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ORIGINAL RESEARCH ARTICLE

REDUCTION OF COLOUR FROM INDIGO DYE-RICH WASTEWATER BY ELECTRO-COAGULATION USING IRON (Fe) AND ALUMINUM (Al) ELECTRODES

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ABSTRACT

Indigo dyes are important substances in the Adire Textile industry which are not environmentally friendly because they produce colored effluent. This study evaluated the performance of a batch-scale electro-coagulation (EC) technique for Indigo dye-rich wastewater treatment. A pilot-scale EC unit using Iron (Fe) and Aluminum (Al) electrodes, having a mono-polar parallel arrangement with a constant inter-electrode distance of 10 mm was developed. The Fe anode and Al cathode had effective areas of 16.50 and 22.75 cm^2 respectively. The impact of pH, Sodium Chloride (NaCl), and electrolysis time on EC performance was assessed by calculating the color reduction. A three-factor three-level Box Benkhen Design with three center points was used to generate fifteen experimental runs using NaCl, pH, and electrolysis time as independent variables and color removal efficiency as the response variable. A regression analysis was performed after the results of the experimental data were fitted to a full quadratic model resulting in the generation of regression coefficients. The model was inversely transformed and modified to make it significant. The results show that the optimum pH and NaCl concentration were 3 and 4 g/l respectively with electrolysis time of 35 minutes. Under these optimum conditions, the percentage of colour removal was 32.78% with a predicted value of 80%. An R^2 value of 83.45% was obtained and ANOVA verified that the accuracy of the proposed polynomial model is acceptable. Therefore, it is noteworthy that an EC technique could be used in reducing color from Indigo Dye-rich wastewater in textile industry.

1.0 Introduction

Adire is an integral part of the Egba culture in Abeokuta, Ogun state, Nigeria. The use of this traditional 'Tie and Dye' has crept into offices and schools where most schools within and outside Abeokuta uses it as Friday and occasional wears. Textile industries are water intensive and Dye-rich effluent discharges from the Adire manufacture flow into surface and groundwater, causing pollution. Conventional textile dyeing used between 120 and 180 litres of water for every 1 kg of fabrics to be dyed (Karthik and Rathinamoorthy, 2015). The color of dye lowers the dissolved oxygen level in rivers and streams by inhibiting sunlight penetration, thus, destroying aquatic life (Durotoye *et al*., 2018; Elango *et al*., 2017; Forgacs *et al*., 2004; Kaur *et al*., 2010; Robinson *et al.,* 2001). Additionally, effluent from Adire textile industry also causes environmental problems which can affect human life (Rajendran, 2018).

These make the treatment of dye-rich wastewater of necessity to protect public health and the environment.

To treat the effluent generated, several methods have been adopted in the treatment of dye-rich wastewater to reduce or eliminate the associated environmental and ecosystem problems. These include physico-chemical, chemical, advanced oxidation, microbiological, enzymatic decomposition and electrochemical methods such as: Adsorption with inorganic and organic supports, coagulation by aluminum, lime, or iron salts, ion exchange and filtration, subsurface flow constructed wetlands, wet electrolytic oxidation and EC (Badejo *et al*., 2012; Carlos *et al*., 2009; Forgacs *et al*., (2004); Martínez-Huitl and Brillas, 2009; Robinson *et al.,* (2001**);** Dai *et al*., 2006). EC treatment technique is the most widely accepted because of its cost efficiency and relatively high removal capacity and low sludge generation (Bharath *et al*., 2018).

EC is the process of dissolution, coagulation and flocculation in the presence of electricity (Moussa *et al.,* 2017). According to Martínez-Huitle and Brillas (2009), EC technique combines coagulation, floatation and electrochemistry for water treatment, it provides an alternative to the use of polymers and polyelectrolyte coagulants for breaking stable emulsions and suspensions. EC and coagulation/flocculation works on the same principle by removing pollutants through destabilizing the repulsive forces that keeps the pollutants suspended in water. Neutralizing the repulsive forces brings about the formation of flocs that can be easily removed after sedimentation. The coagulants in the EC technique are generated by electrolytic oxidation as against the use of chemical coagulants in the coagulation/flocculation process. It is also efficient in treating oily water and has wide applications as compared to other wastewater treatment methods (Abbass *et al*., 2018). EC has been used for various wastewater treatments such as municipal and industrial (including textile, tanneries, pulp and paper, oily and food processing industry) wastewater (Islam, 2019). Bazrafshan *et al*., (2013) observed that the technology removed approximately 98% of COD, BOD, TSS and bacterial indicators on diary wastewater. EC can also be employed in conjunction with the conventional method of dye-rich wastewater treatment.

