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Optimization and PID Control of pH and Temperature in an Electrocoagulation Process

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Abstract: In this work, effects of temperature and pH in batch treatment of pulp and paper mill wastewater using electrocoagulation has been investigated. Conductivity, temperature, and pH are selected as controlled variables; supporting electrolyte, cooling water, acid and base flow rates are selected as manipulated variables, respectively. Real time experimental multi input-multi output (MIMO) control of conductivity, temperature, and pH under constant current conditions are achieved using MIMO Proportional Integral Derivative (PID) control algorithms coded in MATLAB™. A central composite design (CCD) has been applied to the system under controlled conditions and optimum pH and temperature values are obtained using response surface methodology (RSM). Both controlled and uncontrolled experiments are performed using optimum values and results are compared in terms of removal efficiencies of pollutants. Results show that 34.47% chemical oxygen demand (COD), 98.06% total suspended solids (TSS), 99.80% turbidity, 99.93% color, and 13.40% SO_4^{2-} removal is achieved in 45 minutes of process operation under controlled conditions and COD, TSS, turbidity, color and SO_4^{2-} removal are increased by 10.92, 2.97, 4.06, 2.89, 3.17 respectively in comparison with uncontrolled operation. The highest removal percentages are obtained under controlled operating conditions as 98.5% and 98.3% for turbidity and color, respectively, for 10 minutes operation. It is concluded that optimum process operating conditions for removal of turbidity and color of pulp and paper mill wastewater is obtained under constant 6.45 pH, 23.24 °C temperature, 1.78 mS/cm conductivity, and power consumption is reduced by 25.3% under controlled conditions.

Keywords: Pulp and paper mill wastewater, pH, temperature, electrocoagulation, PID control, RSM.

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1. INTRODUCTION

The pulp and paper industry has high pollutant discharges to the environment and they consume considerable amount of wood, water and energy. In terms of freshwater withdrawal, the pulp and paper making industry is one of the most water demanding industry and position third in the world, after the primary metals and the chemical industries (Sridhar et al., 2011). The industrial wastewaters cause several problems such as color, slime growth, thermal impacts, and scum formation in the environment. They also affect the ecosystem, enlarge the quantity of toxic substances in the water, and pollute scenic beauty (Pokhrel & Viraraghavan, 2004). Water turbidity generated by the presence of suspended solids and colloidal particles cannot be treated by conventional methods such as filtration and sedimentation (Özyurt et al., 2021; Terrazas et al., 2010). Electrocoagulation process has been confirmed to be effective in destabilizing colloidal particles and color removal (Linares-Hernández et al., 2009). As an electrolytic technology, it consists of anodic dissolution of a metal electrode within the effluent to be treated, with the simultaneous formation of hydroxyl ions and hydrogen gas. Compared with traditional methods, electrocoagulation, in theory, has the advantage of removing the smallest colloidal particles; the smallest charged particles have better probability of being coagulated because of the electric field that sets them in motion (Mollah et al., 2004; Terrazas et al., 2010). Treatment of pulp and paper mill wastewater by electrocoagulation is highly complex process and is greatly influenced by many factors such as electrode configuration and material, current density, electrolysis time, electrical conductivity, pH and temperature (El-Ashtoukhy et al., 2009; Kalyani et al., 2009; Katal

& Pahlavanzadeh, 2011; Khansorthong & Hunsom, 2009; Soloman et al., 2009; Uğurlu et al., 2008; Zaied & Bellakhal, 2009). Among these operational parameters, temperature and pH were found to be more significant than the others. Temperature affects the pollutant removal in many ways such as rate of reactions, solubility of metal hydroxides, liquid conductivity and kinetics of gas bubbles or small colloidal particles (Attour et al., 2014). It is reported that the increase in the temperature of the solution cause increasing the solubility of aluminum (Vepsäläinen et al., 2009). For that reason the precipitation of the aluminum is increased at lower temperatures, which results in a better removal (Katal & Pahlavanzadeh, 2011). In electrocoagulation process direct electrical current applied between metal electrodes immersed in wastewater causes the dissolution of aluminum or iron electrodes into wastewater. Chemical dissolution of metal electrodes is strongly influenced by the pH of the solution. The dissolved metal ions, at an appropriate pH, can form wide ranges of coagulated species and metal hydroxides that destabilize and aggregate suspended particles or precipitate and adsorb dissolved contaminants (Cañizares et al., 2005; Merzouk et al., 2009). At low pH (2-3) cationic monomeric species Al^{3+} and $Al(OH)^{2+}$ dominate. When pH is between 4 and 9, the Al^{3+} and OH^- ions generated by the electrodes react to form various monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2^{2+}$, and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_{13}(OH)_{34}^{5+}$ that finally transform into insoluble amorphous $Al(OH)_{3(s)}$ through complex precipitation kinetics (Bayramoglu et al., 2004). When pH is higher than 10, the monomeric $Al(OH)_4^-$ anion concentration increases (Alinsafi et al., 2005). It is reported that there is a 1-2 units of increase in pH during electrocoagulation (Camcioglu et al., 2017b). Accordingly formations of ionic compounds that have high solubility in water cause a decrease in removal efficiencies. As a result control of pH and temperature in their optimum values come up as a necessity.

In this study the effects of temperature and pH in treatment of pulp and paper mill wastewater with electrocoagulation has been investigated. A CCD has been applied to the system and optimum pH and temperature values are obtained using RSM. In order to determine the effect of constant electrical conductivity, pH and temperature on removal efficiencies during electrocoagulation, experiments were performed under controlled and uncontrolled conditions of operating parameters using optimum values and the results were compared in terms of COD, TSS, turbidity, color, SO_4^{-2} and Cl^- removal efficiencies.

2. EXPERIMENTAL SECTION

2.1. Experimental Procedure

Experiments were carried out in batch process using a 2000 mL electrocoagulation reactor made of flex-glass. In each run, 1000 mL of sedimented pulp and paper mill wastewater was fed into the reactor. Characteristics of sedimented pulp and paper mill wastewater are given below in Table 1.

