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Electrochemical treatment of Baker's yeast wastewater containing melanoidin: optimization through response surface methodology

E. Gengec, M. Kobya, E. Demirbas, A. Akyol and K. Oktor

ABSTRACT

Effluents from Baker's yeast production plant contain a high percentage of color and a large amount of organic load. In the present study, Baker's yeast wastewater (BYW) is treated with the electrocoagulation (EC) process using Al electrodes. Operating parameters (pH, current density, color intensity and operating time) are optimized by response surface methodology (RSM). Quadratic models are developed for the responses which are removal efficiencies of color, chemical oxygen demand (COD) and total organic carbon (TOC) and operating cost (OC). Optimum operating parameters and responses are determined as initial pH 5.2, current density of 61.3 A/m^2 and operation time of 33 min, and 71% of color, 24% of COD, 24% of TOC removal efficiencies and OC of 0.869 €/m^3, respectively. The quadratic model fits for all responses very well with R^2 (>0.95). This paper clearly shows that RSM is able to optimize the operating parameters to maximize the color, COD and TOC removal efficiencies and minimize the OC.

Key words | Baker's yeast wastewater, electrocoagulation, operating cost, optimization, melanoidin

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INTRODUCTION

Baker's yeast industries generally use molasses as a raw material which is a by-product of the sugar industry, has high organic content and dark brown color. The effluents from the process of production of yeast and molasses contain a large amount of coloring substances (melanoidin, etc.) and give dark brown color to the effluent. The Baker's yeast wastewater (BYW) contains high chemical oxygen demand (COD) (10–80 g/L), is strongly nitrogenous (0.5–1.5 g/L total N), sulfate-rich (2–10 g/L), (Gladchenko *et al.* 2004) color intense (3.5–3.8 Abs_{475nm}/cm), of low pH 5–6 and strongly odorous.

Several treatment processes used for the treatment of BYW are advanced oxidation processes such as ozone, photo-Fenton (Altinbas *et al.* 2003; Catalkaya & Sengul 2006); physico-chemical treatments such as coagulation, adsorption, membrane filtration, and biological processes (Mutlu *et al.* 2002; Gladchenko *et al.* 2004; Kalyuzhnyi *et al.* 2005; Zhou *et al.* 2008; Liang *et al.* 2010). Many of them are cost-effective and limiting their widespread application in biologically treated yeast wastewater. For example, 86% for COD and 95% for color removal efficiencies by ozone at a cost of 1.58 €/m^3 and 71% for COD and 96% for color removal efficiencies by the Fenton process at a cost of 1.33 €/m^3 are reported for biologically treated BYW (Altinbas *et al.* 2003). Moreover, conventional biological treatment has commonly been applied to reduce biochemical oxygen demand (BOD) from molasses wastewater; however, the color remains brown due to the presence of melanoidin. BYW contains a relatively high amount of 'inert' organics (color) which can't be removed biologically. In addition, the disadvantages of these treatment processes are for instance, high equipment and operational cost, less effective decolorization rate and simultaneous generation of hazardous secondary pollutants in treated effluents. Therefore, treatment of BYW requires an alternative treatment technology in terms of relatively simple, cost-effective and effective removals of color, COD and total organic carbon (TOC) before its safe disposal into the environment.

Electrocoagulation (EC) has the potential to extensively eliminate the disadvantages of the classical treatment techniques to achieve a sustainable and economic treatment of industrial wastewater (Chen 2004). EC has also received considerable attention for treatment of wastewater as EC treatment is characterized by simple and easily operated

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equipment, short operation time, no addition of chemicals and low sludge production, respectively. Moreover, the advantages of the EC compared with conventional chemical coagulation include reduced wastewater acidification and salinity, low dosage of coagulant, superior coagulant dispersion and intrinsic electroflotation separation capability. Iron (Fe) or aluminum (Al) is generally employed as a sacrificial electrode material in the EC process. In an EC process, the coagulating ions are produced *in situ* involving three successive stages: (i) formation of coagulants by electrolytic oxidation of the sacrificial electrode such as Fe or Al; (ii) destabilization of the contaminants, particulate suspension and breaking of emulsions; and (iii) aggregation of the destabilized phases to form flocs. Fe/Al gets dissolved from the anode generating corresponding metal ions, which almost immediately hydrolyze to polymeric iron or aluminum oxyhydroxides. These polymeric oxyhydroxides are excellent coagulating agents. The mechanism of the EC process is as follows:

