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Application of Doehlert design for optimization of the electroflotation method for the treatment of wastewater from poultry slaughtering and processing

Aplicação do modelo Doehlert na otimização do método de eletroflotação para o tratamento de efluentes do abate e processamento de aves

Aplicación del modelo Doehlert para la optimización del método de electroflotación para el tratamiento de aguas residuales del sacrificio y procesamiento de aves de corral

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Abstract

In this work, an electroflotation (EF) method for the treatment of poultry slaughterhouse effluent was proposed, and its efficiency in reducing chemical oxygen demand (COD) was investigated. The following operating conditions were optimized through Doehlert design: [$Al_2(SO_4)_3$], pH, treatment time, and current density. Treatment of the effluent was carried out in a reactor with TiO_2–RuO_2 (anode) and iron (cathode) electrodes. The optimum process conditions were obtained with a pH 9, current density of 60 A m^{-2}, electrolysis time of 80 min, and [$Al_2(SO_4)_3$] of 1.8 mg L^{-1}. Under these operating conditions, turbidity, COD, and biochemical oxygen demand (BOD) removal efficiencies of 93.1%, 80.7%, and 89.7%, respectively, were obtained. The operating cost of the process was calculated at 0.9 USD per m^3. The EF method combined with chemical coagulation was shown to be a suitable process for the treatment of effluent from the slaughter and processing of poultry.

Keywords: Electrochemistry; Chemometrics; Wastewater slaughterhouse; COD.

Resumo

No presente trabalho, é proposto o método de eletroflotação (EF) para o tratamento de efluentes do abate e processamento de aves, assim como também é observada sua eficiência na redução da concentração da demanda química de oxigênio (DQO). As seguintes condições operacionais foram otimizadas pelo método Doehlert: [$Al_2(SO_4)_3$], pH, tempo de tratamento e densidade de corrente. O tratamento do efluente foi realizado em um reator equipado com eletrodos de TiO_2–RuO_2 (ânodo) e ferro (cátodo). As condições de operação otimizadas obtidas foram pH 9, densidade de corrente de 60 A m^{-2}, tempo de eletrólises de 80 min e [$Al_2(SO_4)_3$] de 1,8 mg L^{-1}. Sob estas condições operacionais foram obtidas eficiências de remoção de turbidez de 93,1%, DQO de 80,7% e demanda biológica de oxigênio (DBO) de 89,7%. Os custos operacionais do processo foram calculados em US$0,9 por m^3. O método de EF combinado com a coagulação química mostrou-se um método adequado para o tratamento de efluentes do abate e processamento de aves.

Palavras-chave: Eletroquímica; Quimiometria; Efluentes de abate; DQO.

Resumen

En este trabajo, proponemos el método de electroflotación (EF) para el tratamiento de aguas residuales de mataderos avícolas y se investigó su eficiencia en la reducción de la demanda química de oxígeno (DQO). Las siguientes condiciones de funcionamiento fueron optimizadas por el método Doehlert: [$Al_2(SO_4)_3$], pH, tiempo del tratamiento y densidad de corriente. El efluente fue tratado en un reactor equipado con electrodos de TiO_2–RuO_2 (ánodo) y hierro (cátodo). Las condiciones óptimas de funcionamiento encontradas fueron pH= 9, densidad de corriente = 60 A m^{-2}, tiempo de electrolisis = 80 min y [$Al_2(SO_4)_3$]= 1,8 mg L^{-1}. Con estas condiciones de funcionamiento fueron obtenidas las siguientes eficiencias de reducción: turbidez= 93,1%, DQO= 80,7% y demanda biológica de oxígeno (DBO)= 89,7%. El costo operativo del proceso se calculó en US$0,9 por m^3. El método de EF combinado con la coagulación química demostró ser un proceso adecuado para el tratamiento de aguas residuales del sacrificio y procesamiento de aves de corral.

Palabras clave: Electroquímica; Quimiometría; Aguas residuales de matadero; DQO.