Table 1: Characteristics of pulp and paper mill wastewater.

Chemical Oxygen Demand (mg/L)	Total Suspended Solids (mg/L)	Turbidity (NTU)	Color (CU)	Conductivity (mS/cm)	pH	SO_4^{2-} (mg/L)	Cl^- (mg/L)
681.72	118	173	923	1.80	7.5	281	105.42

Six electrodes made of aluminum with dimensions of 60 mm x 60 mm x 2 mm were positioned in mono-polar parallel arrangement with a gap separation between them of 10 mm. In order to conduct the experiments under constant current conditions, the electrodes were connected to a DC power supply (MAY 11-PS Constant Current Power Supply) operating in the range of 0-2 A. 0.04 M NaCl, 0.1 M HCl and 0.1 M NaOH solutions were added to the wastewater with peristaltic pumps (Longer Pump LEAD-2). A stirrer (MTOPS MS-3020) was used to maintain uniform concentration and temperature dispersion in the reactor. During experiments a heating/cooling water circulator (Hoefer RCB 20-PLUS) was used at 12 °C in order to keep temperature constant at a desired value and avoid possible temperature increase.

A pH meter, conductivity meter and a thermocouple were used for on-line measurements of pH, conductivity and temperature during wastewater treatment. These probes were placed into a separate chamber inside the reactor to avoid the measurements to be influenced from charge distribution occurring in the reactor. Sample circulation between the chamber and reactor was carried out using a peristaltic pump (Aspen Standard Pump).

On-line signals of pH, conductivity and temperature from measurement devices are sampled and transferred towards controller using data acquisition device (MAY 11-ESA Electrophoresis Control Unit) and calculated signals from controller is transferred to related manipulated variable via the data acquisition device and input variables are adjusted. In electrical control studies 0.04 M NaCl solution flow rate is the input variable and is adjusted by the electrical conductivity controller signals transferred to the peristaltic pump. pH control studies are carried out using 0.1 M HCl solution and 0.1 M NaOH solution flow rate as the input variables and their values are adjusted by the pH controller signals transferred to the peristaltic pumps. Temperature control studies were performed using on/off position of cooling water valve as the manipulated variable and its position is regulated by the temperature controller. Experimental setup is given in Figure 1.

Experiments were carried out in 1 A constant current and 45 minutes electrocoagulation time conditions.

A multi-purpose real-time MATLAB/Simulink model and a PID controller program were designed for performing electrocoagulation studies, monitoring input and output variables, carrying out dynamic analyses and control experiments in electrochemical reactor. Real-time MATLAB/Simulink model is given in Figure 2.

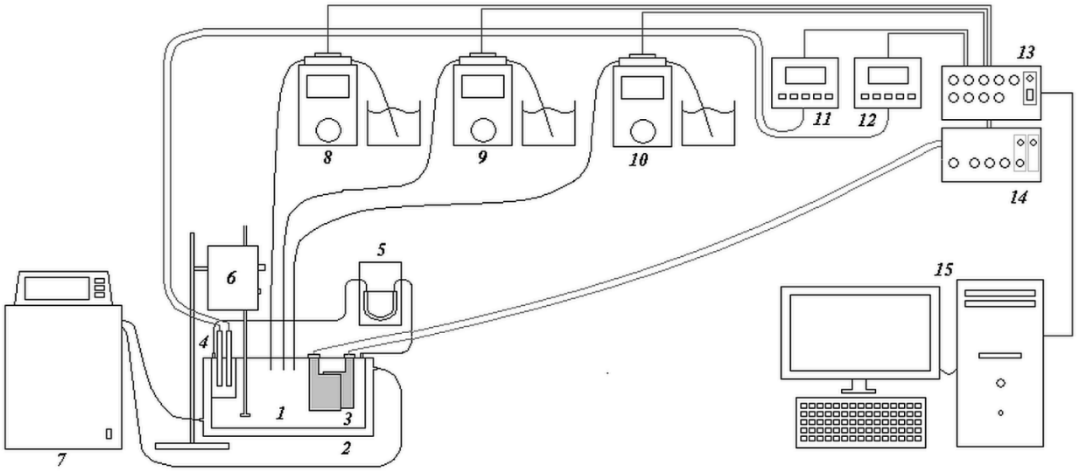

Figure 1: Experimental set-up (1: electrocoagulation reactor, 2: electrocoagulation reactor heating/cooling jacket, 3: electrodes, 4: pH meter, conductivity meter, thermocouple, 5: sample circulation pump, 6: mechanical stirrer, 7: heating/cooling water circulator, 8: acid pump, 9: base pump, 10: supporting electrolyte pump, 11: pH and temperature display, 12: conductivity display, 13: control unit, 14: power supply, 15: computer).

2.2. Analytical Procedure

50 mL of the samples were taken after treatment processes and kept 3 h at 20 °C for sedimentation. Supernatants were collected for analyses. COD analyses were performed according to SM 5220 D (Eaton et al., 2005). 2.5 mL of samples were treated with 1.5 mL of high range digestion solution and 3.5 mL of sulfuric acid reagent in 16 x 100 mm culture tubes. Treated samples were digested at 150 °C for 2 h using a thermoreactor (Velp ECO-16). After digestion process, tubes were cooled down to room temperature and absorbance of the samples were read at 600 nm using a spectrophotometer (PG Instruments T60V). The COD of the samples were calculated with a calibration curve prepared using potassium hydrogen phthalate standard. Color analyses were performed in accordance with SM 2120 C (Eaton et al., 2005). Sample absorbances were read at 456 nm using the spectrophotometer. Color values were calculated by a calibration curve prepared previously using 500 CU Pt-Co stock solution. Sample turbidities were measured using a turbidity meter (Aqualytic AL250T-IR). TSS analyses were performed accordingly according to SM 2540 D (Eaton et al., 2005). SO_4^{2-} and Cl^- analyses are performed in accordance with SM 4500 SO_4^{2-} E and SM 4500 Cl^- B (Eaton et al., 2005) respectively.

