Electrocoagulation-Intensified Peroxidation of Poultry Slaughterhouse Wastewater: A Parametric Study and Process Optimization

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Abstract

In this research, electrocoagulation intensified peroxidation (ECP) using Aluminium electrode was studied as a post-treatment method for poultry slaughterhouse wastewater (SWW) with 4 operational variables (pH, current density, contact time and H_2O_2 dosage). Optimization was carried out using Response Surface Methodology (RSM). The experimental data were analyzed via analysis of variance and a second order model was developed to test the effects of process parameters on the treatment performance. The optimum conditions were chosen as; pH 5.83, 0.18 g/L H_2O_2 dosage, 58.60 min. contact time and current density 4.21 mA/cm^2. The compatibility of the predicted optimum conditions has been verified by experimental data. As a result of the experiments performed under optimum conditions, COD, TSS and color removals were found to be 97.89%, 99.31% and 98.56%, respectively. The difference between experimental and predicted values was found to be less than 0.86%. The final treated effluent met the discharge standards determined by the World Bank, EU, US and Malaysian Department of Environment for SWWs. Under optimum conditions to treat 1 cubic meter of SWW was calculated as 2.52 MYR ($ 0.58).

1.0 Introduction

Poultry slaughterhouse wastewater (SWW) was listed as the most hazardous industrial wastewater among the category of agriculture and food by the United States Environmental Protection Agency (USEPA) in the year 2004. Numerous studies reported that the poultry slaughterhouse, which contains a significant amount of organic content as a result of using water in activities such as animal slaughter and facility cleaning, is one of the sectors that produce a significant amount of wastewater (Bayar et al., 2014; Paulista et al., 2018; Awang et al., 2011; Valladão et al., 2009). The release of this kind of wastewater without proper treatment would leave detrimental effects on both health of humans and the environment. Hence, adequate treatment of poultry SWW is essential to secure the safety of human, welfare as well as the environment (Awang et al., 2011; Tong et al., 2019).

To date, several treatment approaches had been employed to purify high strength wastewater generated from various sources to meet the desired standard including biological, electrochemical and advanced oxidation process (Aziz et al., 2019; Cheng et al.,2017; Charles and Cheng 2019; Bashir et al., 2017). Among them, biological treatments such as aerobic and anaerobic processes are the common conventional treatment for poultry SWW due to its virtue of cost-effective (Bayar et al., 2014). However, biological treatment is often insufficient for poultry SWW enriched with refractory organic content due to difficulty in attaining complete stabilization of organic compounds in this process. This statement is strongly proved there is a need of a research study reported that additional post-treatment is necessary for poultry SWW to ensure the quality of final effluents in complying with the standard discharge requirement before releasing into water bodies (Bayar et al., 2014; Bustillo-Lecompte and Mehrvar 2015). Herein, an electrochemical method such as electrocoagulation was introduced as a post-treatment for poultry SWW as blood pollutants present contain electrolyte and ion will be an extra benefit to enhance the treatment performance (Awang et al., 2011; Ozturk et al., 2019).

Due to their effectiveness, electrochemical oxidation and electrocoagulation have been proposed to treat various types of wastewater in the last few years (Bashir et al., 2013 and 2019). Electrocoagulation had constantly gained popularity in various types of wastewater treatment due to their exhibited high oxidation potential, great performance in decomposing high strength wastewater, cost-effective, easy to handle and most importantly, a small amount of sludge production compared to other electrochemical methods (Bashir et al., 2016 and 2019). However, using electrocoagulation technique may require longer hydraulic retention times and higher energy consumption. Thus, there is a need to enhance and intensify the process by adding a powerful oxidizing agent such as hydrogen peroxide (Bashir et al., 2016 and 2019. Electrocoagulation intensified peroxidation can be applied to improve the activities of electrocoagulation on the removal of pollutants.

In this process, pollutants are oxidized and separated from the solution with electrocoagulation, while intensified oxidation of pollutants with highly active hydroxyl radicals (•OH) is provided by adding H_2O_2. Electrocoagulation intensified peroxidation (ECP) is a viable technology to enhance the process performance ((Bashir et al., 2016: Barrera-Díaz et al., 2008).

Recently, electrocoagulation has slowly gained the interest to be a poultry SWW treatment owing to its capability to purify contaminated water by in situ coagulant generation (Potrich, et al., 2019; Potrich, et al.,2020; Yousefi et al., 2019). Nevertheless, this is the first study to investigate and optimize the process of electrocoagulation-intensified peroxidation using an aluminum plate electrode in the post-treatment of poultry slaughterhouse wastewater.

2.0 Materials And Methods

2.1 Sample collection

Poultry SWW samples were collected from a duck producer, Dynavest Food Industries Sdn. Bhd. located at Kawasan Perindustrian Gopeng, approximately 21.4 km away from UTAR Kampar campus. It is not pure abattoir effluent as it was collected from the settling tank after the Sequencing Batch Reactor (SBR) process. On-site parameter analyzes were carried out by using CyberScan PCD 650 Multi-Parameter to test for the properties of SWW including pH, salinity, resistivity, measuring *Dissolved Oxygen* (DO) and Total Dissolved Solids (TDS). After collection and analysis, the bottles containing the samples were then transferred to the UTAR Faculty of Engineering and Green Technology Environmental Laboratory (EV-Lab) for storage. The samples were kept in the refrigerator under the temperature of 3°C to preserve the integrity of the samples and avoid the compounds present in the samples from undergoing any chemical reactions.

2.2 Laboratory-Scale set up

The experiment was conducted by the set up shown in Fig. S-1 (Supplementary material). It is made up of 2 retort stands holding the positive charge (red) and negative charge (black) crocodile clip that connecting electrodes to Direct Current (DC) power supply. To supply electricity for electrocoagulation treatment, GPS 3303 DC power supply from Gw Instek, Taiwan was used. The cathode and anode used were 2 Aluminium sheets of area 48 cm^2 (12 × 4 × 1), however, only half of it was embedded into a 500 mL sample containing beaker, thus the effective area was only 24 cm^2. Throughout the experiment run, a distance of 2 cm was maintained between electrodes to ensure good performance results due to a high reaction between coagulant species and contaminants (Hakizimana et al., 2017). Besides, to ensure homogeneity mixing of the SWW samples, Froilabo DC 300 Hot Plate Digital Magnetic Stirrer, France was used to agitate the wastewater samples at a rate of 300 revolutions per minute throughout the whole process of the experiment. Certain standard parameters are broadly used to represent the quality of SWW effluent and indirectly indicate the common substances present in the SWW. There were 6 parameters chosen for laboratory characteristic analysis of SWW which are pH, COD, Ammoniacal Nitrogen (NH_3-N), Colour, Turbidity, Total Suspended Solids (TSS) and BOD_5.

2.3 Experimental design and statistical analysis

In experiment design, a software named Design-Expert® (Version 12, Stat-Ease, Inc., Minneapolis, MN, USA) was employed for operating parameters optimization. It consists of a series of statistical and mathematical tools for building an empirical model such as Response Surface Methodology (RSM) to determine the optimum conditions where the performance of ECP is at the greatest. In this study, RSM was widely used to design the experiment by determining which input variable influences the variable of interest (Azmi et al., 2016; Ng et al.,2016). Optimization through RSM was considered a promising approach as it could provide a series of experiments to be carried out to obtain optimal response and subsequently eliminate unnecessary operating costs. Undoubtedly, RSM would result in more effective and time-saving ECP treatment system study due to the treatment performance could be improved (Bashir et al., 2019).

