.0000
x_1^2	0.0555556	1	0.0555556	0.45	0.5059
x_1x_2	0.7	1	0.7	5.73	0.0239
x_1x_3	0.0	1	0	0.00	1.0000
x_2^2	1.34127	1	1.34127	10.97	0.0026
x_2x_3	0.0214286	1	0.0214286	0.6787	0.6787
Total error	3.3	27	0.122222		
Total (corr.)	46.3056	35			
R^2 = 92.8734 %, Adj R^2 = 90.7618 %					

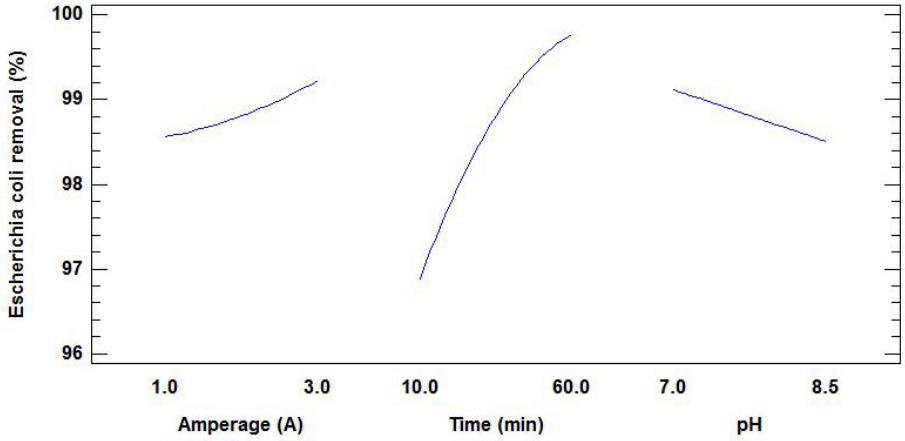

Figure 7. Graph of main effects on the removal of *Escherichia coli*

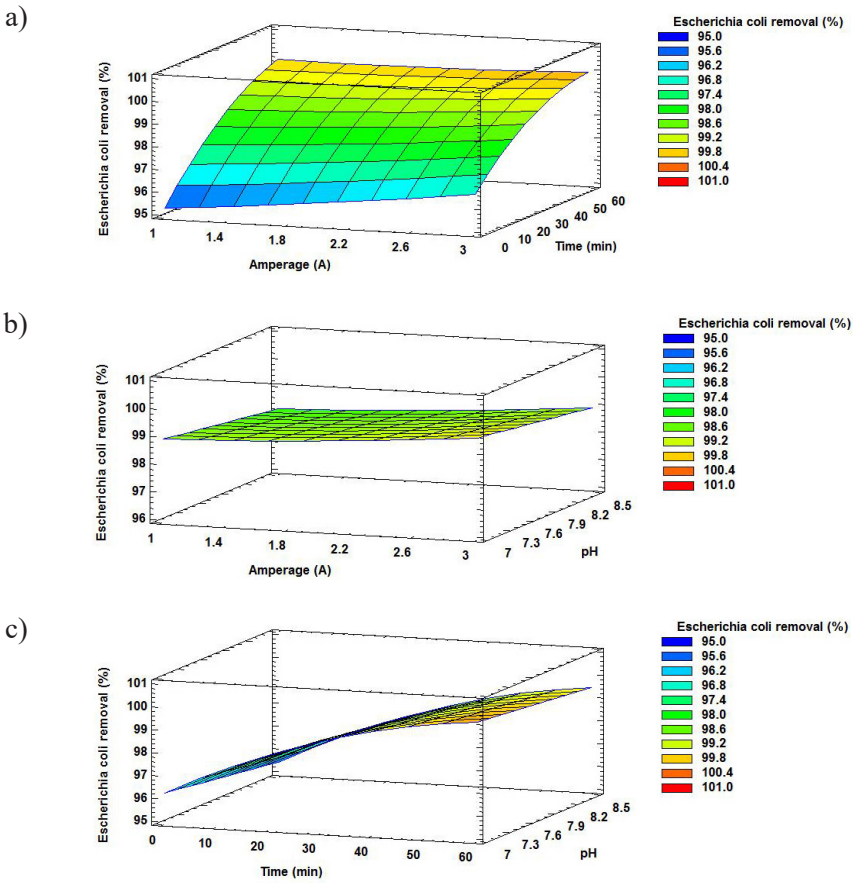

Figure 8. Three-dimensional response surface graphs for *Escherichia coli* percent removal: (a) amperage and time; (b) amperage and pH; and (c) time and pH

Figure 8 presents the response surface graphs, showing *E. coli* percent removal variation according to amperage, time, and pH.

CONCLUSIONS

The proposed experimental design indicated that the three variables, namely amperage, time, and pH, significantly affect the response variable *E. coli* percent removal. Using this design, a regression coefficient (R^2) of 0.9287 was obtained, which means that the proposed statistical model explained 92.87% of the variation in *E. coli* percent removal, thereby representing a good model fit. The highest *E. coli* percent removal achieved was 99.9% at a neutral pH of 7, amperage of 3 A, and treatment time of 60 min. However, similar values of 99% were also achieved at the natural wastewater pH of 8.5. With regards to the

treatment time, after 20 min of treatment, 98% *E. coli* percent removal was achieved at natural pH values, whereas a similar value of 99% was achieved at a pH of 7, during which a large quantity of suspended solids and organic matter present in the domestic wastewater are removed. Although the statistical analyses indicated that manipulating pH to neutrality improved the efficiency, it was not substantial enough to warrant implementation of manipulating pH in the treatment plant.

In this study, it was clarified that electrocoagulation removes a large percentage of pathogenic microorganisms, such as *E. coli*, from domestic wastewater. The removal of pathogenic microorganisms in the primary electrocoagulation reactor is superior to traditional biological processes such as activated sludge, which can typically remove 60%-80% of pathogens. This will substantially reduce the use of chlorine during the final treatment stage (disinfection), thereby preventing the discharge of water with high chlorine concentrations.

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