1. Introduction

The consumption of poultry meat has generated great economic and social growth, with a growth of over 26% in the last decade in Brazil (ABPA, 2018). This growth impacts water resources, once the production of poultry requires a high consumption of water; there is an average consumption of 20 L of water per poultry animal during processing (Eryuruk et al., 2017). These industries have great potential for pollution as there is considerable production of liquid waste with high organic loads (Oller et al., 2011) and it is fundamental to treat the effluents generated to mitigate health and environmental impacts.

Electrochemical treatment techniques are promising technologies, and they have been used as advanced low-cost technologies for the removal of organic compounds (Eryuruk et al., 2017), potentially toxic elements (Belkacem et al., 2008), and coliforms (Zarei et al., 2018) through the application of an electric current through electrodes submerged in the effluent, without the addition of chemical coagulants (Quin et al., 2013, Gomes et al., 2018, Mickova, 2015).

During the electroflotation (EF) technique, hydrolysis of a water molecule leads to the generation of gases, oxygen at the anode ($2H_2O \rightarrow 4H^+ + O_2\uparrow + 4e^-$) and hydrogen at the cathode ($4e^- + 4H_2O \rightarrow 2H_2\uparrow + 4OH^-$), released in the form of bubbles (Wang et al., 2010). These bubbles entrap the suspended solids at the surface of the reactor, facilitating their removal by scraping (Mickova, 2015).

The EF technique is viewed as a clean technology compared to conventional techniques, such as coagulation-floculation (Santo, et al., 2012), anaerobic treatment (Nacheva et al., 2011), and aerobic treatment (Keskes et al., 2012), due to the low generation of sludge (Mickova, 2015), low retention time, and low operating costs when the working conditions are optimized (Phalakornkule et al., 2010, Mollah et al., 2001, Mouedhen et al., 2008).

Kobya et al. (2006) observed the efficiency in the removal biochemical demand oxygen (BOD) and in the removal chemical demand oxygen (COD), value of 80% and value of 85%, respectively, when applying the electrocoagulation technique in the treatment of a poultry slaughterhouse effluent and Bayar et al. (2011) reported efficiency in the removal COD value of 90% and removal turbidity value of 98%, using the same technique in the same effluent, thus demonstrating that the electrochemical technique is efficient in the treatment of poultry slaughterhouse effluent.

In this work, application of the EF method for the treatment of poultry slaughterhouse wastewater was proposed, and its efficiency in the reduction of COD was investigated when using operating conditions optimized through a fractional factorial design. Calculation of the operational cost was carried out in order to show the economic viability of the treatment system based on the EF technique. This paper is based on the principle of the need to bring knowledge produced in academia to society, with wide applicability in problem solving, integrating science, society and technology (Pereira et al., 2018).

2. Methodology

2.1 Sampling

The effluent used was collected in a poultry slaughterhouse, with an average of 3.000 daily slaughter poultry. Wastewater comes from the cutting, bleeding and scalding, plundering, further processing and evisceration, storing, packaging, and washing of the equipment which after said produced passes through a railing to remove coarse particles, following by the equalization tank, where the samples were collected for the experiments.

2.2 Reagents and instrumentation

The COD, BOD, turbidity, and conductivity were determined according to methods 5220 B, 5210 B, 2130 B, and 2510 B (APHA, 1998), respectively, described in the Standard Methods for Water and Wastewater Analysis. The pH was adjusted using 1.0 mol L^{-1} NaOH (Merck), and aluminium sulphate [$Al_2(SO_4)_3$] was applied as the coagulant in the tests determined by the experimental design.