Furthermore, Zailani *et al*. (2018) reported that aluminum electrode had higher removal efficiencies than Iron electrodes in stabilized leachate treatment. Similarly, Manilal *et al.* (2017) and Nasrullah *et al.* (2012) found aluminum to be the superior Iron and Steel in sewage treatment while Eyvaz *et al*. (2009) found alternating pulse current to be superior to direct current for pollution removal in disperse dyes.
Photo-Fenton and EC processes were integrated in removing organic and inorganic pollutants from industrial effluent in dye-rich wastewater treatment by Módenes *et al.* (2012). EC has also been combined with electro-oxidation techniques and ozone processes in the treatment of highly coloured and polluted industrial wastewater (Barrera-Díaz *et al*., 2003; Bernal-Martínez *et al*., 2010).

Also, Response Surface Methodology (RSM) a collection of mathematical and statistical techniques was adopted. RSM is useful for developing the empirical model building, improving and optimizing processes parameter; it is also used for finding the interaction of several affecting factors (Azargohar and Dalai, 2005). Similarly, RSM uses quantitative data from the related experiment to determine regression model and optimize an output variable which is influenced by several input variables (Behera *et al*., 2018).

The technological advancement in man's activities brought about by industrial revolution has resulted in effluents from industries channeled without proper treatment. There is therefore, the need for a wastewater recycling technique to avoid scarcity of clean water. This study focused on optimizing the EC process using Fe and Al electrodes having mono-polar parallel arrangement for colour removal in Indigo dye-rich wastewater with the RSM.

2. Materials and Methods

Indigo dye-rich wastewater was obtained from a local Adire market at Asero, Abeokuta Ogun state, Nigeria. The pH, conductivity, total dissolved solids, dissolved oxygen, temperature and Color were measured using standard methods (APHA, 2005), before and after the experiment to determine the treatment efficiency of the EC process. All experiments were conducted in triplicate. Using RSM, experiment was run 15 times using Box-behnken design. The total volume of raw dye waste-water used was 22.5 litres, with 1.5 litres per run.

2.1 Experimental Set-up

The Reactor was made of transparent cylindrical plastic container (230 x 160 x 110 mm) with an effluent tap. Mono-polar parallel arrangement was obtained with two Fe (30 x 80 x 1 mm) and two Al (50 x 50 x 0.5 mm) electrodes connected in parallel (10 mm distance apart) and circuit completed with two 12 volts battery connected in series to supply power to the whole set-up with a stirrer to achieve uniform mixing (Figure 1). Multi-meters were placed at strategic position to measure the input voltage and current. Distilled water was used for washing the electrodes after each run while H_2SO_4 and NaOH solutions were used for adjusting the pH of the solution.