The operational parameters to be investigated on ECP performance were pH, reaction time, H_2O_2 dosage and current density. To get a series of experiments to be conducted using RSM, the range of the parameters were first needed to be determined. The ranges for each parameter inserted to the software are shown in Table S-2 (Supplementary material). The ranges are taken from the literature that involves treating wastewater with similar target contaminants as poultry SWW. In research by Bashir et al. (2019), the wastewater studied was POME while in literature by Barrera-Diaz et al. (2014), the wastewater treated was industrial water containing high organic pollutants. Both wastewaters were rich in organic contents which were similar to poultry SWW. Hence, the ranges are chosen from these two research studies. The generated 30 sets of experiments were carried out and responding variables were chosen to represent the efficiency of the ECP treatment such as COD, Colour, Turbidity, TSS and NH_3-N were observed and recorded. The purifying efficiency (%) of the ECP could be calculated using Eq. (1).

$$Removal efficiency = \frac{Y - Y_a}{Y} \times 100\%,$$

$$where Y = Initial Reading, Y_a = Final Reading$$

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To ensure the reliability of the results obtained, Analysis of Variance (ANOVA) was used to evaluate and analyze the experiment outcome. The response surface quadratic model developed was examined for its competency and significance by using Eq. (2):

$$Y = \beta_0 + \sum_{j=1}^{n} \beta_j x_j + \sum_{j=1}^{n} \beta_{jj} x_j^2 + \sum_{i<} \sum_{j=2}^{n} \beta_{ij} x_i x_j + \varepsilon \qquad (2)$$

where Y is the response, x_i and x_j are the operational variables, β_o is the constant coefficient, β_j, β_{jj} and β_{ij} are the interaction linear coefficient, quadratic and second-order term respectively, n is the number of operational variables and lastly, ε is a random error (Bashir et al.,2019). By referring to the Coefficient of determination, R^2 from the graph, the percentage of perfectly fit sample variation to the model's estimated data point could be known. This represented how much the experimental result data fit the model's estimated data points. Next, probability (p-value) of 95 % confidence level was tested to evaluate the model's statistical significance of each response. The best conditions under which removal percentage is the highest was then predicted by models that illustrated the respective response's interaction.

2.4 Analytical methods

On-site parameter test was done on the spot at the industry in which the fresh wastewater sample was taken by using the CyberScan PCD 650 multi-parameter. The parameters tested include pH, Temperature, Oxidation Reduction Potential (ORP), Electrical conductivity, Total Dissolved Solids (TDS), Salinity and Resistivity.

COD, TSS, colour and NH_3-N concentration were measured by using Hach DR 6000 Spectrophotometer. COD measurement is based on the Chemical-reactor Digestion Method that involved HR COD Vials (20mg/L-1500mg/L) while the Nessler method was adopted to measure the total ammonia nitrogen present. pH of poultry SWW samples collected was measured by Hanna HI 2550 model pH meter. The laboratory test for turbidity of collected poultry SWW was carried out by using HI 98703 Turbidity Portable Meter. BOD_5 indicates the amount of biodegradable organic matter present in the wastewater and it is measured by DO meter. In the measurement of BOD_5, standard method 5210B was adopted. Firstly, the initial DO of diluted wastewater samples (DO_0) was measured by using the DO meter. After 5 days, the final DO of the samples was measured as DO_5 and the reading BOD_5 was calculated by using Eq. (3). Lastly, Atomic Absorption Spectroscopy (AAS) was used to determine the amount of Aluminium ions present in SWW samples.

$$BOD_5 = \frac{DO_0 - DO_5}{DilutionFraction}$$

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3.0 Results And Discussion

The initially treated poultry SWW collected was appeared in dark brown with a layer of sludge settled down at the bottom together with some repulsive odor. All parameter tests were carried out according to APHA standard methods (ABHA 2005). Table S-3 (Supplementary material). shows the characteristic readings of poultry SWW collected. As from Table S-3, all readings had exceeded the discharged standard limit by Malaysian DOE. In measurement, the reading obtained for COD was 2625 ± 275 mg/L while for BOD_5 was 140.75 ± 9.25 mg/L. From this reading, it could be seen that COD was much higher than BOD_5, indicating that there were more substances present in a non-biodegradable form which are organic and inorganic matter. Thus, COD measurement is emphasized in this study rather than BOD_5. 30 sets of experiments generated were conducted, and removal efficiencies for each parameter are recorded in Table 1.

Table 1
Experimental Sets and Results.

Operational Variables					Responses			
Std	Initial pH	H_2O_2 Dosage, g/L	Contact Time, minutes	Current Density, mA/cm^2	COD Removal, %	TSS Removal, %	Colour Removal, %	Treated pH
8	6.00	1.00	60.00	4.00	98.2	99.25	97.49	7.04
1	3.00	0.00	10.00	4.00	89.0	99.25	98.50	4.10
29	4.50	0.50	35.00	9.50	92.3	97.00	94.85	5.84
4	6.00	1.00	10.00	4.00	97.6	98.00	98.87	6.88
10	6.00	0.00	10.00	15.00	97.8	99.75	99.39	6.99
26	4.50	0.50	35.00	9.50	92.2	97.25	95.14	5.81
18	5.25	0.50	35.00	9.50	97.1	97.00	98.34	6.33
14	6.00	0.00	60.00	15.00	98.1	99.00	98.22	7.54
25	4.50	0.50	35.00	9.50	92.0	95.50	94.73	5.72
16	6.00	1.00	60.00	15.00	98.7	97.75	98.38	7.94
28	4.50	0.50	35.00	9.50	92.2	97.50	95.14	6.00
22	4.50	0.50	47.50	9.50	95.5	97.00	95.30	6.23
13	3.00	0.00	60.00	15.00	95.0	93.50	92.50	5.10
30	4.50	0.50	35.00	9.50	92.0	96.50	94.81	5.58
23	4.50	0.50	35.00	6.75	91.0	96.75	96.15	5.88
2	6.00	0.00	10.00	4.00	93.0	99.00	97.81	7.48
11	3.00	1.00	10.00	15.00	97.6	99.25	98.62	4.21
3	3.00	1.00	10.00	4.00	96.5	99.50	98.66	3.68
24	4.50	0.50	35.00	12.25	93.6	97.75	96.27	6.17
9	3.00	0.00	10.00	15.00	96.8	99.00	98.54	6.14
20	4.50	0.75	35.00	9.50	94.3	95.75	94.81	6.16
5	3.00	0.00	60.00	4.00	95.1	96.75	96.80	4.81
21	4.50	0.50	22.50	9.50	93.4	97.75	96.72	5.61
6	6.00	0.00	60.00	4.00	98.6	99.75	99.35	6.90

Operational Variables					Responses			
12	6.00	1.00	10.00	15.00	97.9	99.50	99.11	6.46
7	3.00	1.00	60.00	4.00	96.5	98.00	97.28	4.04
27	4.50	0.50	35.00	9.50	92.2	97.25	94.65	5.72
19	4.50	0.25	35.00	9.50	90.2	98.00	97.16	6.01
17	3.75	0.50	35.00	9.50	97.1	95.75	93.39	5.82
15	3.00	1.00	60.00	15.00	98.6	90.50	87.84	5.62

3.1 Analysis of Variance (ANOVA)

The outcomes of experimental sets obtained were analyzed and interpreted by using ANOVA to test for competency and significance of the response surface quadratic model. The analysis was conducted by RSM and the results are tabulated in Table S-4 (Supplementary material). Fisher's test value (F value) is used to test the model by comparing the explained response to still unexplained responses. It is obvious from Table S-4 that the model F value for all responses refers to the model is significant. This is highlighted by the small difference between actual and predicted responses (Gunst and Myers 1996). The probability that such a large F value will occur is only 0.01% due to noise (Ozturk, and Yilmaz 2020). Another term is the p-value that represents the probability of seeing the observed F-value if the null hypothesis is true. According to the software, the desired p-value is usually less than 0.05 which is the alpha value so that the model is tested as significant. It can be seen from Table S3 that the p-value of the model for all responses is less than 0.05. R^2 is often used to represent the statistical measure of how close the experimental data are fitted to the regression model line. The perfect model fit has R^2 reading of 1 which indicates that all actual experimental data are fitted perfectly to the predicted response value (Jami et al., 2015). Based on the R^2 values in Table S-5 (Supplementary material)., it means that the model does not express only 12.02%, 10.68, 13.61 and 7.6% of the variation for COD, TSS, colour removals and treated pH values, respectively. Abbasi et al. (2020) reported that features such as the largest F value, the highest R^2 value and the smallest p-value are in the best regression model. Coefficient of Variance (C.V.), also called as relative standard deviation is widely used to represent how the data points in the data series will disperse around the mean value. Normally, acceptable C.V. reading is less than 10% which means that the variation among the responses is small and thus making the data reliable (Chowdhury et al.,2020). As can be seen from Table S4, the C.V values for all responses are less than 6% and the low C.V clearly showed that the deviations between the predicted and experimental values were low. Also, it was implied that the experiments were conducted with sufficient reliability. Furthermore, Predicted Residual Error Sum of Square (PRESS) measures how the model fits each point in the design. The obtained low PRESS (Table S-5) shows a good model fitting for all responses (Gunst and Myers 1996). In addition, adequate precision (AP) is the signal-to-noise ratio that is used to compare the useful information to the false or irrelevant data in the reading. Based on what indicated from the software, the desired value for AP should be greater than 4. This condition is satisfied for all dependent variables (Table S-5).