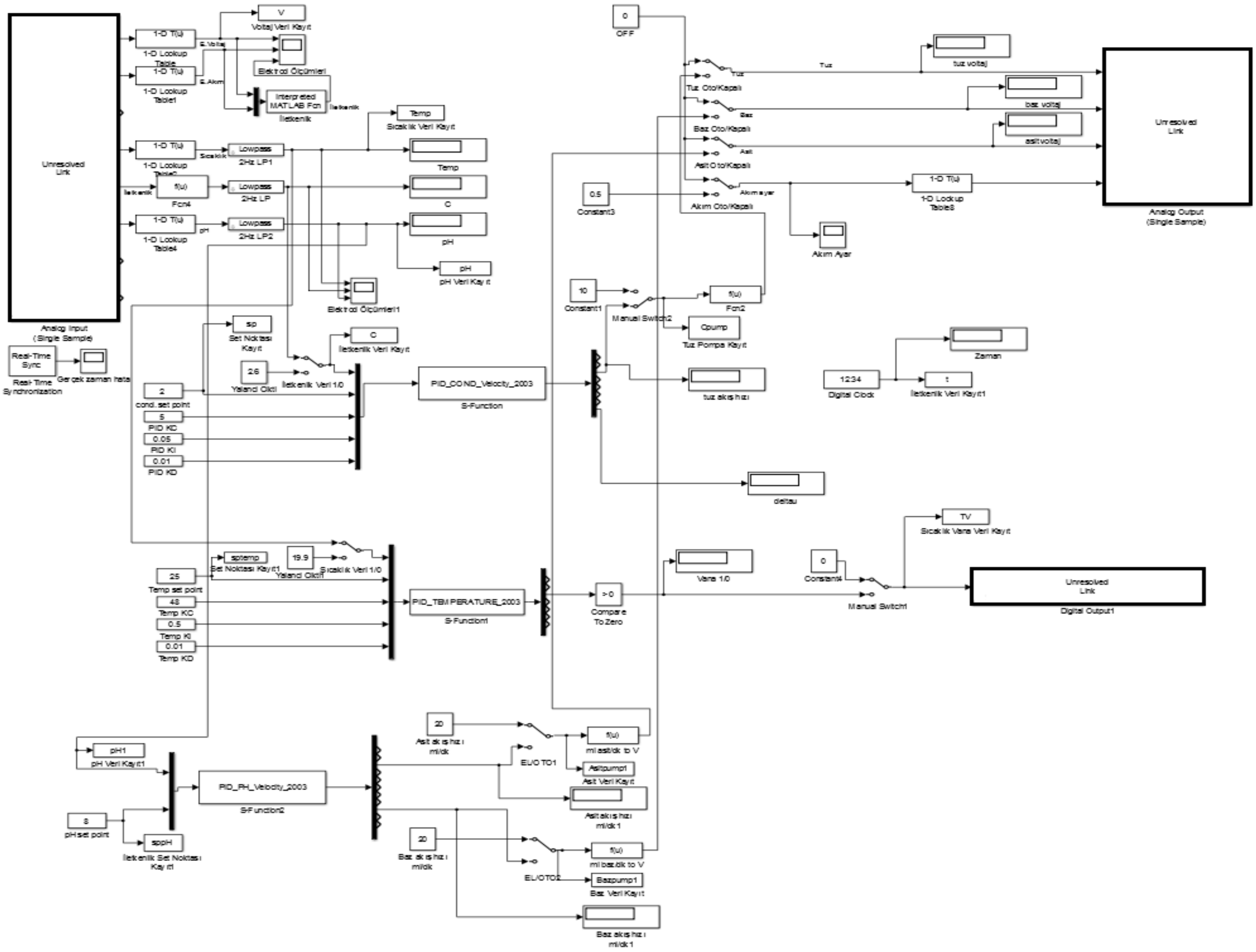

Figure 2: Real-time MATLAB/Simulink model.

3. RESULTS AND DISCUSSION

3.1. Determination of Optimum pH and Temperature

RSM was used to determine the model that gives the relation between dependent variables turbidity removal (y_1) and color removal (y_2) and the independent variables pH (x_1) and temperature (x_2). Experiments were also carried out to find the optimum values of operating parameters for maximum turbidity and color removal. CCD with 2 factors was applied using Minitab 17 statistical software. A total number of 14 experiments consisting of 4 factorial points, 4 axial points and 6 replicates in the center points were employed in this work. Experimental design matrix in terms of uncoded factors and measured responses are given in Table 2.

Table 2: Design of experiments and results for electrocoagulation treatment.

Run	x_1 pH	x_2 Temperature (°C)	y_1 Turbidity removal (%)	y_2 Color removal (%)
1	7.50	22.50	100.00	100.00
2	12.45	22.50	14.20	10.33
3	7.50	22.50	99.99	100.00
4	7.50	22.50	98.12	98.33
5	7.50	11.89	98.68	99.23
6	2.55	22.50	74.68	76.43
7	7.50	33.11	99.34	99.17
8	11.00	15.00	50.18	43.44
9	7.50	22.50	100.00	99.52
10	4.00	30.00	74.43	75.29
11	4.00	15.00	69.43	70.29
12	11.00	30.00	54.70	37.80
13	7.50	22.50	98.83	98.53
14	7.50	22.50	98.59	98.94

Color and turbidity removal results of the samples taken at 15 minute time intervals for each run are presented in Figure 3. A nonlinear regression method was used to fit the experimental data to second-order polynomial equation to identify model terms for turbidity and color removal using Minitab 17 statistical software. Mathematical form of second-order polynomial equation considering all the linear, square and linear by linear interaction terms is given in Equation (1).