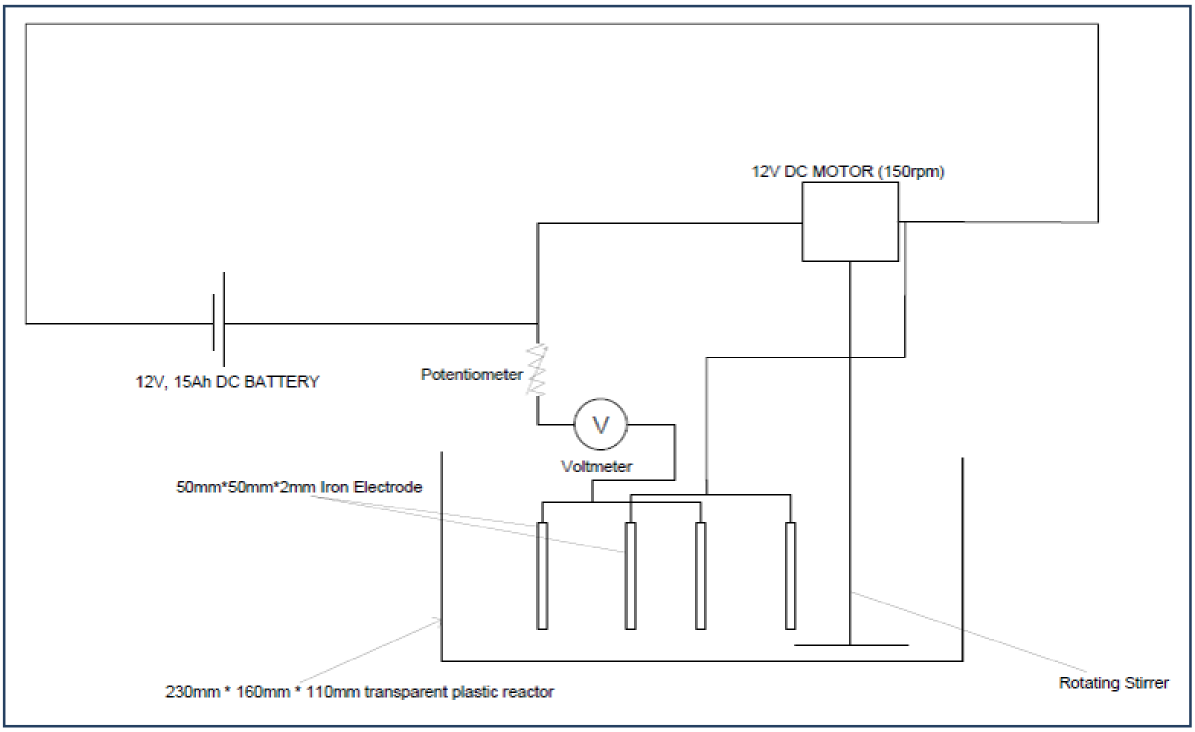


Figure 1: Experimental Setup

Based on RSM, 15 runs of experiment were performed using the Box-Behnken Design (BBD). The BBD has 3 levels (-1, 0 and +1) for the design matrix to observe the effect of the treatment parameters (pH, NaCl and electrolysis time) on colour removal efficiency by the EC process. The significance and adequacy of the predicted model was determined by ANOVA. The statistical significance was verified by the F-test which uses F-value that describes the variation in the mean of the data. The accuracy of the fitted polynomial model was determined by the coefficient of R^2. The model was transformed according to the diagnostic information obtained from the former model.

The pH, NaCl and electrolytic time were kept in the range of 3 - 10, 0 - 4 mg/l and 10 - 60 minutes respectively in this experiment. The coded and actual values of the variables of the design of experiments for the overall EC optimization are shown in Table 1. For a typical run, 1.5 L of the raw wastewater was poured into the reactor after all the initial tests. The initial pH was adjusted using H_2SO_4 and NaOH solutions to reach the desired values for each run.

The effluent collected at each run was allowed to settle before the final test is done. After each run, the electrodes and the reactor were washed with distilled water to avoid errors as residues on treated wastewater from the former run may affect the characteristics of the next run thereby leading to false results. After the electrocoagulation reaction is completed, the wastewater was left to naturally precipitate for 30 minutes for the removal of flocs formed during the process, and the various analyses were carried out with the supernatant. Design Expert 11.0 software was used to investigate the effect of chosen treatment/operating parameters in the EC process of the dye by optimizing experimental conditions in the RSM category. The response surface plot was carried out to check the influence of three operating parameters on colour removal.

Table 1: Variables and their levels

Factor	Independent process variable (unit)	Coded levels of variables		
		-1	0	+1
X_1	Electrical Conductively EC (mg/l)	0	2	4
X_2	pH	3	7	11
X_3	Electrolysis time (mins)	10	35	60

The influence of the process variables was evaluated by the Color removal efficiency of the process. The center points were used to evaluate the experimental error and the replicability of data. The RSM provides that it is possible to indicate independent process variables quantitatively with equation 1:

$$y = F(x_1, x_2, x_3, \dots, x_n) \pm \varepsilon \qquad (1)$$

where:
y is the response (efficiency of colour removal),
F is the response function,
ε is the experimental error, and
x_1, x_2, x_3, …, x_n are independent variables.

3. Results and Discussion

The results obtained from this study show good correlation between actual and predicted responses as shown in Table 2. The experimental inputs for the various factors in each of the 15 runs with their corresponding colour removal response as shown in Table 2 for both the experimental and predicted values confirmed the reliability of the model. The inverse transformation has been applied on both the experimental and predicted values respectively.