$$\text{At the anode: } \mathrm{M} \rightarrow \mathrm{M}^{n+} + n\mathrm{e}^- \quad (1)$$

$$\text{At the cathode: } n\mathrm{H_2O} + n\mathrm{e}^- \rightarrow \frac{n}{2}\mathrm{H_2} + n\mathrm{OH}^- \quad (2)$$

where M is anode material (Al) and n is number of electrons involved in the oxidation/reduction reaction. Soluble metal ions are generated at the anode and react with the hydroxide ions formed at the cathode, and the metal hydroxides are produced as shown in Equation (3). These insoluble metal hydroxides react with the suspended and/or colloid solids and precipitate:

$$\mathrm{M}^{n+} + n\mathrm{OH}^- \rightarrow \mathrm{M(OH)}_{n(\mathrm{s})} \quad (3)$$

M^{n+} and OH^- ions generated by electrode reactions (1) and (2) react to form various hydroxo monomeric and polymeric species, depending on pH range, which transform finally into $\mathrm{M(OH)}_{n(\mathrm{s})}$ according to complex precipitation kinetics.

There are some studies about effluents of BYW treated firstly by a biological treatment process and followed by the EC in the literature (Kobya *et al.* 2008; Gengec *et al.* 2012). But, there are no studies directly yet for treatment of raw BYW by EC, which contributes to the novelty of this study. This study aims to investigate the removal efficiencies of color, COD and TOC from BYW by the EC process for the first time through a response surface methodology (RSM) to get acceptable discharge limits. The optimum operating conditions and operating costs (OCs) are determined with RSM which produces a number of equations with both the maximum removal efficiencies and minimum OC.

MATERIALS AND METHODS

Baker's yeast wastewaters

BYW is obtained from the Baker's Yeast factory in Kocaeli, Turkey. The existing treatment systems consist of two-stage processes which are aerobic and anaerobic. The flow rate of the treatment plant is 1,050 m^3/day. The high strength wastewaters are treated initially in the anaerobic stage and then are combined with the low strength wastewaters to be treated in the aerobic stage. The wastewater is collected from the anaerobic influent of the treatment plant and kept in a refrigerator at 4 °C. The chemical characterizations of the wastewater are pH 5.5, suspended solid of 850 mg/L, alkalinity of 3,800 mg/L $CaCO_3$, COD of 27,082 mg/L, TOC of 6,092 mg/L, color of 3.5 Abs/cm (6,905 as Pt-cobalt), total Kjeldahl nitrogen of 1,100 mg/L and phosphate of 18 mg/L (6 mg/L as PO_4-P).

Experimental set-up and procedure

EC treatment is carried out in a batch electrolytic reactor made from plexiglas material with dimensions of $100 \times 100 \times 130$ mm (Gengec *et al.* 2012). Four aluminum electrodes in the reactor are used as cathode and anode with effective dimensions of $80 \times 50 \times 3$ mm. The total effective area of the electrodes is 240 cm^2. The electrodes are situated 0.5 cm apart from each other and connected in monopolar parallel connection mode. The EC reactor is agitated at 200 rpm to maintain uniformity throughout the system.

The sample is diluted with deionized water to obtain desired color intensity and pH of sample is adjusted with H_2SO_4 or NaOH. pH of the solutions is measured by pH meter (WTW inolab pH 720 model). An amount of 0.80 L wastewater sample is placed into the EC reactor and current density is adjusted by a digital DC power supply (TDK-Lambda Genesys model; 50 V–30 A) operated at galvanostatic mode and the experimental operation is started. After each experimental run, the electrodes are dipped into solutions of HCl (35%) and hexamethylenetetramine (2.8%) to remove the oxide and/or passivation layers from the electrodes. The electrode surfaces are rinsed several times with deionized water and dried and weighed before

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and after the EC experiment. All experiments are performed at 25 °C.