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta_{ij} x_i x_j + \varepsilon$$

(Eq. 1)

Where Y is the predicted response, x_i and x_j are the input variables (i and j range from 1 to k), β_0 is the model intercept coefficient, β_i, β_{ii}, β_{ij} are the regression coefficients for the linear, quadratic and interaction terms respectively, k is the number of independent variables and ε is the interaction effect (Korkmaz et al., 2017).

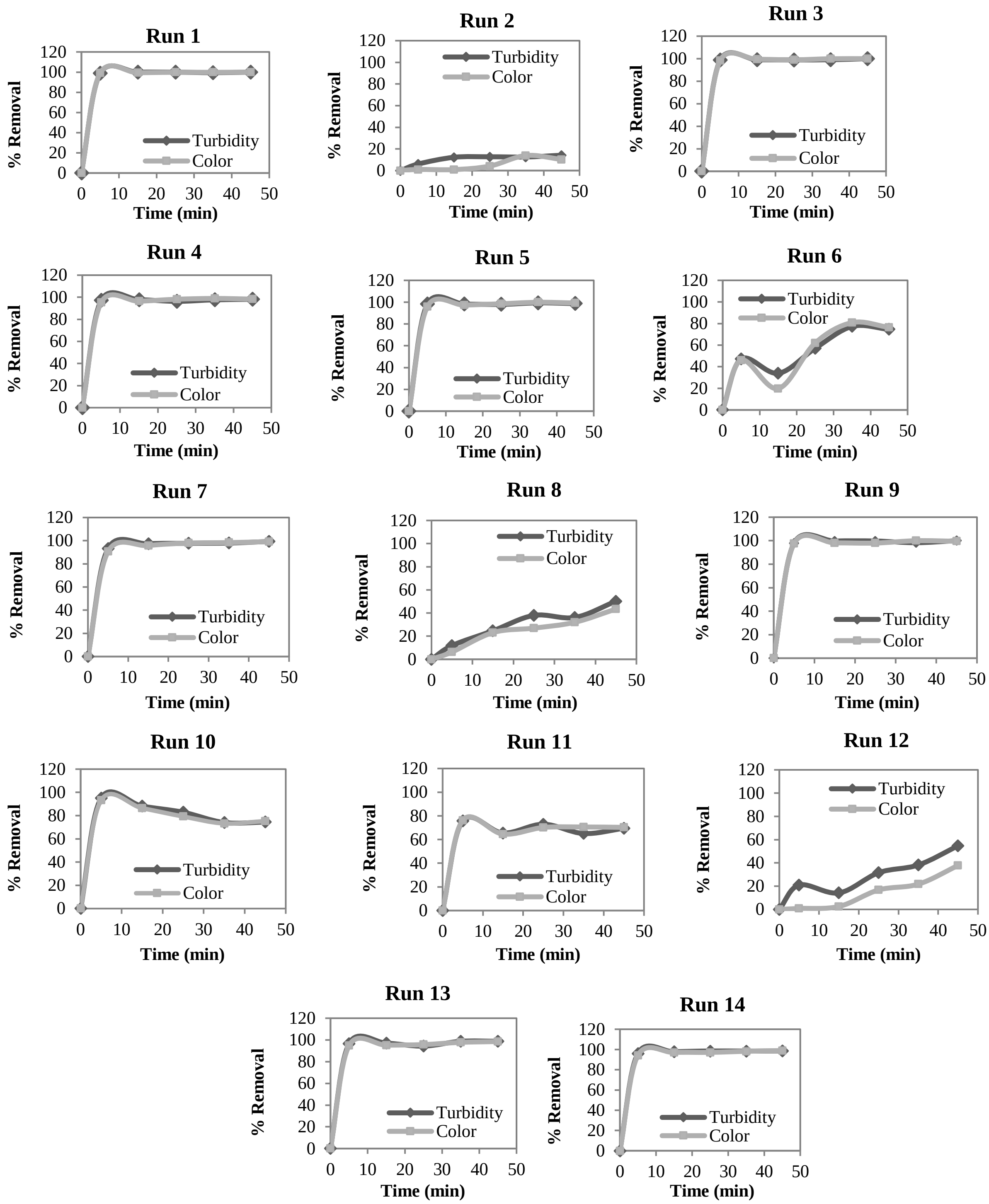

Figure 3: Variation of turbidity and color removal with time for run 1-14.

The second-order polynomial equations for turbidity removal and color removal in terms of uncoded factors are given in Equation 2 (2) and Equation (3), respectively.

$$y_1 = -30.7 + 32.03x_1 + 2.18x_2 - 2.432x_1^2 - 0.0446x_2^2 \quad (2)$$

$$y_2 = -36.1 + 33.02x_1 + 2.91x_2 - 2.577x_1^2 - 0.065x_2^2 \quad (3)$$

The statistical significance of the models were justified through analysis of variance (ANOVA) with F-test at 95% confidence level. Results of regression analysis are shown in Tables 3 and 4 for turbidity and color removal, respectively.

Table 3: Regression analysis results for turbidity removal in terms of coded factors.

Predictor	**Coef**	**SE Coef**	**T**	**P**
Constant	99.25	2.73	36.35	0.000
x_1	-15.56	2.36	-6.58	0.000
x_2	1.31	2.36	0.55	0.596
x_1^2	-29.79	2.46	-12.11	0.000
x_2^2	-2.51	2.46	-1.02	0.338

Analysis of Variance

Source	**DF**	**SS**	**MS**	**F**	**P**
Model	5	8614.04	1722.81	38.51	0.000
Blocks	1	107.88	107.88	2.41	0.159
Linear	2	1951.13	975.56	21.81	0.001
pH	1	1937.48	1937.48	43.31	0.000
Temp	1	13.65	13.65	0.31	0.596
Square	2	6555.03	3277.52	73.27	0.000
pH*pH	1	6554.68	6554.68	146.54	0.000
Temp*Temp	1	46.50	46.50	1.04	0.338
Error	8	357.85	44.73		
Lack-of-fit	4	354.37	88.59	101.77	0.000
Pure Error	4	3.48	0.87		
Total	13	8971.89			