The predicted removal efficiency (%) for COD, TSS, colour and treated pH were calculated using the Coded Eq. (4), (5), (6) and (7). In the Eqs. A, B, C and D represent pH, H_2O_2 Dosage (g/L), Contact Time (min.) and Current Density (mA/cm^2) respectively. The purpose of the formulation is to compare the factor coefficients and identification of the relative effects of factors.

COD Removal (%)

$$= 93.06 + 0.89*A + 1.23*B + 0.82*C + 0.99*D + 13.09*A^2 – 6.26*B^2 + 2.66*C^2 – 5.95*D^2 – 0.51*A*B + 0.13*A*C – 0.42*A*D – 0.50*B*C – 0.58*B*D – 0.83*C*D \quad (4)$$

TSS Removal (%)

$$= 96.87 + 1.02*A – 0.33*B – 1.16*C – 0.65*D – 2.10*A^2 – 0.096*B^2 + 1.90*C^2 + 1.40*D^2 – 0.11*A*B + 1.11*A*C + 0.70*A*D – 0.17*B*C – 0.27*B*D – 0.92*C*D \quad (5)$$

Colour Removal (%)

$$= 95.45 + 1.35*A – 0.37*B – 1.19*C – 0.73*D – 0.13*A^2 + 0.35*B^2 + 0.43*C^2 + 1.25*D^2 + 0.19*A*B + 1.31*A*C + 0.96*A*D – 0.26*B*C – 0.28*B*D – 1.16*C*D \quad (6)$$

Treated pH

$$= 5.93 + 1.20*A – 0.19*B + 0.20*C + 0.32*D + 0.13*A*B + 0.011*A*C – 0.24*A*D + 0.24*B*C + 0.0069*B*D + 0.11*C*D \quad (7)$$

Figure 1(a), (b), (c) and (d) show the correlation between the actual value and the predicted values for the removal efficiency of COD, TSS, Colour and treated pH, respectively. It could be seen that the experimental data points are scattered around the expected outcome. Undoubtedly, this explained why the standard deviation reading showed values of 1.39 for COD, 0.90 for TSS, 1.28 for Colour and 0.35 for treated pH as tabulated in Table S-5. On the other hand, Hassan et al. (2019) reported that the random distribution observed along the zero axis in a fixed interval indicates that there is no clear model that confirms the fixed variance assumption of that model.

3.2 Effects of operational variables on COD

COD is an important parameter that needed to be observed as it contributes to most of the impurities, resulting in high pollute effects of water. The combined effects of independent parameters are shown in Fig. 2(a) contact time and current density (b) contact time and H_2O_2 dosage, (c) current density and pH. The conditions are pH of 4.50 and H_2O_2 dosage of 0.5 g/L for Fig. 2(a), pH of 4.50 and current density of 9.35 mA/cm² for Fig. 2(b), contact time of 35 min. and H_2O_2 dosage of 0.5 g/L for Fig. 2(c). As seen from Fig. 2(a), there was an improvement in COD removal when the contact time was increased from 10 min.

to 60 min. and the current density was increased from 4 mA/cm^2 to 15 mA/cm^2. The same effect of current density and contact time can be observed from Fig. 2(c) and Fig. 2(b), respectively. This situation obeyed Faraday's laws of electrolysis which stated that the generation of coagulants is directly proportional to the sum of the electricity applied through the system (An et al., 2017). Higher current density caused more production of metal ions at both anode and cathode. Subsequently, this leads to more metal hydroxide which acts as a coagulant to be produced and agglomerate the pollutants (Nasrullah et al., 2017). On the other hand, there is no guarantee that higher values of applied current density will result in higher removal efficiency. At higher current densities, the turbulence of electro-produced gases may increase, and excessively formed flocs can lead to break them (Galvão et al.,2020). Also, at higher current densities, the life of the electrodes may be shortened, and the energy consumption values of the system may increase. For this reason, it is very important to optimize the current density, which is a parameter that affects the system very much. Besides, a suitable pH is needed for good COD removal too. As seen from Fig. 2(c), COD could be removed better when pH is about 6. Although there is research by Bayar et al. (2014), stated that the poultry SWW could be treated best at lower pH. However, the content of the wastewater varies according to the source where it is obtained, making the optimum pH for them to be different too. In this study, the wastewater was obtained from the duck processing industry which was totally different from the chicken industry in the research study by Bayar et al. (2014). Until now, there is no study regarding the treatment of wastewater from the duck industry using electrocoagulation peroxidation. The statement below explains why the effluent from duck processing industry could be treated best at high pH which is different from other literature.

When electricity is applied through the system, dissolution of Aluminium occurred at anode as in Eq. (8), producing Aluminium ions (Al^{3+}) and a secondary reaction (Eq. 9) occurs at anode in the situation of sufficiently high potential. Over a wide range of pH, these Al^{3+} ions produced would undergo hydrolysis immediately to generate various monometric species such as $Al(OH)^{2+}$ and $Al(OH)_2^+$ as shown in Eq. (10) and Eq. (11) (Mouedhen et al.,2008). Al^{3+} and $Al\ (OH)^{2+}$ result from a dissolution of Aluminium anode at low pH will then be transformed into coagulants $Al(OH)_3$ which have a large surface area for the absorption of soluble organic compounds and metal ions (Eq. 12). In fact, the transformation to $Al(OH)_3$ could only occur at an appropriate pH, depending on the type of aqueous solution (Daneshvar et al.,2006; Mollah et al.,2001). Hence, an optimum pH varies according to the type of wastewater, depending largely on the content present.

$Al \rightarrow Al^{3+} + 3e^-$ (8) $2H_2O \rightarrow O_2 + 4H^+ + 4e^-$ (9) $Al^{3+} + H_2O \rightarrow Al(OH)^{2+} + H^+$ (10) $Al(OH)^{2+} + H_2O \rightarrow Al(OH)_2^+ + H^+$ (11) $Al(OH)_2^+ + H_2O \rightarrow Al(OH)_3 + H^+$ (12)

Among the operational parameters, H_2O_2 plays a large role in eliminating COD. The removal efficiency varied when H_2O_2 dosage changed. From Fig. 2(b), it could be seen that the COD removal efficiency increased when the H_2O_2 dosage was raised from 0 g/L to 1.0 g/L. Addition of H_2O_2 would lead to the initiation of AOP which is a process claimed with various degrees of effectiveness. Unlike conventional methods, AOP capable to remove a wide range of recalcitrant organic compounds, colour and turbidity (Li et al.,2019). Thus, by adding more H_2O_2, more •OH which with high oxidizing potential will be appeared during H_2O_2 reduction to oxidize the pollutants (Barrera-Díaz et al.,2014). The chain reactions that happened between organic compound (R) and hydrogen radicals are described as followed (Ozyonar et al.,2015):

$RH + OH* \rightarrow H_2O + R*$ (13)

$R * + O_2 \rightarrow ROO*$ (14)

$ROO* + RH \rightarrow ROOH + R*$ (15)

In short, COD removal highest at 60 min. of contact time, 15 mA/cm^2 of current density, pH 6 and 1 g/L H_2O_2 dosage, attaining the highest removal of 98.7% (see Table 1, std 16). Although these conditions maximize the COD removal efficiency, however, the aim of this study is to optimize the overall process performance by considering other parameters.