Instruments and analysis procedures

The samples are analyzed for COD and TOC according to Standard Methods (APHA 1998). COD is measured by closed reflux titrimetric method, TOC levels are determined through combustion of the samples at 680 °C using a non-dispersive IR source (Tekmar Dohrmann Apollo 9000). Color content of wastewater is measured using a UV–Vis spectrophotometer (Perkin-Elmer 550 SE) at 475 nm (Raghukumar & Rivonkar 2001). The analysis of COD, TOC and color removals are carried out after filtering with Whatmann 42 filter paper. The following equation is used to calculate for COD, TOC and color removal efficiencies (R_e):

$$R_e(\%) = \frac{C_0 - C_f}{C_0} \times 100 \quad (4)$$

where C_i and C_f are the initial and final concentration of the wastewater.

Experimental design and data analysis

The Design Expert 8.0.4 software (trial version) is used for the statistical design of experiments and data analysis and performed in duplicate. The four most important operating variables: initial wastewater pH (x_1), current density (x_2), operating time (x_3) and color (x_4) are optimized. Their range and levels are given in Table 1. The independent variables' range and levels are chosen after preliminary experiments.

RSM is a collection of mathematical and statistical techniques, commonly used for improving and optimizing processes (Myers & Montgomery 2002). RSM uses an experimental design such as the Central Composite Design (CCD) to fit a model by least squares technique. RSM usually contains three steps: (i) design and experiments; (ii) response surface modeling through regression; and (iii) optimization. RSM makes it possible to represent independent process parameters in quantitative form as:

$$y = f(x_1, x_2, x_3, \ldots\ldots\ldots x_n) \pm \varepsilon \quad (5)$$

where y is the response (yield), f is the response function, ε is the experimental error, and $x_1, x_2, x_3, \ldots\ldots x_n$ are independent parameters.

By plotting the expected response of y, a surface known as the response surface is obtained. The form of f is unknown and may be very complicated. Thus, an RSM aims at approximating f by a suitable lower-ordered polynomial in some region of the independent process variables. If the response can be well modeled by a linear function of the independent variables, the function in Equation (5) can be written as:

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i=1}^{k-1} \sum_{i=1}^{k} \beta_{ij} x_i x_j + \varepsilon \quad (6)$$

The predicted response (y) is therefore correlated to the set of regression coefficients (β): the intercept (β_0), linear ($\beta_1, \beta_2, \beta_3$), interaction ($\beta_{12}, \beta_{12}, \beta_{23}$) and quadratic coefficients ($\beta_{11}, \beta_{22}, \beta_{33}$).

Analysis of variance (ANOVA) is used for graphical analyses of the data to obtain the interaction between the process variables and the responses. The quality of the fit polynomial model is expressed by R^2, and its statistical significance is checked by the Fisher F-test in the same program. Model terms are evaluated by the p-value (probability) with 95% confidence level.

The operating cost

The OC of the EC process includes consumption of electrodes (ELC) and energy (ENC), as well as labor, maintenance, sludge dewatering and disposal, and fixed costs (Kobya *et al.* 2010). In this study, ENC and ELC are taken into account as

Table 1 | Process factors and their levels used for the treatment process

			Range of actual and coded variables				
Factor	**Variables**	**Units**	**−2**	**−1**	**0**	**+1**	**+2**
x_1	Initial pH	–	2.50	4.00	5.50	7.00	8.50
x_2	CD	A/m^2	15.00	35.00	55.00	75.00	95.00
x_3	Time	min	10.00	20.00	30.00	40.00	50.00
x_4	Initial color intensity	Abs/cm	0.50	1.25	2.00	2.75	3.50

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major cost items in the calculation of the OC, €/m^3 of wastewater treated:

$$OC = aENC + bELC + cCC \quad (7)$$

where ENC, ELC and CC are energy (kWh/m^3), electrode (kg/m^3) and chemical consumptions, respectively. a is electrical energy price (0.072 €/kWh), b is electrode material price (1.65 €/kg Al) and c is constant for chemical consumption such as NaOH (0.73 €/kg) and H_2SO_4 (0.29 €/kg). Values of a, b and c are given for the Turkish market in March 2011. ENC and ELC are calculated from Faraday's Law (Equations (8) and (9)).

$$ENC = \frac{U \times i \times t_{EC}}{v} \quad (8)$$

$$ELC = \frac{i \times t_{EC} \times M_W}{z \times F \times v} \quad (9)$$

where U is cell voltage (volt), i is current (Ampere), t_{EC} is the operating time (min) and v is the volume (m^3) of the wastewater, M_W are molecular mass of aluminum (27 g/mole), z is number of electrons transferred ($z = 3$) and F is Faraday's constant (96,487 C/mole).