P and t tests were used to determine the compatibility of second order polynomial equations with the experimental results. Terms including temperature were found to be the least effective in the model. ANOVA results presented in Table 4 and 5 indicate higher F-values for regression of turbidity and color models than F distribution table value of 3.69. The large F-value shows that most of the variation in the output can be explained by the developed regression model (Camcioglu et al., 2017a). The associated P-value is also used as an indicator for whether F is large enough to indicate statistical significance. The P-values of reduced quadratic models are <0.001 which clearly confirm good fit of experimental data. The values of R^2, adjusted R^2, predicted R^2 and lack of fit of models are obtained to investigate the accuracy of the suggested polynomials. In order to emphasize the relationships between factors and responses and also to determine the optimum conditions, Equations (2) and (3) are expressed as response surfaces. The value of R^2 gives the correlation of total variation in the effluent removal efficiencies predicted by developed models.

Table 4: Regression analysis results for color removal in terms of coded factors.

Predictor	Coef	SE Coef	T	P
Constant	99.22	2.53	39.16	0.000
x_1	-19.73	2.19	-8.99	0.000
x_2	-0.09	2.19	-0.04	0.968
x_1^2	-31.57	2.28	-13.82	0.000
x_2^2	-3.66	2.28	-1.60	0.148

Analysis of Variance

Source	DF	SS	MS	F	P
Model	5	10737.1	2147.42	55.76	0.000
Blocks	1	254.3	254.29	6.60	0.033
Linear	2	3113.5	1556.74	40.42	0.000
pH	1	3113.4	3113.41	80.84	0.000
Temp	1	0.1	0.07	0.00	0.968
Square	2	7369.3	3684.67	95.67	0.000
pH*pH	1	7358.1	7358.12	191.05	0.000
Temp*Temp	1	98.8	98.77	2.56	0.148
Error	8	308.1	38.51		
Lack-of-fit	4	305.8	76.44	129.81	0.000
Pure Error	4	2.4	0.59		
Total	13	11045.2			

For turbidity and color removal, R^2 values are calculated as 0.9601 and 0.9721 for reduced quadratic models which ensure an acceptable fit to experimental data (Rai et al., 2016). Adjusted R^2 values for the models are 0.9352 and 0.9547 which also high enough to support acceptable correlation between experimental and predicted value. The predicted R^2 values for the present models are 0.7459, 0.8613 and suggest how good the models predict the effluent removal. The adjusted R^2 and predicted R^2 should be within 20% of each other to be in reasonable agreement (Bozoglu et al., 2015). These values offer 74.59 and 86.13% of variability in predicting new observation in comparison to approximately 96.01 and 97.21% variability in the original data.

Main effect plot was drawn in order to determine the effects of factors causing significant changes in turbidity and color removal efficiencies when factor levels were changed. Mean turbidity and color removal efficiencies were drawn against pH and temperature with levels and shown in Figure 4.

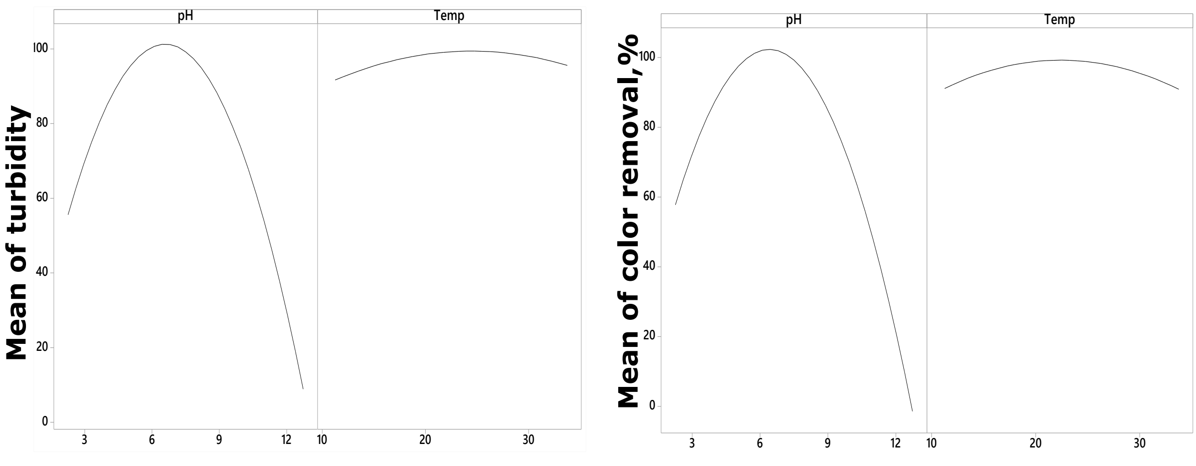

Figure 4: Main effects plot for turbidity and color removal.

As can be seen from the figure, pH and temperature have various effects since the variation in factor levels from low to middle range caused increase in turbidity and color removal efficiencies while a significant decrease is observed in the middle – high factor levels range.

Single interaction plots for two factors were plotted. An interaction plot is a plot of means for each level of a factor with the level of a second factor held constant. Interaction is present when the response at a factor level depends upon the levels of other factors. Parallel lines in an interaction plot indicate no interaction (Bozoglu et al., 2015). The greater the departure of the lines from the parallel state, the higher the degree of interaction. However, the interaction plot does not indicate statistical significance. The interaction of pH and temperature was examined in Figure 5.