2.3 Experimental design

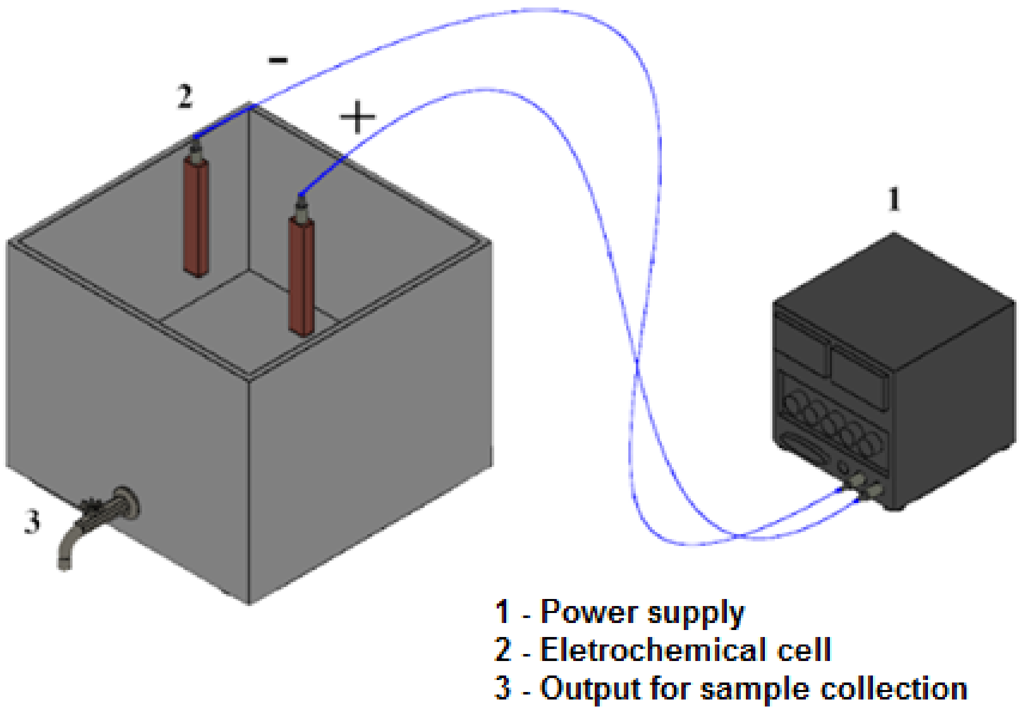
The experimental matrix was developed by applying the 2^{4-1} fractional factorial design for initial pH (4, 7, and 10), current density (10, 35, and 60 A m^{-2}), electrolysis time (10, 45, and 80 min), and aluminium sulphate dosage (0, 2.5, and 5.0 mg L^{-1}). A Doehlert design for two variables was applied for initial pH (4, 5.5, 7, 8.5 and 10), and aluminium sulphate dosage

(0, 1.5 and 2.5 g L^{-1}), achieving as response the COD reduction efficiency (expressed in %). A simultaneous experimental design was performed using triplicate the central point to evaluate the experimental error of design. The test combinations were developed using Statistica® software version 6.0 (StatSoft, Tulsa, USA).

2.4 Electrochemical process

The optimization of the process was performed with a real sample of poultry effluent using a polymethylmethacrylate (PMMA) reactor with a volume of 16 cm^3 and two electrodes connected in parallel, as shown in Figure 1.

Figure 1. Schematic diagram of the experimental configuration of the electroflotation process.



Source: Authors.

The anode was composed of TiO_2–RuO_2 with an effective area of 214.95 cm^2, and the iron cathode (stainless steel 316) consisted of a perforated plate with an effective area of 53.296 cm^2. The electrodes were connected to a continuous power supply (model ITFA 5000, Instrutemp, China). The distance between the electrodes was defined based on the works of Paulista et al. (2018) and Adjeroud et al. (2015), who found an optimal distance of 1.0 cm.

Five samples of slaughter and poultry processing wastewater, collected on consecutive days, were analyzed. The percent removal of COD, BOD, and the turbidity were calculated by the final value/initial value ratio for each parameter. All analyses were performed in triplicate.

For the evaluation of the operational cost of the treatment, the cost of the electricity tariff was considered according to $OC_{EF} = t \times V_{med} \times I \times E_{power}/V_{waste}$, where OC_{EF} is the operational cost (USD/m^3), *t* is the electrolysis time (h), V_{med} is the medium voltage (V), *I* is the electric current (A), E_{power} is the cost of electricity (USD/Wh), and V_{waste} is the treated effluent volume (m^3).