RESULTS AND DISCUSSION

Statistical analysis

The RSM is used for analyzing the relationship of four variables (initial pH, current density, operating time and initial color intensity) and process responses (removals of color, COD and TOC) in this study. The adequacy of the RSM is justified through ANOVA and the results are shown in Table 2. A higher value of R^2 (0.97 for color, 0.97 for COD and 0.96 for TOC) shows that the model can explain the response successfully (Figure 1).

CV as the ratio of the standard error of estimate to the mean value of the observed response (as a percentage) is a measure of reproducibility of the model. As shown in Table 2, CV (%) for color, COD and TOC removals is found to be 16.07, 14.81 and 16.65% respectively. The

Table 2 | ANOVA results for response parameters for Al electrode

Response	R^2	Adj. R^2	SD	CV	PRESS	F-value	$Prob > F$	AP
Color removal (%)	0.97	0.93	5.97	16.07	2,728.7	29.7	<0.0001	20.561
COD removal (%)	0.97	0.93	1.93	14.81	249.4	37.9	<0.0001	18.373
TOC removal (%)	0.96	0.92	2.06	16.65	279.8	26.0	<0.0001	18.012
ENC (kWh/m^3)	0.99	0.99	0.068	3.30	0.30	630	<0.0001	88.461
ENC (€/m^3)	0.99	0.99	0.0005	3.30	0.0002	630	<0.0001	88.461
ELC (kg/m^3)	0.99	0.99	0.011	3.20	0.017	277.9	<0.0001	57.455
ELC (€/m^3)	0.99	0.99	0.017	3.10	0.017	464.9	<0.0001	71.848
OC (€/m^3)	0.99	0.99	0.018	2.59	0.019	725.6	<0.0001	91.157

SD: Standard Deviation, CV: Coefficient of Variance, PRESS: Predicted Residual Error Sum of Squares, AP: Adequate Precision, P: Probability of Error.

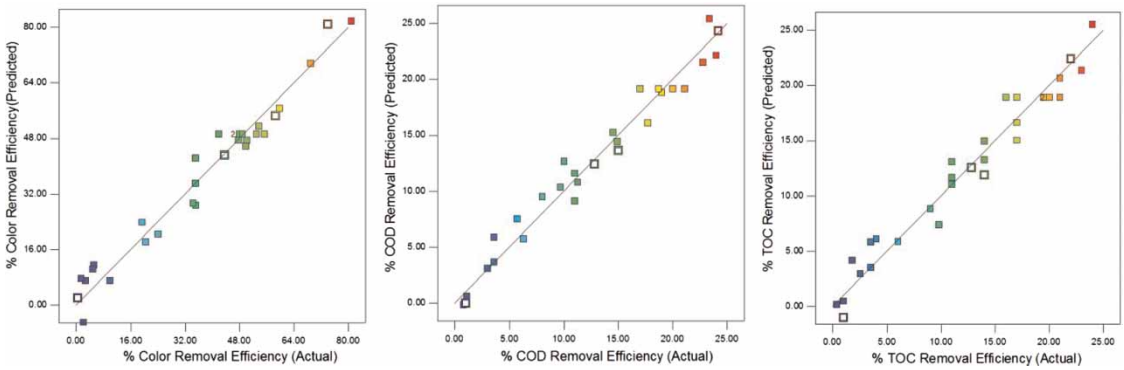


Figure 1 | Comparison of predicted and experimental values for COD, color and TOC removals.