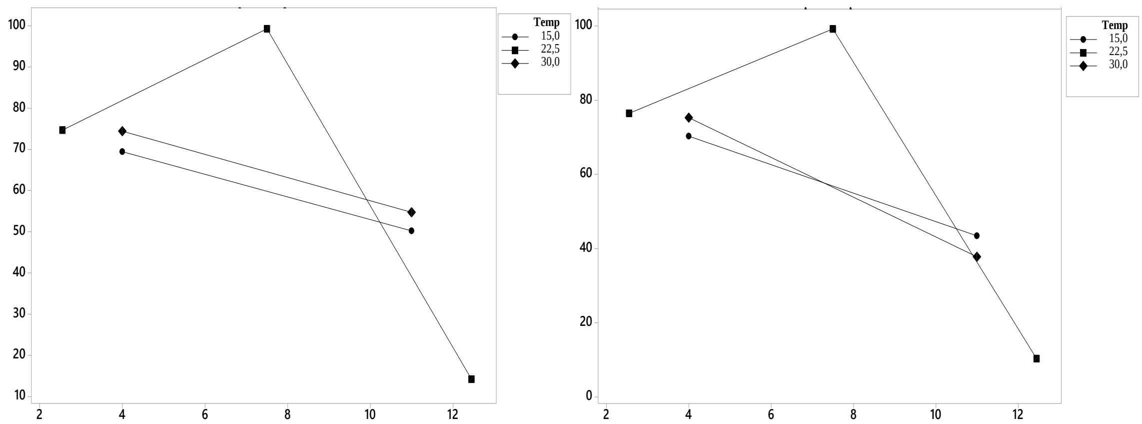

Figure 5: Interaction plot for turbidity and color removal.

The two-factor interaction of pH and temperature was observed for both turbidity and color removal cases. If low and high levels of factors were compared only, the results would be elusory. Although midpoint experiments indicated the interaction between factors, it could not be detected by experiments at low and high range for turbidity removal. The highest mean removal was reached at 22.5 °C and pH 7.5 while the lowest mean removal was obtained at 22.5 °C and pH 12.45.

The optimal condition giving maximum turbidity and color removal was determined by evaluating obtained models in Minitab 17 and reported as 6.45 for pH and 23.25 °C for temperature. The surface response and contour plots of the reduced quadratic models for turbidity and color removal varying with temperature and pH are shown in Figure 6 and 7, respectively.

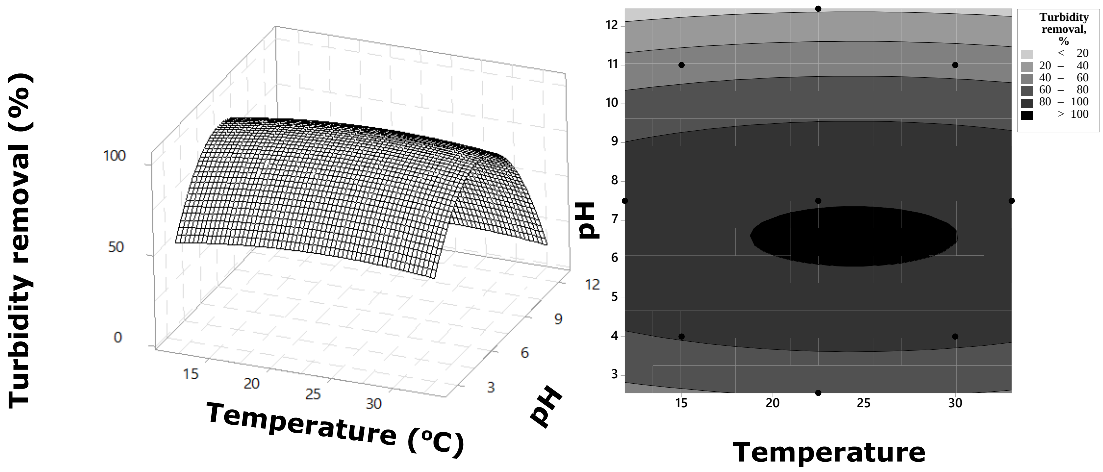

Figure 6: Surface and contour plot for turbidity removal as a function of temperature and pH.

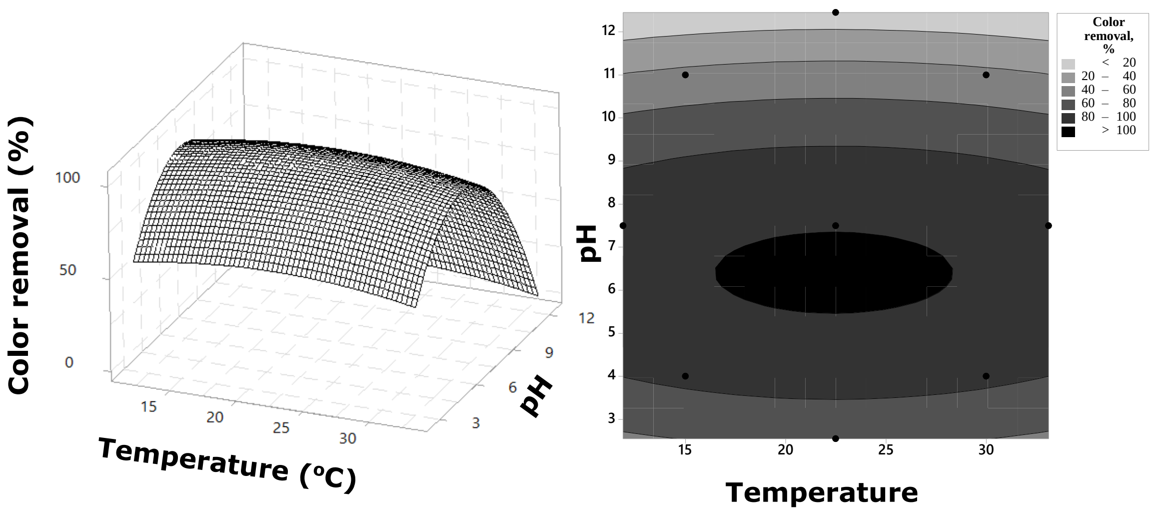

Figure 7: Surface and contour plot for color removal as a function of temperature and pH.