Accordingly, the current density applied in electrochemical systems is one of the important parameters affecting the process cost, precisely for this reason, it is not reasonable to consider these conditions (Table 1, std. 16) with a current density of 15 mA/cm² as optimum conditions. As required by multi-parameter optimization, all conditions should be considered and optimized for the whole system. At a constant contact time, current density and pH, an increment of H_2O_2 dosage from 0 g/L to 1 g/L could result in a boost of 10.3% in COD removal efficiency. On the contrary, the worst condition for COD removal was 10 min. contact time, 4 mA/cm^2 of current density, pH 3 and 0 g/L H_2O_2 dosage with removal percentage of 89% only (see Table 1, std 1).

3.3 Effects of operational variables on TSS

Total suspended solid is another parameter being studied in this research. TSS contain in most of the wastewater needed to be eliminated as it results in a high reading of colour and turbidity.

The combined effects of independent parameters are shown in Fig. 3(a) contact time and H_2O_2 dosage (b) pH and H_2O_2 dosage. The conditions are pH of 4.50 and current density of 9.50 mA/cm² for Fig. 3(a), contact time of 60 min. and current density of 15 mA/cm² for Fig. 3(b). Figure 3(a) shows a reduction in the treatment of TSS despite the increment of contact time from 10 min. to 60 min. High TSS reading showed during longer contact time attributed to a large amount of Aluminium hydroxide produced as described in Eq. (8) to Eq. (12) (Amarine et al., 2020). Longer contact time would produce more Al^{3+} and OH^- ions, subsequently, resulted in more formation of Aluminium hydroxide. Initially, Aluminium hydroxide present as a grey colour floc suspended in the solution before settling down to the bottom. However, if the flocs are not large and dense enough, it will remain dispersed and contribute to the cloudy properties of the solution.

When the pH is about 6, it can be said that it provides the desired condition for TSS removal (Fig. 3(b)). However, to give a clear value for the optimum value of the pH, a complete optimization was determined by numerical and graphical optimization for all parameters. According to Kobya et al., (2003) and Hernandez et al., (Linares-Hernández et al.,2009), excellent treatment performance will be observed when the pH is below 8. On the contrary, pH greater than 10 would undergo a decrement in treatment performance. This is because the predominant Aluminium chemical species, $Al(OH)_3$ would be presented as coagulants to trap the colloidals at pH 4-9.5. Contrarily, when the pH is greater than 10, there will be other Aluminium complexes such as $Al(OH)_4^-$ species present which is incapable of removing contaminants (Jotin et al.,2012).

On the other hand, no significant increase (approximately 1%) in TSS removal was observed when the H_2O_2 dosage increased from 0 g/L to 1.0 g/L. A high dose of H_2O_2 should speed up the elimination process of TSS. This might due to the amount of coagulants, $Al(OH)_3$ formed was enough to remove the TSS present without the need of H_2O_2 in this case. In TSS removal, two conditions maximize the efficiency which were at pH 6, 0 g/L H_2O_2 dosage, 10 min. contact time, 15 mA/cm^2 current density and pH 6, 0 g/L, 60 min. contact time, 4 mA/cm^2 current density respectively. This implied that long contact time may require a low current density to achieve good removal efficiency, while short contact time is sufficient at high current density.

3.4 Effect of operational variables on colour

River contamination due to impropriate removal of colour by various industries has become a serious issue nowadays. Hence, wastewater should be discharged with colour removed so that it will not affect the aesthetic and clarity of the river. The combined effects of contact time and H_2O_2 dosage are shown in Fig. 4(a) and (b). The conditions are pH of 3.00 and current density of 4 mA/cm^2 for Fig. 4(a), pH of 6 and current density of 4 mA/cm^2 for Fig. 4(b). From Fig. 4, higher pH was claimed to have better colour removal than lower pH. It is obvious from Fig. 4 that there was poor colour removal efficiency when the value of pH was 3 while the removal was improved with a gradual increase from pH 3 to pH 6. The removal reduction is due to when it is in acidic condition, collapsing hydroxide ions generated at the cathode by protons would occur, resulting in the insufficient formation of coagulants, Aluminium hydroxide for pollutants agglomeration (Bashir et al.,2013). Thus, better treatment was obtained at high pH 6 compared to pH 3.

The combined effects of current density and H_2O_2 dosage are shown in Fig. 5(a) and (b). The conditions are pH of 4.46 and contact time of 10 min. for Fig. 5(a), pH of 4.46 and contact time of 60 min. for Fig. 5(b). Differently, Fig. 5 depicts that high current density enhances the clarity of wastewater. This phenomenon could be explained by the fact that high current density leads to an increase in the dissolution of Aluminium anode, causing more precipitate for the removal of contaminants that contribute to colour property. Also, colour removal by H_2 flotation could occur at a faster rate when bubbles are generated more rapidly and with smaller size when high current density is applied. Bubbles with smaller sizes were good in assisting colour removal as they were reported to be more efficient in trapping pollutants compared to large bubbles size (Kobya et al., 2006). In the study of contact time, colour treatment experience a decrement trend when the contact time was slowly increased. This statement could be proved by that when at constant pH, H_2O_2 dosage, and current density (pH 6, 1 g/L and 4 mA/cm^2), an increase in contact time caused a 1.38% reduction in colour removal. On the other hand, addition of H_2O_2 did not result in much difference in treatment performance. As mentioned earlier, this might be due to high current density which cause more formation of coagulants which could reduce colour to a certain extend. Hence, there is no H_2O_2 required for colour removal when high electricity is employed. From the result, colour could be eliminated maximum at optimal condition of pH 6, 0 g/L, 10 min. contact time and 15 mA/cm^2 of current density which fulfills the above explanations that colour treatment work best at alkaline condition, low contact time and high current density.

3.5 Effect of operational variables on treated pH

From the results as shown in Table 1, the final pH of all sets increases at all initial pH value. For every set of experiments, the final treated pH was always larger than the initial pH. Same results were obtained in a previous research by Bayar et al. (2014). It was observed that the larger the initial pH, the larger of the final treated pH. pH tends to rise rapidly for low initial pH, while rise more slowly for high initial pH. The combined effects of pH and current density are shown in Fig. 6 (a) and (b). The conditions are H_2O_2 dosage of 1 g/L and contact time of 10 min. for Fig. 6 (a), H_2O_2 dosage 1 g/L and contact time of 60 min. for Fig. 6 (b). From Fig. 6, it could be noted that pH tends to increase when longer contact time. The increase in pH of the wastewater after treatment was due to more OH^- ions were produced from hydrogen evolution reaction at the cathode when there were longer contact time and higher current density, resulting in alkaline properties that increase the pH value of the wastewater. Similar comments were reported in the literature (Jotin et al., 2012; Kobya et al., 2006). From this, it was proven that electrocoagulation peroxidation using Al electrodes would possess the virtue of pH adjustment mitigation for reuse purposes due to its pH neutralizing property (Nagaraju et al., 2006). According to Malaysian DOE, the final effluent is only allowed to discharge at pH 5.5 to 9.0. In this study, the final pHs obtained for all sets of experiments were within the discharge standard issued by the government.

3.5 Optimization and verification for ECP

One of the main vital objectives of using Design-Expert® is optimization which involves obtaining optimum conditions for the whole system (Azmi et al., 2016; Ng et al., 2016). Traditional graphical optimization that provides results in the form of overlay contour plots for system optimization and numeric optimization were used together, so it was possible to predict the solution when varying independent parameters. Undeniably, with optimum conditions, there would be a substantial reduction in unnecessary chemical used, duration and subsequently saving operating cost. After 30 sets of experiments generated by Design Expert® were carried out, several optimum sets were produced with numerical optimization according to the result inserted. However, in the selection of optimum conditions, the focus has been on conditions that maximize all pollutant parameters and minimize energy consumption. The parameter that has a significant role in energy consumption is the current density.