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F-statistics values are higher and their values are 29.7 for color, 37.9 for COD and 26.0 for TOC. The large *F*-values indicate that most of the variation in the response can be explained by the regression model. The *prob* > *F*-value from the ANOVA is less than 0.05, which shows that the model terms are significant. In this study *prob* > *F is* <0.0001 for all response values. Adequate precision (AP) compares the range of the predicted values at the design points to the average prediction error. The AP measures the signal to noise ratio and a ratio greater than 4 is desirable. The actual design of this work for Al electrodes is presented in Table 3. Table 3 shows the CCD in the form of a 2^4 full factorial design with six additional experimental trials as replicates of the central point. The quadratic regression model for color, COD and TOC removal efficiencies and OC including energy and electrode consumptions obtained from the CCD design in terms of process factors (x_1,x_2, x_3 and x_4) are expressed as follows:

$$\begin{aligned}\mathrm{Re}_{(\mathrm{Color})}(\%) &= -113.8 + 15.7{*}\mathrm{pH} + 1.5{*}\mathrm{CD} \\ &+ 3.1{*}\mathrm{t} + 25.7{*}\mathrm{Abs} + 8.3.10^{-3}{*}\mathrm{pH}{*}\mathrm{CD} + 0.05{*}\mathrm{pH}{*}\mathrm{t} \\ &- 1.6{*}\mathrm{pH}{*}\mathrm{Abs} + 0.01{*}\mathrm{CD}{*}\mathrm{t} - 0.2{*}\mathrm{CD}{*}\mathrm{Abs} - 0.18{*}\mathrm{t}{*}\mathrm{Abs} \\ &- 1.53{*}\mathrm{pH}^2 - 9.5.10^{-3}{*}\mathrm{CD}^2 - 0.04{*}\mathrm{t}^2 - 5.89{*}\mathrm{Abs}^2 \end{aligned} \quad (10)$$

$$\begin{aligned}\mathrm{Re}_{(\mathrm{COD})}(\%) &= -66.6 + 12.6{*}\mathrm{pH} + 0.32{*}\mathrm{CD} + 1.8{*}\mathrm{t} \\ &+ 15.4{*}\mathrm{Abs} - 0.04{*}\mathrm{pH}{*}\mathrm{CD} + 0.05{*}\mathrm{pH}{*}\mathrm{t} - 0.39{*}\mathrm{pH}{*}\mathrm{Abs} \\ &+ 5.3.10^{-3}{*}\mathrm{CD}{*}\mathrm{t} - 0.05{*}\mathrm{CD}{*}\mathrm{Abs} - 0.09{*}\mathrm{t}{*}\mathrm{Abs} \\ &- 1.03{*}\mathrm{pH}^2 - 4.95.10^{-4}{*}\mathrm{CD}^2 - 0.03{*}\mathrm{t}^2 - 3.69{*}\mathrm{Abs}^2 \end{aligned} \quad (11)$$

$$\begin{aligned}\mathrm{Re}_{(\mathrm{TOC})}(\%) &= -87.8 + 15.6{*}\mathrm{pH} + 0.4{*}\mathrm{CD} + 2.2{*}\mathrm{t} + 21.1{*}\mathrm{Abs} \\ &- 0.042{*}\mathrm{pH}{*}\mathrm{CD} - 0.017{*}\mathrm{pH}{*}\mathrm{t} - 0.78{*}\mathrm{pH}{*}\mathrm{Abs} \\ &+ 5.6.10^{-3}{*}\mathrm{CD}{*}\mathrm{t} - 0.067{*}\mathrm{CD}{*}\mathrm{Abs} - 0.13{*}\mathrm{t}{*}\mathrm{Abs} \\ &- 1.07{*}\mathrm{pH}^2 - 1.04.10^{-3}{*}\mathrm{CD}^2 \\ &- 0.032{*}\mathrm{t}^2 - 4.07{*}\mathrm{Abs}^2 \end{aligned} \quad (12)$$

$$\begin{aligned}\mathrm{ENC}(\mathrm{kWh/m^3}) &= -0.01 + 0.22{*}\mathrm{pH} - 0.015{*}\mathrm{CD} - 0.03{*}\mathrm{t} \\ &+ 0.14{*}\mathrm{Abs} + 9.3.10^{-3}{*}\mathrm{pH}{*}\mathrm{CD} - 2.2.10^{-3}{*}\mathrm{pH}{*}\mathrm{t} + 0.05 \\ &{*}\mathrm{pH}{*}\mathrm{Abs} + 1.8.10^{-3}{*}\mathrm{CD}{*}\mathrm{t} - 9.6.10^{-3}{*}\mathrm{CD}{*}\mathrm{Abs} \\ &- 3.2.10^{-3}{*}\mathrm{t}{*}\mathrm{Abs} - 0.03{*}\mathrm{pH}^2 + 2.210^{-4}{*}\mathrm{CD}^2 \\ &+ 2.4.10^{-4}{*}\mathrm{t}^2 - 5.7.10^{-3}{*}\mathrm{Abs}^2 \end{aligned} \quad (13)$$