3.2. Effect of Process Controlled Operation on Treatment Performance

The conventional PID feedback control is the most frequently applied feedback control strategy because of its robustness, ease of operation and the lack of specified process knowledge required for the controller designs (Camcıoğlu et al., 2017). Real time experimental MIMO control of conductivity, temperature and pH under constant current conditions are performed using Simulink™ based digital MIMO PID controllers designed in our previous study (Camcioglu et al., 2017b). Algorithms are adapted to a designed real time Simulink™ model which has the ability to transfer real time data of input measurement signals from conductivity, temperature and pH sensors to designed controller and calculated controller output signals to supporting electrolyte, acid and base pumps and cooling water valve simultaneously. Conductivity, temperature and pH are selected as controlled variables; cooling water, supporting electrolyte, acid and base flow-rate are selected as manipulated variables. PID parameters of controllers are given in Table 5.

Table 5: PID parameters of controllers.

Controller	Parameters		
	Proportional	**Integral**	**Derivative**
Conductivity	5	0.05	0.01
Temperature	48	0.5	0.01
pH (Acid)	1000	0.05	0.01
pH (Base)	1	0.06	0.04

In order to determine the effect of constant conductivity, pH and temperature on removal efficiencies and power consumption during electrocoagulation treatment, experiments were performed under controlled and uncontrolled conditions using optimum pH and temperature values and the results were compared in terms of turbidity and color removal. Initial values of pH and temperature were set to 6.45 and 23.24 ℃ respectively at natural wastewater conductivity in both uncontrolled and controlled electrocoagulation studies. Variation of pH and temperature under uncontrolled and controlled conditions are presented in Figures 8 and 9, respectively.

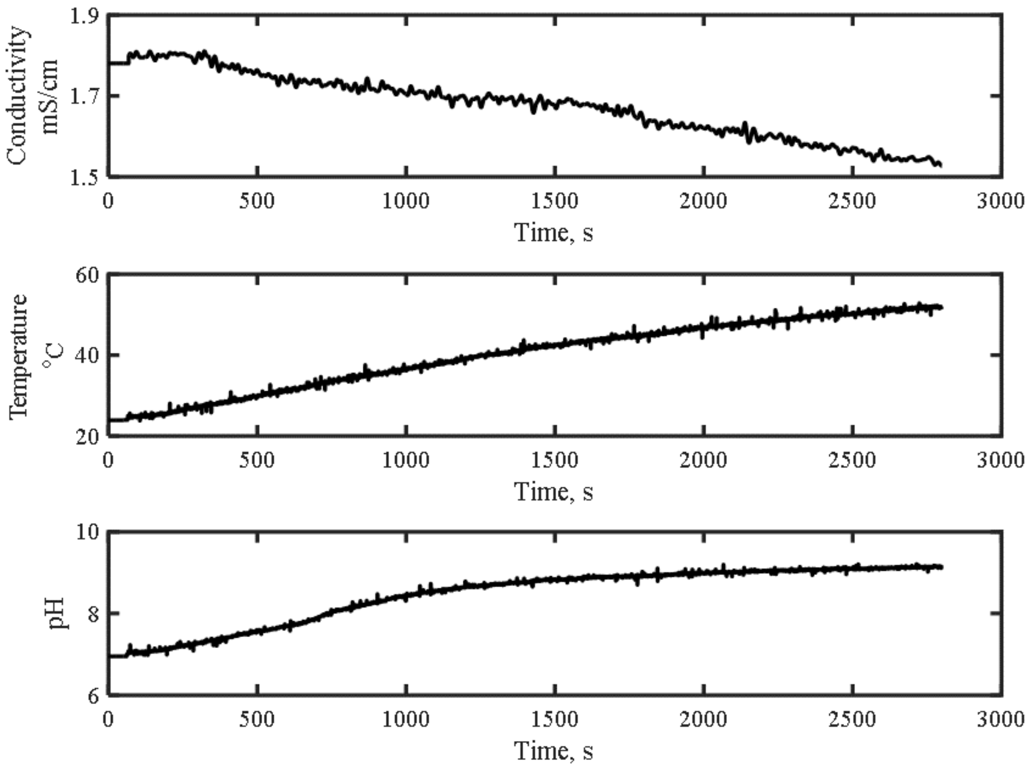

Figure 8: Variation of operating conditions with time for uncontrolled case.