Then, other parameters that affect the system cost, such as H_2O_2 dosage and contact time, were focused on. While it is possible to minimize and maximize target parameters in numerical optimization, options such as target setting, value fixation and range determination are possible. Thus, in optimization, it is possible to direct the optimization in line with the experimental experience and foresight of the designer. In this study, all parameters were assigned with the 'in range' option. Since the optimum value of the pH value is close to 6.0, the range was determined as 5.8-6. The range that is close to the optimum value of H_2O_2 dosage and requires minimum usage to reduce system cost was determined as 0.18–0.25. Since the contact time is estimated to be around 60 min. from the experimental data, the range was determined as 58-59.9; The current density was determined to be in the range 4.2–4.23 based on both experimental data and to reduce system cost. All responses are assigned the same with 'in range'. The optimum independent variables obtained from numerical optimization are; pH: 5.83, H_2O_2 dosage: 0.18 g/L, contact time: 58.60, current density: 4.21 mA/cm². Under these conditions, the system predicts that the dependent variables will be COD: 98.74%, TSS: 100%, Color: 98.90% and the final pH value 6.88.

A practical visual examination of the field of optimum response values in the field of parameters to select the optimum combination of parameters is possible with the overlay plots (Nagaraju et al.,2019). Figure S-6 (Supplementary material) shows the area of optimum response values of optimum dependent variables in factor space. Figure S-6 shows the area of optimum response values of optimum dependent variables in factor space. Fig S-6 (a), pH and H_2O_2 dosage; Fig S-6 (b) pH and contact time, Fig S-6 (c) Current density and contact time overlay graphs showing the effect of them on response areas. In Fig S-6 (a), X1 shows the pH, X2 shows the H_2O_2 dosage, while the contact time and current density are 58.60 min. and 4.21 mA/cm², respectively. Fig S-6 (b), X1 shows pH, X2 shows the contact time, while the H_2O_2 dosage and current density are 0.18 g/L and 4.21 mA/cm², respectively. In graphical optimization, regions that do not meet the optimization criteria are shaded gray. Any "window" that is not shaded in gray meets the target of each response. Considering the optimization of the combination of parameters in contours where critical response contours overlap, it is seen that the optimum system conditions are pH: 5.83, H_2O_2 dosage: 0.18 g/L, Current density: 4.21 mA/cm² and contact time: 58.60 min. Under these conditions, the system predicts that the dependent variables will be COD: 98.73%, TSS: 100%, Color: 98.90% and the final pH value 6.88. As seen graphical optimization values and numerical optimization values are almost the same. Similar observations were reported in the literature (Powar et al.,2019; Bayramoglu et al., 2006; Singh et al., 2008). Experiments were carried out under these optimum conditions to the validation of the optimized parameters and the results obtained are shown in Table 2. From Table 2, it could be observed that there were only small differences between the predicted removal and the actual removal for all responses. All experimental removal readings were slightly lower than the predicted value obtained by the software.

Table 2
Optimization and Verification for Optimum Condition.

Responses	Predicted Removal, %	Actual Removal, %	Percentage Difference, %	Final Reading	Discharge Standard
COD	98.74	97.89	0.86	53	100
TSS	100	99.31	0.69	16	100
Colour	98.90	98.56	0.34	5.08	200
Final pH	6.88	6.84	0.58	6.84	5.5-9.0
Desirability	1				

The condition selected giving desirability of 1, meaning that all conditions were fulfilled to the goal set. Although there were some minor discrepancies between the predicted value and actual reading, however, effluent characteristics (Table 2) such as COD (53 mg/L), Colour (16 mg/L) TSS (5.08 mg/L) and final pH(6.84) met the discharge standards determined by World bank, EU, US (Bustillo-Lecompte and Mehrvar, 2015) and Malaysian DOE for SWWs.

3.6 Cost estimation

Cost estimation was made based on the amount of chemical and energy used during treatment to ensure the cost-effectiveness of the treatment process in an industrial application. The amount of energy spent to treat 1 cubic meter of wastewater was calculated from Eq. (16), the cost calculation resulting from the use of H_2O_2 from Eq. (17), and the total system cost from Eq. (18).

$$E\left(\frac{kWh}{m^3}\right) = \frac{VxIxt}{v}(16)$$

$$CostofH_2O_2\left(\frac{MYR}{m^3}\right) = costofH_2O_2(\$)xMalaysiarate(17)$$

$$TotalCost\left(\frac{MYR}{m^3}\right) = ExTariffrate + CostofH_2O_2(18)$$

V (volt) potential difference in the system, I (Ampere) express the current applied to the system, t states the reaction time (min), v (m³) symbolizes the wastewater volume in the reactor. Usually, the category of customers and the supply voltage determine the rate of electricity charges. Tenaga National Berhad (TNB) is a Malaysian multinational electricity company and according to TNB the wastewater treatment plant is classified as a medium voltage commercial plant. According to the medium voltage commercial tariff rate received from TNB (Tenaga Nasional Berhad 2020) electricity is charged at 0.365 MYR/kWh. For H_2O_2 price estimation, the cost of industrial-grade H_2O_2 was $ 445, taken from the average between $

390–500 per ton (Asghar, et al., 2015). After conversion, it is $ 0.64 per liter H_2O_2. Under optimum conditions (pH 5.83, H_2O_2 dosage: 0.18 g/L, contact time: 58.60 min, current density: 4.21 mA/cm^2) energy consumption to treat 1 cubic meter of SWW was calculated as 1.42 kWh. The cost of electricity based on energy consumption values was found as 0.52 MYR/m³. The cost of H_2O_2 use was determined as 2 MYR/m³. The total cost for treating 1 cubic meter of SWW was found as 2.52 MYR ($ 0.58). Electrochemical technologies are frequently preferred in the treatment of industrial wastewater. One of the most important factors limiting the application of electrochemical processes is cost. Another factor is whether discharge/reuse standards are met. Therefore, we can say that sufficient treatment and low cost affect the preferability of the electrochemical process. Table 3 gives a comparison of COD removal efficiency and system cost for SWWs.

Table 3
A Comparison of the literature on COD removal efficiency and system cost of Electrochemical processes.

Treatment	Slaughterhouse Wastewater Type	Electrode	COD Removal Efficiency	Cost	References
Electro coagulation	Poultry	Aluminium	92%	0.7 $/m^3	[45]
Electro Coagulation + Flotation	Swine	Aluminium	85 %	1.03 $/m^3	[49]
ultrasound-assisted electrocoagulation-flotation	Swine	Aluminium	86.9 %	0.74 $/m^3	
Electro coagulation	Pig	Aluminium	81.01%	4.28 $/m^3	[50]
Electro coagulation	Poultry	Aluminium	65%	3.89 $/m^3	[16]
Electro coagulation + peroxidation	Poultry	Aluminium	97.89%	0.58 $/m^3	**This Study**

It is clear from Table 3 that electrochemical technologies are successful in COD removal in SWW treatment. Although there is secondary waste generation due to the nature of the process in the electrocoagulation technique, it is preferred due to the low amount of waste generated, high pollutant removal efficiency and economical energy consumption values (compared to processes such as electrooxidation). Ozturk and Yilmaz (2019) used Ti/Pt electrodes to purify cattle-SWW in their studies. While providing 92.2% COD removal, they determined the energy consumption values as 153.57 kWh/m³ (0.45 kWh/m³ in this study). As can be seen, the electrocoagulation process is advantageous in terms of

COD removal efficiency and energy consumption. As can be seen from Table 3, low energy consumption is possible in processes where electrocoagulation is applied. Another factor contributing to this situation is the high electrical conductivity of blood-containing wastewater. Blood can transmit electric current thanks to the electrolytes it contains. Therefore, electrochemical treatment of multi-component SWWs (because of activities such as animal cutting, chopping, intestinal washing) may be possible without using support electrolyte. This was proven with this study. As a result, although H_2O_2 was added, it certainly helped to improve purification performance without being more expensive than the traditional electrocoagulation methods.