$$\begin{aligned}\mathrm{ELC}(\mathrm{kg/m^3}) &= -0.11 + 0.02{*}\mathrm{pH} - 1.50.10^{-3}{*}\mathrm{CD} \\ &- 5.2.10^{-6}{*}\mathrm{t} + 0.10{*}\mathrm{Abs} + 2.1.10^{-5}{*}\mathrm{pH}{*}\mathrm{CD} + 4.2.10^{-5} \\ &{*}\mathrm{pH}{*}\mathrm{t} - 0.01{*}\mathrm{pH}{*}\mathrm{Abs} + 2.10^{-3}{*}\mathrm{CD}{*}\mathrm{t} + 2.08.10^{-4}{*}\mathrm{CD}{*}\mathrm{Abs} \\ &- 5.8.10^{-4}{*}\mathrm{t}{*}\mathrm{Abs} + 4.6.10^{-3}{*}\mathrm{pH}^2 + 9.6.10^{-3}{*}\mathrm{CD}^2 \\ &+ 1.3.10^{-3}{*}\mathrm{t}^2 - 8.7.10^{-3}{*}\mathrm{Abs}^2 \end{aligned} \quad (14)$$

$$\begin{aligned}\mathrm{OC}_{(€/\mathrm{m^3})} &= -0.18 + 0.05{*}\mathrm{pH} - 3.5.10^{-3}{*}\mathrm{CD} - 2.1.10^{-3}{*}\mathrm{t} \\ &+ 0.17{*}\mathrm{Abs} + 1.10^{-4}{*}\mathrm{pH}{*}\mathrm{CD} - 9.6.10^{-5}{*}\mathrm{pH}{*}\mathrm{t} \\ &- 0.02{*}\mathrm{pH}{*}\mathrm{Abs} + 4.6.10^{-4}{*}\mathrm{CD}{*}\mathrm{t} - 3.4.10^{-4}{*}\mathrm{CD}{*}\mathrm{Abs} \\ &- 1.2.10^{-3}{*}\mathrm{t}{*}\mathrm{Abs} - 1.7.10^{-3}{*}\mathrm{pH}^2 \\ &+ 3.210^{-5}{*}\mathrm{CD}^2 + 4.0.10^{-3}{*}\mathrm{t}^2 - 0.02{*}\mathrm{Abs}^2 \end{aligned} \quad (15)$$

A positive and negative sign in front of the terms refer to a synergistic effect and antagonistic effect, respectively.

To examine the effects of all factors on response, the perturbation plot shows how the response changes as each factor moves from the chosen reference point, with all other factors held constant at the reference value. As seen in Figure 2, a reference point is chosen at the middle of the design space. Perturbation plot (2a) shows that color removal efficiency was higher between pH 4.0 and 6.0. Color removal efficiency decreases in the EC process when pH is <4.0 and >6.0. As seen in Table 3 (runs 22, 26 and 29), color removal efficiency is realized 35, 49 and 34% for initial pH of 2.5, 5.5 and 8.5, respectively as all other factors hold constant.

In the experiments, the electrochemical generated aluminum ions may form monomeric and polymeric Al-hydrolysis species such as $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al(OH)_4^-$, $Al_6(OH)_{15}^{-3}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$ and $Al_{13}(OH)_{34}^{5+}$ at pH 4–6. Above pH > 6, oversaturation and formation of amorphous hydroxide precipitate $Al(OH)_{3(s)}$ enmeshes colloidal particles in a sweep floc (Rebhun & Lurie 1993). Formation rates of the different species play an important role in the removal process. Four major interaction mechanisms in the treatment of BYW are considered as co-precipitation, charge neutralization, adsorption and sweep coagulation and each one is being proposed for a separate pH range. Thus, charge neutralization and co-precipitation between these hydrolysis products and hydrolyzed negatively charged organic molecules such as melanoidin in the BYW occur in the pH range of 4–6. The EC process in the low pH range is explained as co-precipitation while the higher pH range (>6) is adsorption and sweep coagulation (Rebhun & Lurie 1993; Zhou *et al.* 2008).