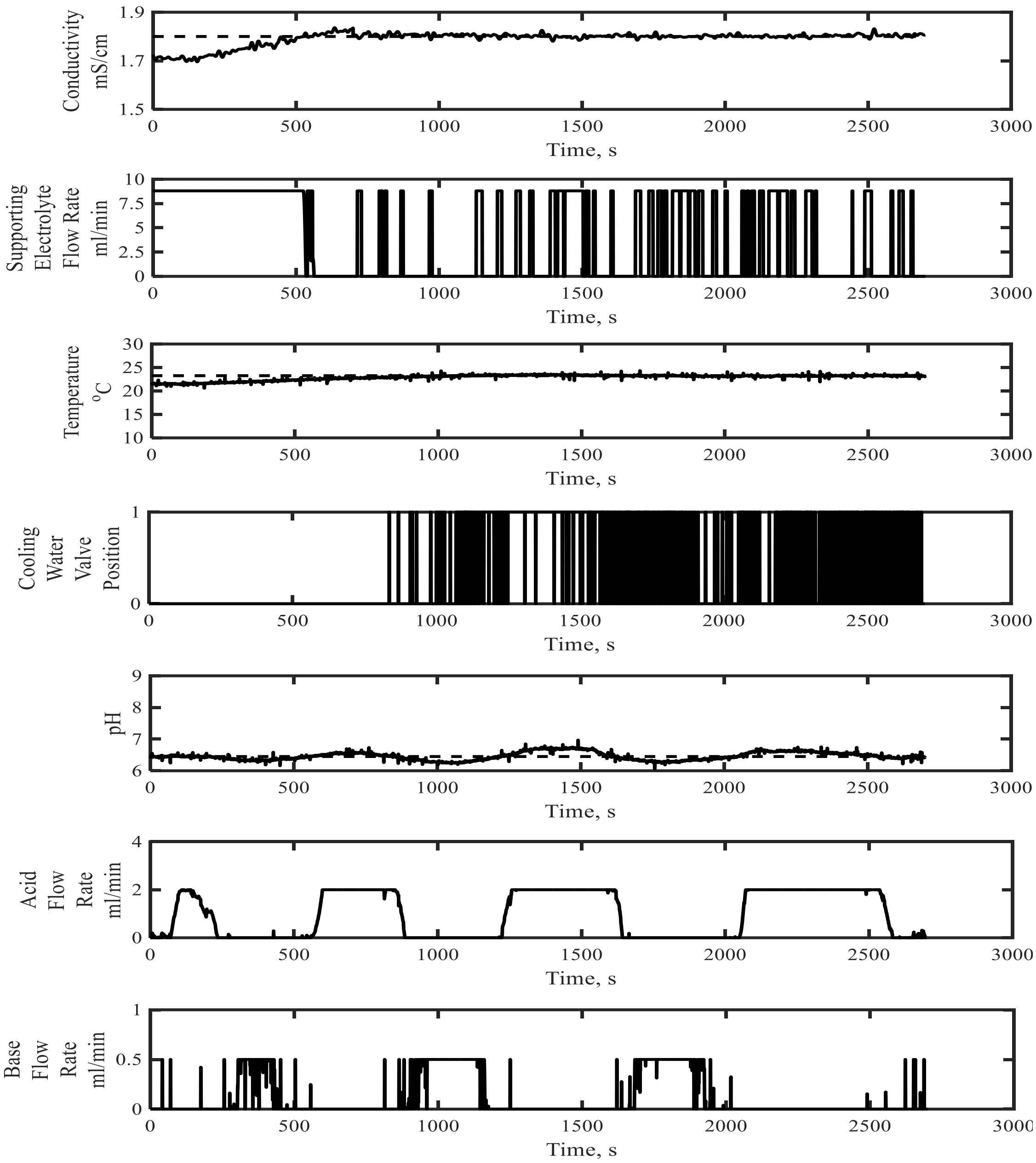

Figure 9: Variation of operating conditions with time for controlled case.

Removal of turbidity and color by time were investigated for uncontrolled and controlled cases at the optimum operating conditions and the results were given in Figure 10.

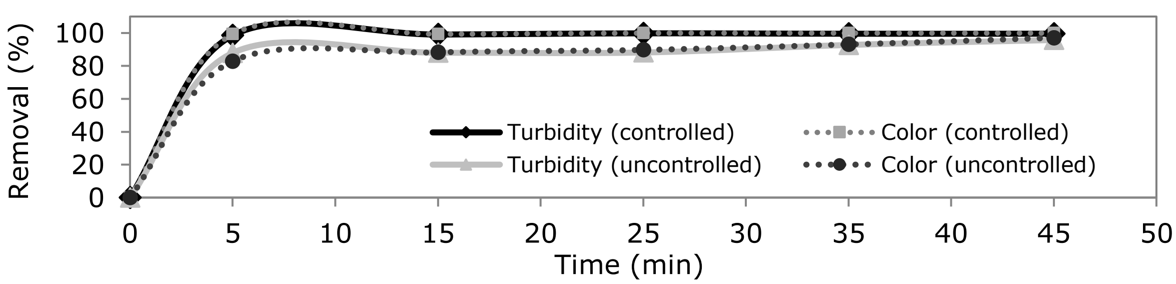

Figure 10: Removal of turbidity and color for uncontrolled and controlled cases.

Results show that 10 minute batch process operation is sufficient for complete removal of turbidity and color under controlled conditions. Power consumption of uncontrolled and controlled cases are calculated as 26.14 kWh/m^3 and 19.53 kWh/m^3 respectively. Results show that conductivity control leads to a 25.3% power consumption reduction. Treatment results of pulp and paper mill wastewater by means of electrocoagulation for 45 minutes under optimum operating conditions are given below in Table 6.

Table 6: Effluent removal results.

Treatment	COD removal (%)	TSS removal (%)	Turbidity removal (%)	Color removal (%)	SO_4^{2-} removal (%)	Cl^- removal (%)
Controlled	34.47	98.06	99.80	99.93	13.40	-
Uncontrolled	23.55	95.09	95.74	97.04	10.23	9.84

Experimental results, throughout the present study, have shown that MIMO control of conductivity, pH and temperature increased removal efficiency of COD, TSS, turbidity, color and SO_4^{2-} compared with uncontrolled condition. Besides less power consumption is required for a higher effluent removal. Cl^- removal cannot be achieved under controlled conditions due to supporting electrolyte addition for conductivity control.

4. CONCLUSION

Removal of COD, TSS, turbidity, color and SO_4^{2-} from pulp and paper mill wastewater is successfully achieved in a batch electrocoagulation reactor. Operating parameters of process were selected as conductivity, pH and temperature respectively. It is observed that under uncontrolled conditions conductivity was dropped 0.28 mS/cm, pH was increased 2.10 units and temperature was increased 28.39 °C during 45 min process. Results show that 87.9% turbidity and 82.9% color removal is achieved in 10 minutes of process operation under uncontrolled conditions. Removal percentages are increased as 7.8 and 14.1% for turbidity and color, respectively. Power consumption is reduced by 25.3% under controlled conditions. The highest removal percentages are obtained under controlled operating conditions as 98.5% and 98.3% for turbidity and color, respectively, for 5 minutes of operation. Under controlled optimum operating conditions, COD, TSS, turbidity, color and SO_4^{2-} removal is increased by 10.92, 2.97, 4.06, 2.89, 3.17, respectively for 45 minutes of process operation in comparison with uncontrolled operation. It is concluded that optimum process operation for removal of turbidity and color of pulp and paper mill wastewater is obtained under constant 6.45 pH, 23.24 °C temperature, 1.78 mS/cm conductivity and 5 minutes time.

5. CONFLICT OF INTEREST

The authors have no conflict of interest.

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