4.0 Conclusion

In this research study, ECP had proved to have a great performance on the treatment of poultry SWW. Performance on the removal of each parameter varied according to different operational variables. ECP process performance was high for TSS and color removals at a high current density and low contact time; For COD removal, was high at high current density, long contact time and high H_2O_2 dosage. Optimum conditions for all dependent parameters were determined for pH, H_2O_2 dosage, contact time and current density as 5.83, 0.18 g/L, 58.60 min. and 4.21 mA/cm², respectively. The compatibility of the predicted optimum conditions has been verified by experimental data. Under optimum conditions the removal efficiencies for COD, TSS and colour were found as 97.89%, 99.31% and 98.56%, respectively. Also, the final treated effluent had all readings complied with the standard discharge limit by the World Bank, EU, US and Malaysian DOE for SWWs. The total cost to treat 1 cubic meter of SWW was found as 2.52 MYR ($ 0.58). Besides, this is a new research study that performs electrocoagulation intensified peroxidation treatment on poultry SWW. Hence, ECP is a treatment method that worth to invest and further research should be conducted on a large scale since it was proven to be an effective post-treatment method.

Abbreviations

AOPs:Advanced Oxidation Processes

ANOVA: Analysis of Variance

BOD_5: Biochemical Oxygen Demand

COD: Chemical Oxygen Demand

C.V: Coefficient of Variance

DC: Direct Current

DO: Dissolved Oxygen

DOE: Department of Environment

ECP: Electro-Coagulation-Peroxidation

ORP: Oxidation Reduction Potential

POME: Palm Oil Mill Effluent

RSM: Response Surface Methodology

SBR: Sequencing Batch Reactor

TDS: Total Dissolved Solids

TSS: Total Suspended Solid

USEPA: United States Environmental Protection Agency

Declarations

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Competing interests

The authors declare that they have no competing interests

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Yoke Peng Toh: Writing- Original draft preparation,Investigation,Methodology,**Mohammed JK Bashir:**Supervision, Conceptualization,Methodology,Writing- Reviewing and Editing, **XinxinGuo:**Investigation,Methodology, **Lai Peng Wong:**Investigation,Methodology, Analysis,

DilaraOzturk: Visualization,Data analysis, Cost analysis,**Salem S. Abu Amr:** Visualization, Reviewing and Editing,Validation, **Lim Jun Wei:**Writing- Reviewing and Editing.

Conflicts of interest

The authors declare that they have no conflict of interest.

Availability of data and material

(Not applicable)

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Figures

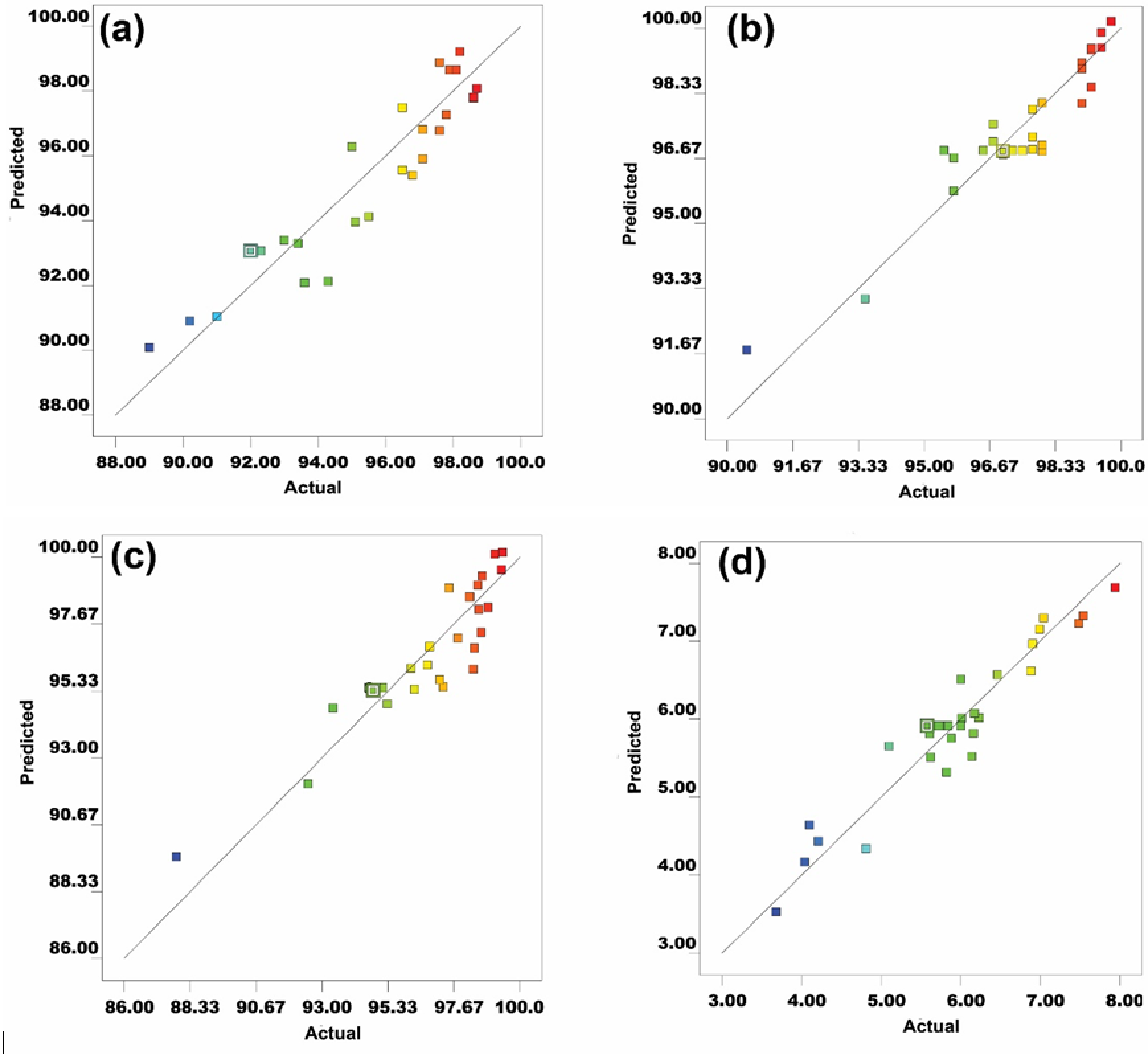

Figure 1

Correlation Relationship between Predicted and Actual Value for Removal of (a) COD, (b) TSS, (c) Colour and (d) Final pH