3. Results and Discussion

Effluents generated from the slaughtering and processing of poultry presented a high organic load. The collected raw effluent presented COD values of 752–1135 mg L^{-1}, BOD values of 120–600 mg L^{-1}, pH values of 6.1–6.6, turbidity values of 140–192 NTU, and conductivity values of 345–767 μs cm^{-1}. The BOD values and COD values were higher than those reported by Bazrafshan et al. (2012) and Drogui et al. (2008), BOD values of 60 mg L^{-1} and COD values of 500 mg L^{-1}, respectively. However, BOD and COD values are lower than those reported by Bayar et al. (2011), COD values of 2171 mg L^{-1} and BOD values of 1123 mg L^{-1}, and Kobya et al. (2006), COD values of 29000–26000 mg L^{-1} and BOD values of 12000–10000 mg L^{-1}. The turbidity values were also lower than those reported by Bustillo-Lecompte & Mehrvar (2015) and Sunada (2011), 271–279 NTU. It is suggested that the concentration differences may be due to the size of the industrial plant and the lack of control of water consumption in the enterprise, which may promote dilution of the effluent. The pH and conductivity values, 6.1–6.5 and 330–500 μs cm^{-1}, respectively, are in agreement with the values measured by Drogui et al. (2008).

3.1 Optimization of the electroflotation process

The experimental design consisted of analysis of the initial pH (4, 7, and 10), current density (10, 35, and 60 A m^{-2}), electrolysis time (10, 45, and 80 min), and aluminium sulphate dosage (0, 2.5, and 5.0 mg L^{-1}). Table 1 shows the matrix of the 2^{4-1} fractional factorial design, the experiments were performed in random order, with 3 replicates of the central point, used to estimate the experimental error of the factorial design, and a response to COD reduction efficiency (expressed in %).

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Table 1. Experimental conditions and responses for 2^{4-1} fractional factorial design with 3 replicates of the central points.

Experiment	pH	Current density ($A.m^{-2}$)	Time (min)	[$Al_2(SO_4)_3$] ($g\ L^{-1}$)	COD ($mg\ L^{-1}$)	Removal COD (%)
1	4/(-1)	10/(-1)	10/(-1)	0/(-1)	-351.0	56.3
2	10/(+1)	10/(-1)	10/(-1)	2.5/(+1)	-75.9	90.6
3	4/(-1)	60/(+1)	10/(-1)	2.5/(+1)	-281.7	64.9
4	10/(+1)	60/(+1)	10/(-1)	0/(-1)	-267.0	66.8
5	4/(-1)	10/(-1)	80/(+1)	2.5/(+1)	-220.8	72.5
6	10/(+1)	10/(-1)	80/(+1)	0/(-1)	-231.3	71.2
7	4/(-1)	60/(+1)	80/(+1)	0/(-1)	-291.1	63.8
8	10/(+1)	60/(+1)	80/(+1)	2.5/(+1)	-45.4	94.3
9	7/(0)	35/(0)	45/(0)	1.25/(0)	-174.6	78.3
10	7/(0)	35/(0)	45/(0)	1.25/(0)	-212.4	73.6
11	7/(0)	35/(0)	45/(0)	1.25/(0)	-206.1	74.4

Source: Authors.

The experiments with short durations and acidic pH values presented inferior performance (36–52% COD removal). It was observed that the tests with the addition of chemical coagulant showed greater COD removal, indicating that the EF with added of the coagulant is efficient to treat effluents poultry slaughterhouse.