Increasing current density (x_2) and time (x_3) contributes noticeably to removal efficiency in the perturbation plot

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Table 3 | CCD for the run of four experimental variables and the obtained results

					Removal efficiency (%)			Consumptions		
Run	x_1 pH	x_2 CD	x_3 t_{EC}(min)	x_4 Color	Color	COD	TOC	ENC (kWh/m^3)	ELC (kg Al/m^3)	OC (€/m^3)
1	5.5	95	30	2.00	60	24	23	4.50	0.60	1.314
2	7.0	35	20	2.75	2	3	3	0.69	0.13	0.265
3	5.5	55	50	2.00	59	15	14	3.55	0.56	1.180
4	7.0	75	40	2.75	35	8	4	3.80	0.59	1.247
5	7.0	35	40	1.25	50	19	17	1.28	0.29	0.571
6	7.0	35	20	1.25	19	11	11	0.77	0.16	0.319
7	4.0	35	40	1.25	48	15	14	1.57	0.28	0.575
8	4.0	75	20	1.25	54	18	17	2.30	0.29	0.644
9	4.0	35	20	1.25	35	10	9	0.78	0.13	0.271
10	7.0	75	20	2.75	3	1	1	1.82	0.30	0.626
11	4.0	35	20	2.75	10	4	4	0.62	0.15	0.292
12	5.0	55	30	2.00	48	17	16	1.99	0.35	0.721
13	4.0	75	20	2.75	20	4	4	1.59	0.33	0.659
14	5.5	55	30	0.50	69	23	21	2.40	0.32	0.701
15	5.5	15	30	2.00	5	15	14	0.33	0.09	0.172
16	7.0	35	40	2.75	5	6	2	1.32	0.26	0.524
17	5.5	55	30	2.00	48	21	20	2.06	0.33	0.693
18	5.5	55	10	2.00	2	1	1	0.77	0.11	0.237
19	5.5	55	30	2.00	53	19	20	2.06	0.33	0.693
20	7.0	75	20	1.25	50	10	11	2.33	0.30	0.663
21	4.0	75	40	1.25	81	23	24	4.59	0.59	1.304
22	2.5	55	30	2.00	35	11	11	1.77	0.33	0.672
23	5.5	55	30	2.00	42	21	21	2.05	0.34	0.709
24	5.5	55	30	2.00	55	17	17	2.09	0.32	0.679
25	5.5	55	30	3.50	1	1	1	1.70	0.30	0.617
26	5.5	55	30	2.00	49	20	20	1.90	0.32	0.665
27	7.0	75	40	1.25	74	24	22	4.50	0.63	1.364
28	4.0	75	40	2.75	44	13	13	3.70	0.61	1.273
29	8.5	55	30	2.00	34	11	10	1.90	0.33	0.681
30	4.0	35	40	2.75	24	6	6	1.27	0.29	0.570

(Figure 2(a)). Increasing current density and time in the EC process increases the amount of metal species formed by dissolution of the anode (Faraday's law, Equation (8)). However, if operation time and current density are further increased, removal efficiencies would not be the same as increasing operation time increases the pH of BYW and $Al(OH)_4^-$ is the dominant soluble Al species leading to lower removal efficiencies. Higher operation times increase energy and electrode costs as well. When all factors hold constant except for operating time in Table 3 (runs 3, 12 and 18), color removal efficiencies are realized 59, 48 and 2% for operation times of 50, 30 and 10 min, respectively. On the other hand, runs 1, 12, 15 indicated that color removal efficiencies are 60, 48 and 5% for 95, 55 and 15 A/m^2, respectively.