Table 2: Colour Removal Responses of Dye-rich Wastewater for Experimental and Predicted values

Run	R Independent Variables			Colour Response Values (%)	
	NaCl Concentration (EC) (g/l)	pH	Electrolysis Time (mins)	Experimental	Predicted
1	4	3	35	0.0305	0.0398
2	4	11	35	0.0329	0.0608
3	2	7	35	0.1592	0.0909
4	0	7	10	0.2653	0.2486
5	2	3	60	0.2252	0.2221
6	0	11	35	0.1206	0.1420
7	0	3	35	0.1057	0.1211
8	2	7	35	0.0458	0.0909
9	0	7	60	0.1379	0.1473
10	4	7	60	0.1712	0.1804
11	2	11	10	0.2193	0.2300
12	2	11	60	0.1209	0.1055
13	4	7	10	0.0700	0.0530
14	2	7	35	0.1416	0.0909
15	2	3	10	0.0486	0.0716

3.1 Model Fitting

The Box-cox plot indicated that the current power of the model (y^{λ}) was 1 (Figure 2). This represented no transformation of the actual response data (y^{1}). The area bounded by the red line indicated the 95% confidence interval (CI) beyond which the model was not significant. The 'power' at the green line (-1) that is, the inverse transformation was used since it falls within the 95% CI. The green line is often called the minimum model residual.

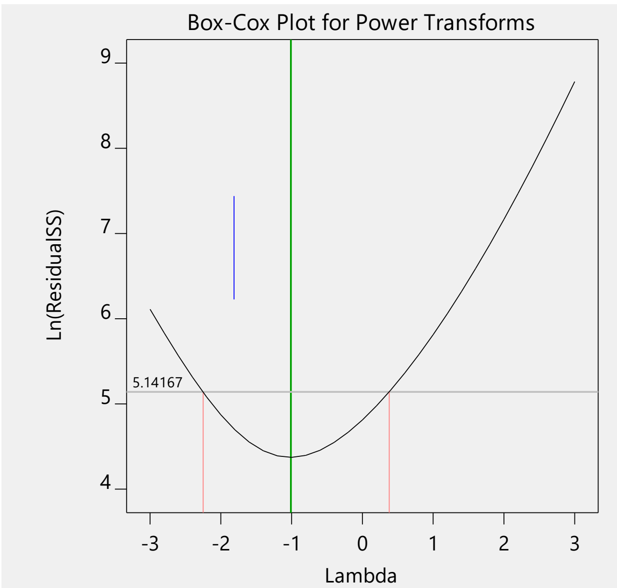


Figure 2: Box-Cox plot for Electrocoagulation data

3.2 Model Reduction and Transformation

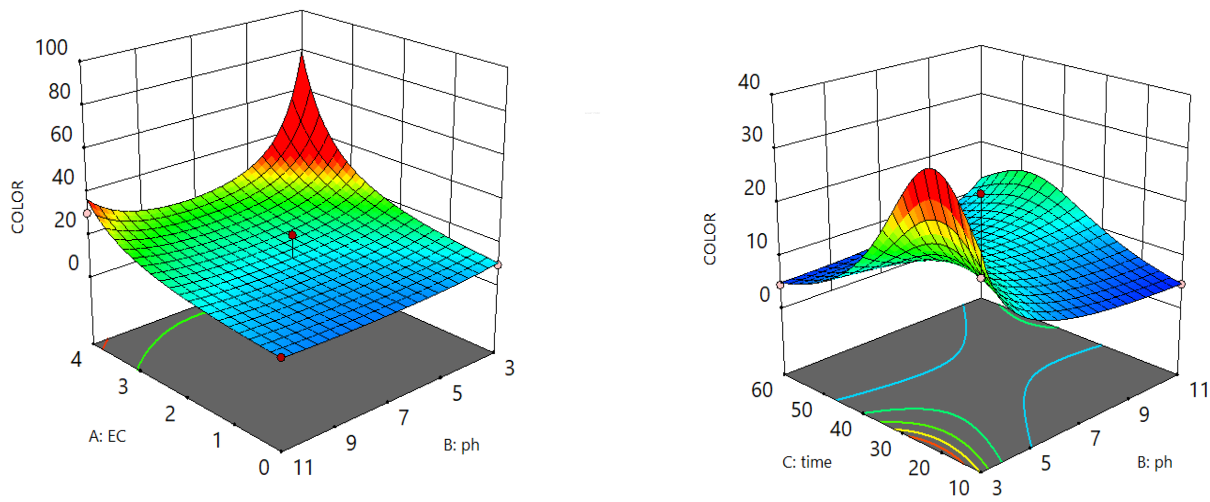
The new model still did not attain significance until the quadratic model was reduced. The model's F-valve of 4.74 and the p-value of 0.0369 indicated the employed model is significant, there is only a 3.69% chance that the F-value occur due to noise. The model was modified by the removal of the A^2 quadratic co-efficient in-order to make the model fit properly. The high P-values 0.5018, 0.6717, 0.8846 and 0.3068 for factors B, C, AB and B^2 respectively indicated that they were not significant to the model. However, only factors AB and B^2 were eliminated in order to improve the quality of the regression model. Furthermore, the lack of fit F-value of 0.9244 is not significant relative to the pure error, and this demonstrated that the model successfully fits. That is, it approximately predicted the effect of the factors on the treatment process.