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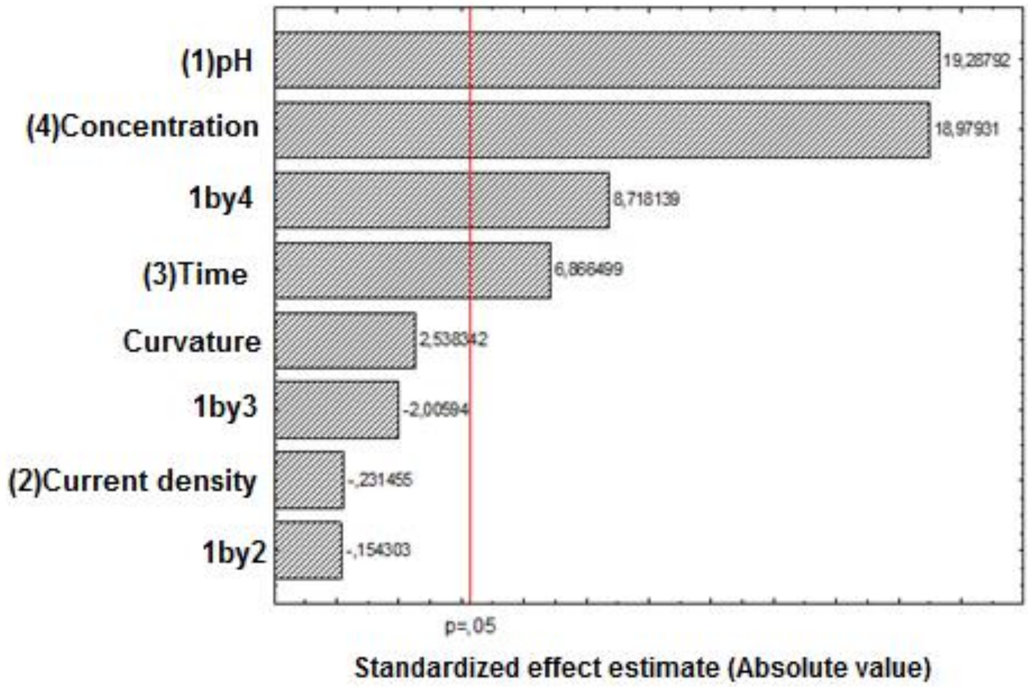
The results obtained for the main effects and interaction effects are shown in the Pareto graph (Figure 2).

Figure 2. Pareto chart for 2^{4-1} factorial design with 3 central points (effects of pH, electrolysis time, coagulant concentration and current density).



Source: Authors.

The Pareto graph suggested that the current density was not significant for the treatment. On the other hand, the combination of the maximum values of [$Al_2(SO_4)_3$] (5 g L^{-1}), pH (10), and time (80 min) showed higher COD reduction efficiency (73%). Similar results were described by Un et al. (2009), who demonstrated that the COD removal efficiency (98.9%) increased with increasing PAC1 coagulant dosage. In EF processes, the pH influences the size of the bubbles generated. According to Montes-Atenas et al. (2010), the hydrogen bubbles produced have smaller diameters in neutral and alkaline medium. Moreover, according to Hann (1984) the effluent treatment is more efficient when small bubbles are formed, which justifies the increase in removal with increasing pH.

The analysis of variance (ANOVA) (Table 2) showed that the linear model was significant (p-value, 0.1264>0.05), showing no adjustment to a 95% confidence level.

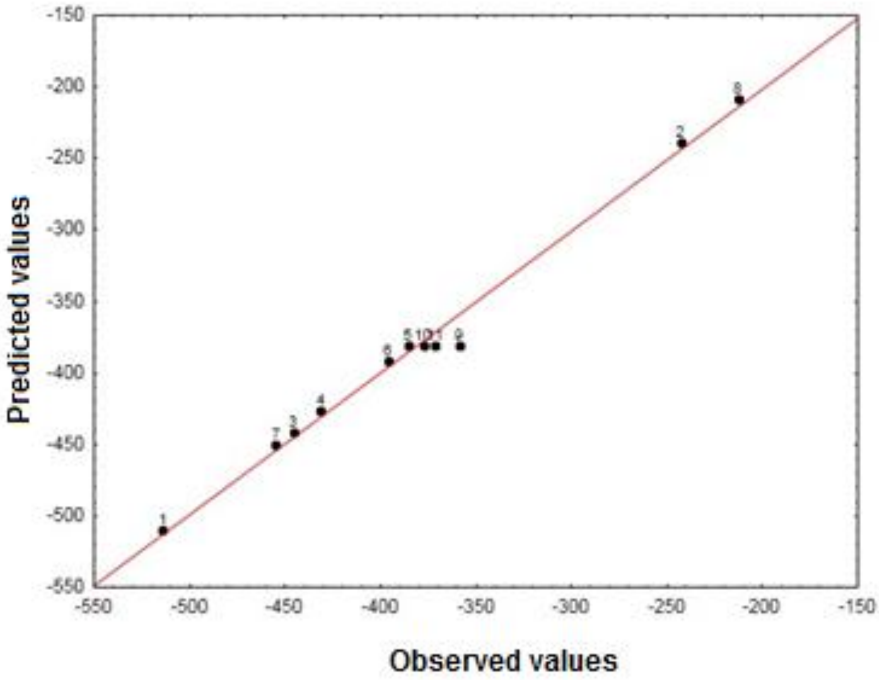
Table 2. ANOVA for 2^{4-1} fractional factorial design for COD removal.