ENC is closely related to over potential and time as described in Equation (5). The perturbation plot of energy (Figure 2(b)) and electrode (Figure 2(c)) consumption shows that there is no significant effect of initial pH on energy and electrode consumptions, and initial color

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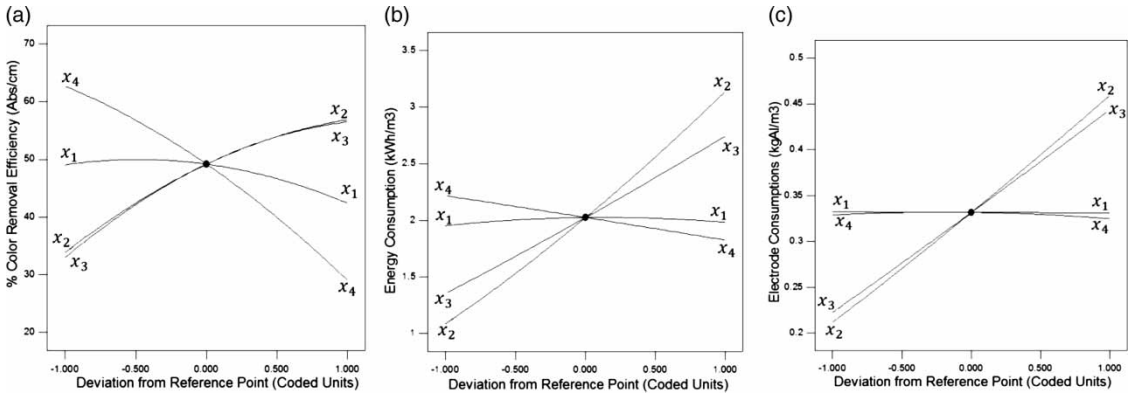


Figure 2 | Perturbation plot of color removal efficiency, ENC (kWh/m^3) and ELC (kg/m^3).

Table 4 | Optimization's constraints and results

Name	Goal	Lower limit	Upper limit	Optimization results	Importance
x_1: pH	In range	4	7	5.2	3
x_2: CD (A/m^2)	In range	35	75	61.3	3
x_3: time (min)	In range	20	40	33	3
x_4: color (Abs/cm)	In range	1.25	2.75	1.25	3
COD removal (%)	Maximize	0.8	24.2	24	3
Color removal (%)	Maximize	0.46	80.92	71	5
TOC removal (%)	Maximize	0.4	24	24	3
OC (€/m^3)	Minimize	0.17226	1.3635	0.869	3

intensity causes an increase in conductivity which leads to a decrease in ENC (Mollah *et al*. 2004). The result is presented for runs of 14, 25 and 26 (Table 3). When initial color intensity is 0.5 Abs/cm, the value of ENC is 2.4 kWh/m^3 and its values are decreased from 1.9 to and 1.7 kWh/m^3 as initial color intensity is increased from 2 to 3.5 Abs/cm.

The desired goals in different runs, selected as maximum color, COD and TOC removals at minimum OC are optimized. Optimization constraints and results are presented with a desirability of 0.802 in Table 4. Corresponding importances of goals are selected as 5 for color removal and 3 for the dependent and independent variables. As seen in Table 4, the minimum value of OC is 0.869 €/m^3 which is set for maximum color removal with an importance of 5.

CONCLUSIONS

The RSM is applied to optimize the operating variables viz. initial pH, current density, initial color intensity and operating time on treatment of BYW by the EC process using aluminum electrodes in a batch reactor. Quadratic models are developed for the responses, such as removal efficiencies of color, COD and TOC, and OC including energy and electrode consumptions. The actual removals of color (81%), COD (23%) and TOC (24%) at optimum operating conditions (pH 4, 75 A/m^2, 40 min and 1.25 Abs/cm) agree well with the optimized conditions (pH 5.2, 61.3 A/m^2, 33 min, 1.25 Abs/cm) which are 71% of color (influent 2,623 Pt-Co and effluent 760 Pt-Co), 24% of COD (influent 7,222 mg/L COD and effluent 5,488 mg/L COD) and 24% of TOC (influent 4,780 mg/L TOC and effluent 3,632 mg/L TOC), respectively. The proposed model also fits very well with the experimental data and values of R^2 are 0.97 for color, 0.97 for COD and 0.96 for TOC. The OC from the model predicts 0,869 €/m^3 as opposed to 1.304 €/m^3 obtained from the actual experiment. ANOVA showed a high coefficient of determination value, thus ensuring a satisfactory adjustment of the second-order regression model with the experimental data.

According to the results, the RSM is a suitable method to optimize the operating conditions (maximize the removal

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efficiencies of color, COD and TOC) while keeping the OCs minimal. The methodology helped in accurate predictions for removal efficiencies of color, COD and TOC from BYW under different process conditions.

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