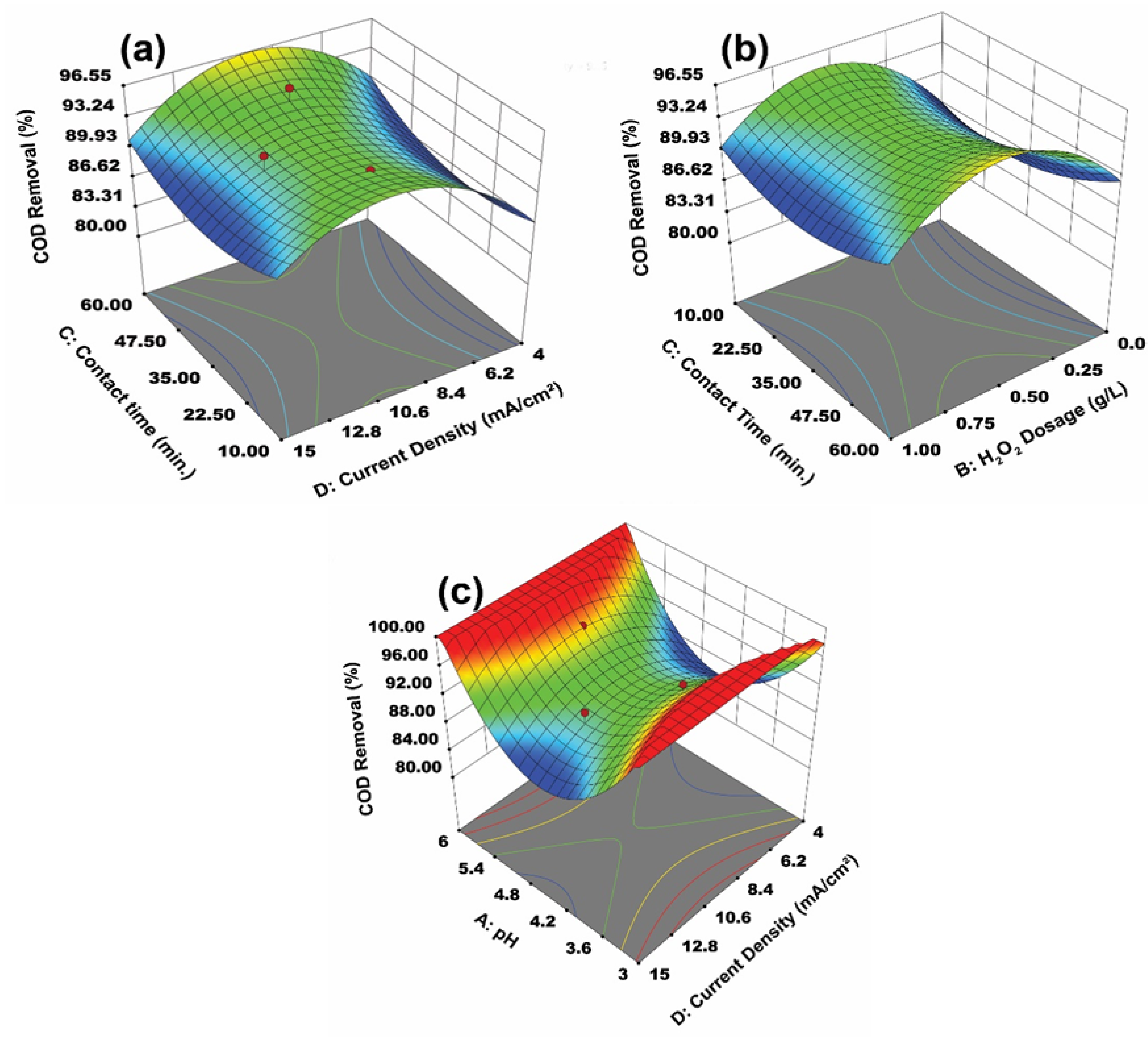

Figure 2

3D Surface plot model and combined effects of (a) contact time and current density, (b) contact time and H2O2 dosage, (c) current density and pH on COD removal ((a): pH:4.50, H2O2 dosage: 0.5 g/L; (b): pH:4.50, Current density: 9.35 mA/cm²; (c): Contact time: 35 min. H2O2 dosage:0.5 g/L)

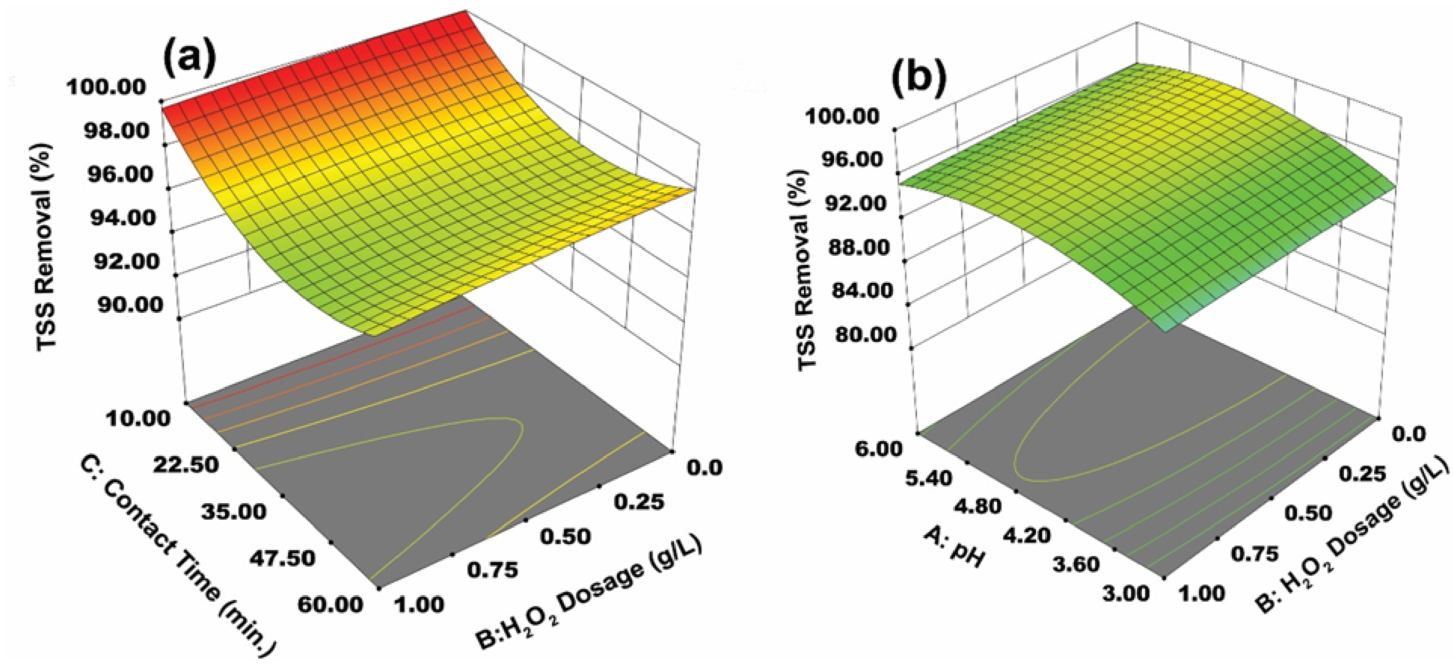

Figure 3

3D Response surface model and combined effects of (a) Contact time and H2O2 dosage, (b) pH and H2O2 dosage on TSS removal ((a): pH: 4.50, current density: 9.50 mA/cm²; (b): Contact time: 60 min., current density: 15 mA/cm²)