Source	Sum of squares	Degrees of freedom	Mean of squares	*F*-value	*p*-value
(1)pH	33462.85	1	33462.85	372.0238	0.002677
(2)Current density	4.82	1	4.82	0.0536	0.838485
(3)Time	4240.95	1	4240.95	47.1488	0.020558
(4)Concentration	32400.60	1	32400.60	360.2143	0.002765
Interaction 1 x 2	2.14	1	2.14	0.0238	0.891535
Interaction 1 x 3	361.93	1	361.93	4.0238	0.182697
Interaction 1 x 4	6836.59	1	6836.59	76.0060	0.012903
Lack of fit	**579.55**	**1**	**579.55**	**6.4432**	**0.126431**
Pure Error	179.90	2	89.95		
Total SS	78069.33	10			

Source: Authors.

Other evidence for the fit of the model was provided by the good correlation (r=0.990) found between the experimental values and predicted values in (Figure 3). Thus, for the analysis of the efficiency of the treatment of poultry effluent by the EF method, the condition assumed for current density and electrolysis time was 60 A m^{-2} and 80 min, respectively.

Figure 3. Linear representation of model fit.



Source: Authors.

3.2 Evaluation of electroflotation process operating conditions through Doehlert Design

Evaluation of reduction COD, experimental conditions of initial pH and aluminium sulphate dosage, was performed through a Doehlert design, as shown in Table 3.

Table 3. Doehlert design for EF with coded values and real values of the pH and coagulant concentration variables, evaluating the final COD concentration.