Therefore, the model for predicting the colour removal using the EC process includes a set of coefficients and uncoded factors as shown in Eqn. 2.

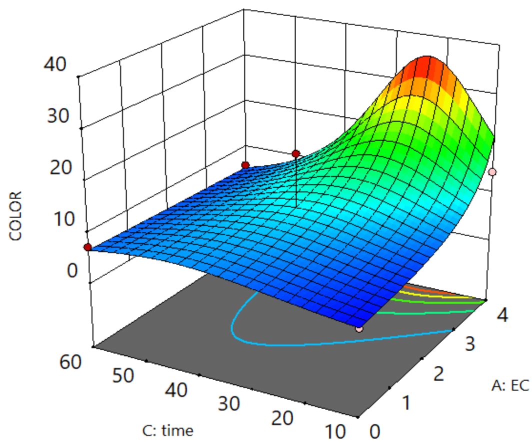
$$Y_{Colour}^{-1} = 0.0772 - 0.0576\,A + 0.0484\,B - 0.0045\,C + 0.0011\,AC - 0.0007\,BC + 0.0001\,C^2 \quad (2)$$

3.3 Effect of Operating Parameters

The response surface plot obtained from the combined effects of the 3 operating parameters: NaCl concentration, Electrolyte time and pH on colour removal is shown in Figure 3.



(a) (b)



(c)

Figure 3: Three-dimensional surfaces for Colour removal efficiencies: (a) EC vs. initial pH at electrolysis time 35 min (b) initial pH vs. electrolysis time at EC 2g/l, and (c) EC vs electrolysis time at initial pH 7.

3.3.1 Effect of NaCl concentration and the initial pH

It could be observed from Figure 3a that there was no noticeable colour removal at constant electrolysis time of 35 minutes until NaCl was added. The removal efficiency increased up to about 20% as the NaCl concentration increased. This was as a result of the increase in current density as the solution conductivity increased (Naje *et al*., 2017). Higher removal efficiency was obtained at lower (acidic) pH (<3) as the model predicts a maximum removal of 80% at a pH of 3 and a NaCl concentration of 4 mg/l.

3.3.2 Effect of initial pH and electrolysis time

The 3-D interaction between the initial pH and the Electrolysis time at a constant NaCl concentration of 2 mg/l is shown in Figure 3b. At 60 mins electrolysis time, the plot is linear as the pH moves from the acidic medium up to the alkaline. The best colour removal efficiency predicted by the response surface plot for this combination is about 36%. The optimum parameters are pH of 3 at 20 mins electrolysis time.

3.3.3 Effect of NaCl concentration and electrolysis time

Figure 3c shows 3-D interaction between the NaCl concentration and the Electrolysis time at the constant initial pH of 7. The effect of increasing the NaCl concentration on the Color removal efficiency was more than that of increasing the electrolysis. From ANOVA, it was observed that Factor C is not significant to the model. This therefore, explains the little effect that the electrolysis time have on the process. The best removal efficiency predicted by the plot for this combination is also about 36% with optimum NaCl concentration of 4 g/l and electrolysis time of 20 mins.

3.4 Optimizing the Treatment Process

The optimum conditions were obtained by taking the top summit point of the various response surface plots (Figure 2). The Response surface model gives a predicted maximum Colour removal rate of 80% at an optimum initial pH of 3 and NaCl concentration of 4 mg/l during 35 mins electrolysis time. The predicted value is very far from the experimentally observed value of 32.4%. This is largely due to the variation and constant degradation in the current supplied by the battery. It could also be partly due to the purity of the metals used as electrodes.

4. Conclusion

This study investigated the performance of EC technique for decolorization of Indigo-dye wastewater in a laboratory pilot-scale batch study. The findings of this study revealed that the pH of the medium and Electrolyte concentration were the most influential parameters than the electrolysis time in colour removal through electrocoagulation. Acidic pH of 3 and increased concentrations of NaCl will produce higher colour removal. Electro-coagulation technique could be used in reducing color from Indigo Dye-rich wastewater in textile industry.

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