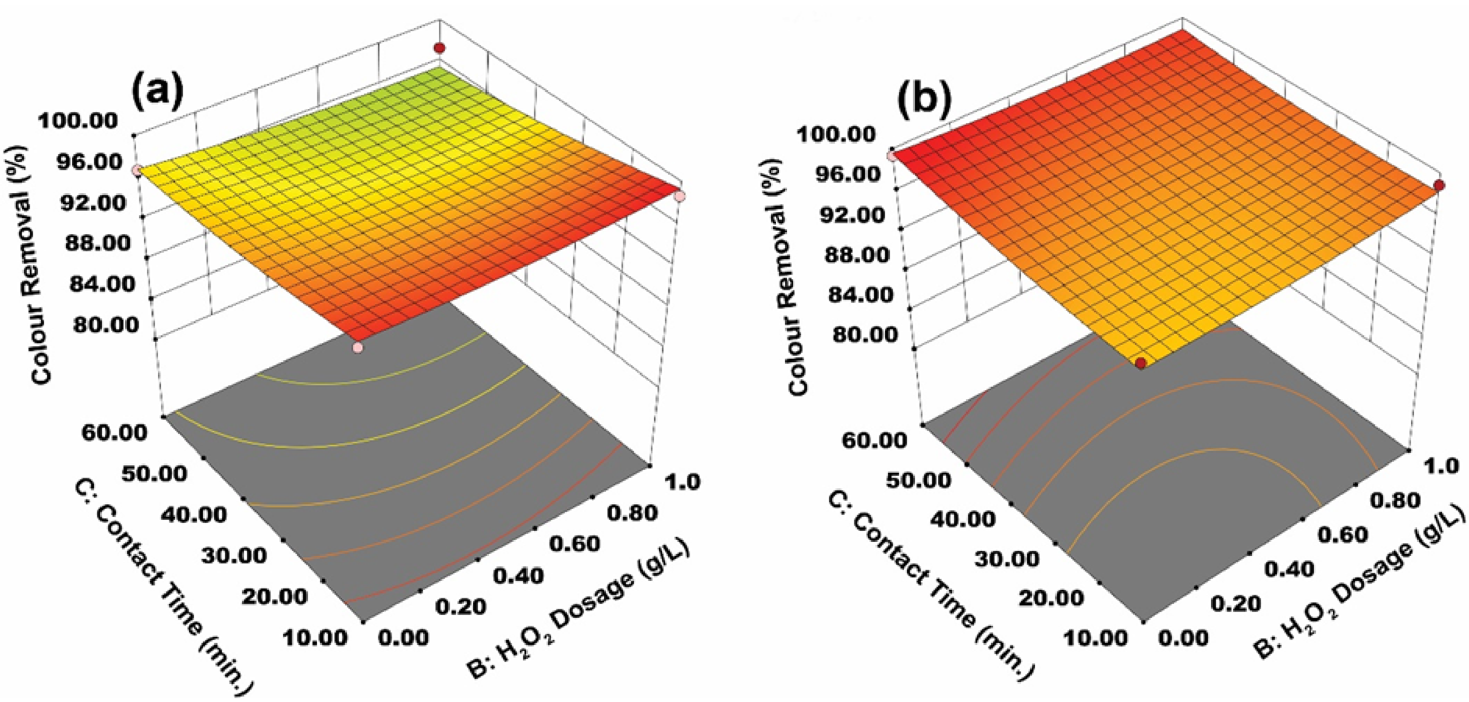

Figure 4

3D Response surface model for colour removal at (a) pH 3, (b) pH 6 ((a): pH: 4.46, contact time: 10 min.; (b) pH: 4.46, contact time: 60 min.)

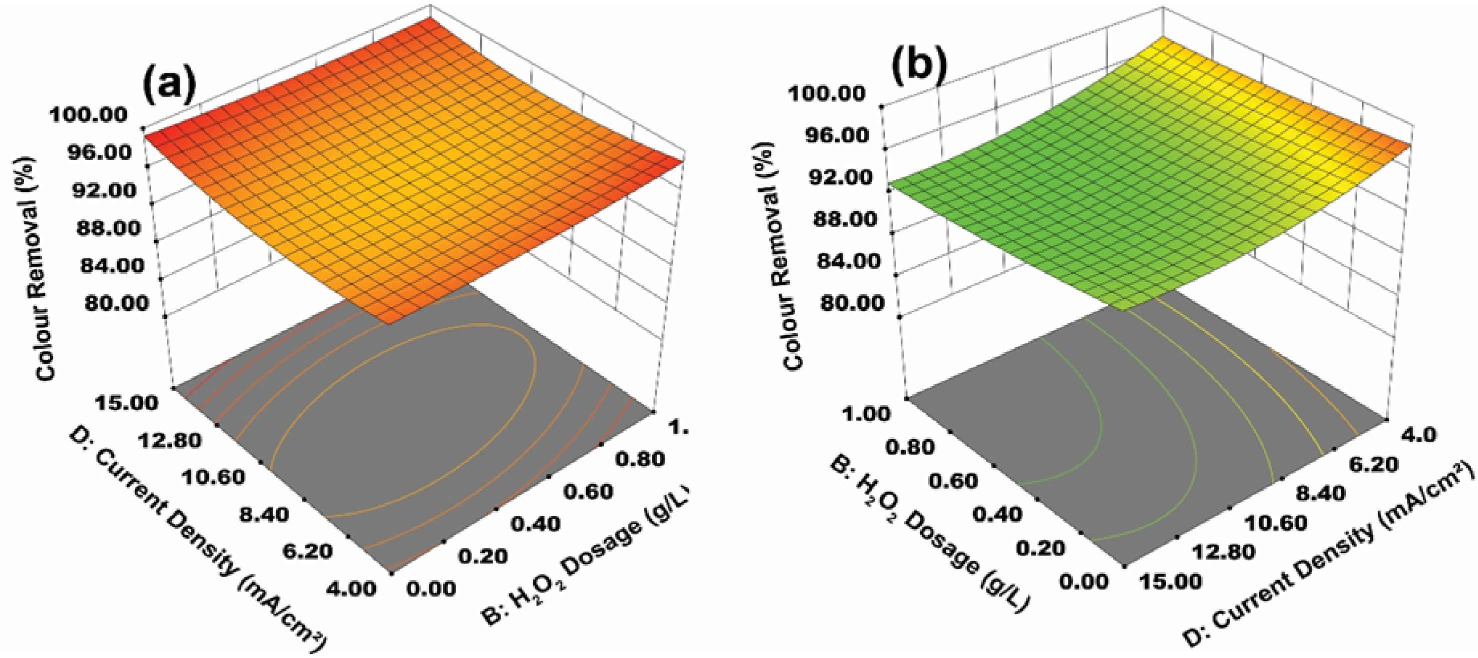

Figure 5

3D Response surface model for colour removal at contact time of (a) 10 min, (b) 60 min.(a): pH: 4.46, contact time: 10 min.;(b): pH: 4.46, contact time: 60 min.)

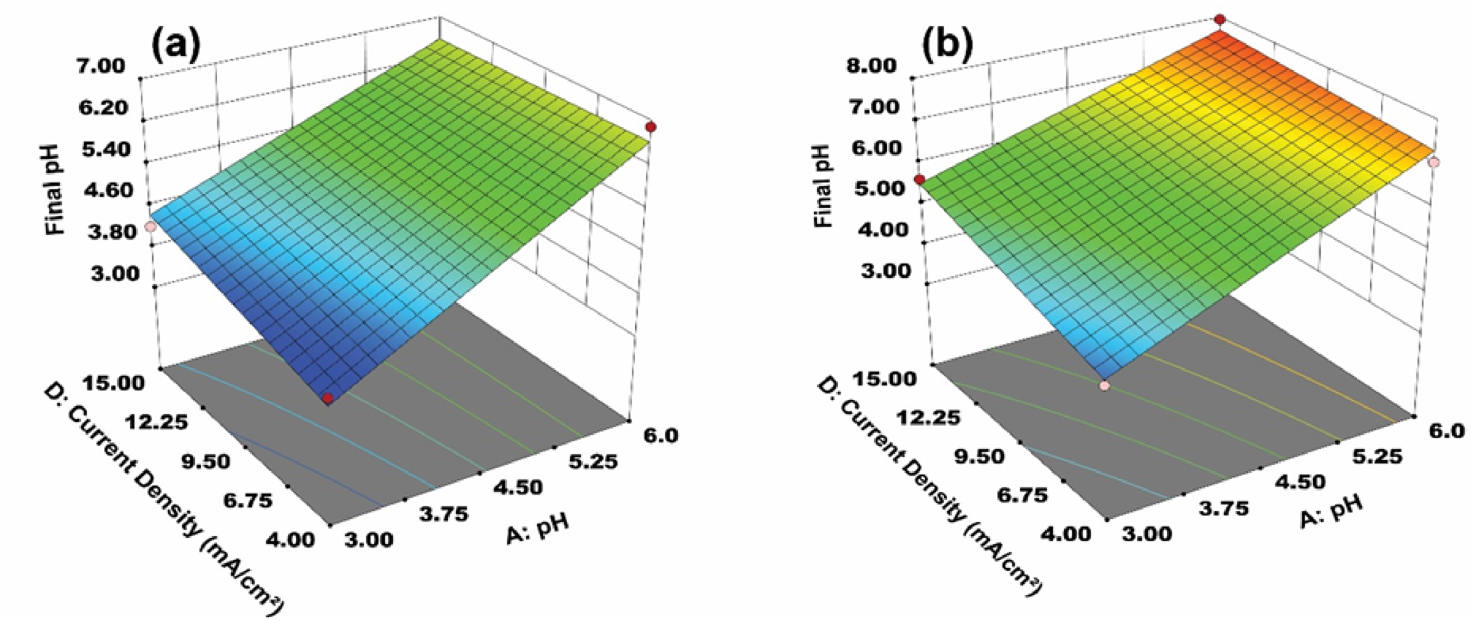

Figure 6

3D Response surface model for treated pH at contact time of (a) 10 min, (b) 60 min. ((a): H2O2: 1 g/L, contact time: 10 min.; (b): H2O2: 1 g/L, contact time: 60 min.)

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- SupplementaryMaterial.docx