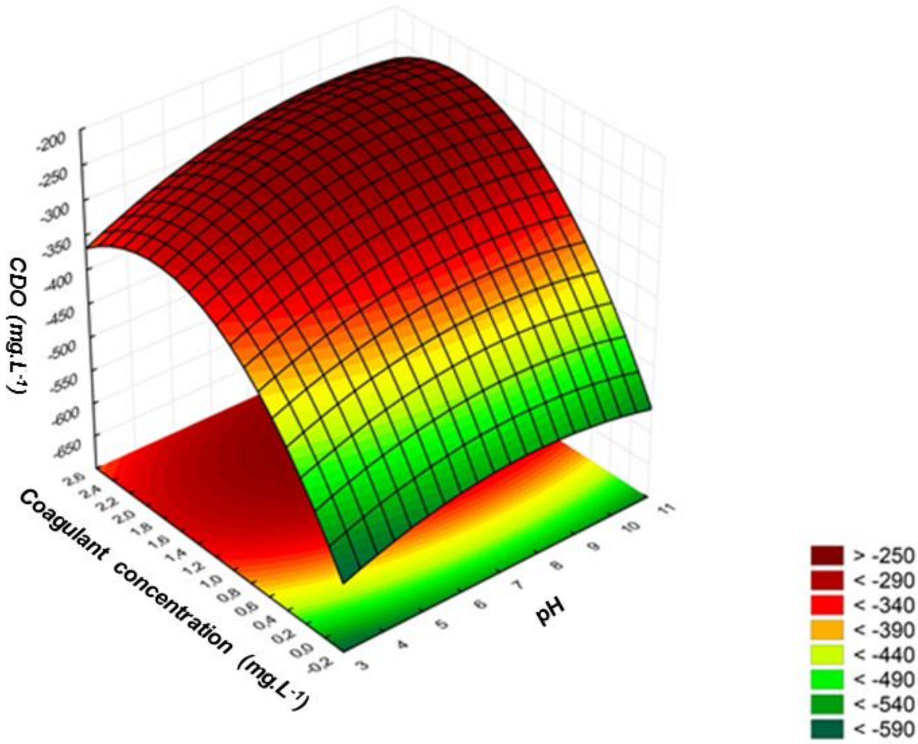
	Coded values / Real values			
Experiment	**pH**	**Coagulant concentration (g. L^{-1})**	**COD (mg. L^{-1})**	**Removal COD (%)**
1	(+1) / 10	(0) / 1.25	-259.6	75.0
2	(+0.5) / 8.5	(+0.866) / 2.5	-272.2	73.8
3	(-1) / 4	(0) / 1.25	-325.7	68.7
4	(-0.5) / 5.5	(+0.866) 2.5	-287.9	72.3
5	(+0.5) / 8.5	(-0.866) / 0	-495.8	52.3
6	(-0.5) / 5.5	(-0.866) /0	-491.6	52.7
7	(0) / 7	(0) / 1.25	-278.5	73.2
8	(0) / 7	(0) / 1.25	-265.9	74.4
9	(0) / 7	(0) / 1.25	-269.0	74.1

Source: Authors.

It was observed that in the tests with addition of chemical coagulant more significant COD removals were obtained, up to 75%, indicating that the coagulation added to electroflotation is an interesting combination to treat poultry slaughterhouse effluents. It was also observed that the increase in the coagulant dosage causes a reduction in the efficiency of the electroflotation process, suggesting that the increase in the size of the flakes formed makes the flotation process more difficult. Bazrafshan et al. (2012) reported in their research that the removal rate increased linearly with increasing doses of polyaluminum chloride (PACl). Their experiments infer that the combined processes are superior to those isolated for the removal of organic compounds.

After analysis of the proposed Doehlert design, response surface graph was generated to evaluate reduction efficiency COD as a function initial pH and aluminium sulphate dosage (Figure 4).

Figure 4. Response surface plots generated from the Doehlert design for reduction COD.



Source: Authors.

As can be seen in Figure 4, COD is found to decrease as the pH values are increased and the concentration of chemical coagulant is reduced. An optimum region is observed when the concentration of aluminum sulfate varies between 1.6 and 2.5 g L^{-1} and the pH is neutral to alkaline.

It was also observed that in EF treatment there is a decrease in efficiency when the coagulant concentration is increased, above 2 g L^{-1}. It is suggested that by increasing the amount of coagulant, the flakes formed become denser, making the flotation process more difficult. Hanauer et al. (2019) used aluminum sulfate to treat poultry slaughterhouse effluent and found that the performance of this coagulant was 72.61 ± 2.60% at 100 mg L^{-1} and when the dose was increased to 125 and 150 mg L^{-1} the efficiency decreased to 64.74 ± 6.64% and 68.57 ± 0.46%, respectively.

The best compromise conditions for operation of the EF process were therefore selected by visual inspection of the results of the Doehlert design. These values were pH (9), coagulant concentration (1.8 g L^{-1}), electrolysis time (80 min) and current density (60 A m^{-2}).

3.3 Application of the electroflotation technique in the treatment of poultry slaughterhouse effluent

Application of the method was performed using the condition most favourable to treatment by EF, defined from the optimization with Doehlert design. Table 4 shows the results of the effluent analysis after the application of EF treatment.

Table 4. Efficiency of EF performance in the treatment of poultry slaughterhouse effluent (reaction time = 80 min, pH = 9, current density = 60 A m^{-2} and [$Al_2(SO_4)_3$] = 1.8 g L^{-1}).

Sample	**Raw effluent**			***Turbidity**	***COD**	***BOD**	****Energy cost**
	Turbidity mg L^{-1}	**COD** mg L^{-1}	**BOD** mg L^{-1}	**%**	**%**	**%**	**(USD)/m^3**
EF-01	242.3	811.3	120	98.6	69.2	85	0.6
EF-02	156	1193.1	150	95.9	85.6	90	0.6
EF-03	180.6	895.0	300	94.5	86.1	91	0.7
EF-04	155.6	822.1	210	87.2	85.3	84.2	0.6
EF-05	192.6	826.8	450	89.5	77.7	98.6	0.7
Average	185.4	909.7	246	93.1	80.7	89.7	0.6
SD	35.6	161.8	133.1	4.6	7.3	5.8	0.0

* % turbidity, COD, BOD (removal).
** trading dollar quotation: 1.00 R$ = 5.33 US$ (04/20/2020).
Source: Authors.

It is observed that there was a reduction in COD ranging from 69.2–85.5%, with an average of 80.7%. Similar results were observed in studies developed by Phalakornkule et al. (2010) 64% COD removal, who used an electrochemical process with aluminium electrodes in a pre-treatment stage of industrial effluent from palm oil processing, and Ahmadian et al. (2012) 62% COD removal, who treated wastewater from slaughterhouses by electrocoagulation using Fe electrodes.

The Brazilian Environment Council (*Conselho Nacional do Meio Ambiente* - CONAMA) (Brasil, 2005) does not inform limit values for the COD parameter. It only establishes that the discharge does not change the characteristics of the receiving body.

For turbidity and BOD, mean reductions of 93.1% and 89.7% were achieved, that is, values of 11.1 NTU and 34 mg L^{-1}, respectively, which are adequate to CONAMA (<100 NTU turbidity and 60% BOD removal) for effluent releases (Brasil, 2005). Similar removal efficiencies were obtained by Drogui et al. (2008) wich 90% turbidity and 86% BOD removal using electrocoagulation in the treatment of poultry slaughterhouse effluent.

The operating cost was estimated based on the value for industrial electric power (0.0001 USD/Wh) (Energisa, 2019). The average cost of electric energy spent in each experiment was 0.6 USD/m^3. The coagulant [$Al_2(SO_4)_3$] (1.8 g L^{-1}) was added to the process at a cost of 0.3 USD/kg (Leroymerlin, 2019). Therefore, the average total cost to treat the poultry slaughterhouse effluent per EF was 0.9 USD per m^3 of treated effluent. Ozyonar & Karagozoglu (2014) who treated bovine slaughterhouse effluent by electrocoagulation obtained a removal efficiency of 78.8% COD and 90.2% turbidity with a total cost of 2.76 USD/m^3. Eryuruk et al. (2018) subjected the effluent wastewater to electrocoagulation with peroxidation and obtained a removal efficiency of 95.5% COD with an operational cost of 9 USD per m^3 of treated effluent.

4. Conclusion

Through Doehlert design, it was possible to optimize the operational conditions of the EF process by achieving a reduction of more than 80.7% COD, 89.7% BOD, and 93.1% turbidity of the poultry slaughterhouse effluent under conditions of a pH value of 9, current density of 60 A m^{-2}, time of 80 min, and [$Al_2(SO_4)_3$] value of 1.8 g L^{-1}. Analysis of the operational cost indicated that the EF process presented an average operating cost of 0.9 USD per m^3 of treated effluent, considering a low cost in relation to other treatments. Thus, coupled electrocoagulation with chemical coagulation was shown to a viable alternative process suitable for the treatment of wastewater from the slaughter and processing of poultry.

The present work can be continued by analyzing the electrochemical process of electrocoagulation (EC) in the same way as the EF process was analyzed. Investigating their efficiency in reducing COD, when optimized through Doehlert planning, the operating conditions of the same: pH, time and current density. The results of both would be compared in terms of COD removal efficiency and total treatment costs, thus indicating which one would have the best cost-benefit ratio.

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