Korean J. Chem. Eng., **30**(3), 664-670 (2013)
DOI: 10.1007/s11814-012-0164-3

Statistical optimization of chemical oxygen demand removal from wastewater by electrochemical oxidation

Dongseog Kim*, Youngwoong Song, and Youngseek Park***,†**

*Department of Environmental Science, Catholic University of Daegu, Gyeongbuk 712-900, Korea
**Department of Occupational Health, Catholic University of Daegu, Gyeongbuk 712-900, Korea
***Faculty of Liberal Education, Daegu University, Gyeongbuk 712-714, Korea
(*Received 17 July 2012 • accepted 27 September 2012*)

Abstract−The independent and combined effects of four variables (current density, electrolyte concentration, air flow rate and pH) on COD removal from wastewater by electrochemical oxidation were optimized using 2^4 full factorial experimental design. ANOVA was conducted to test the combined effects of the independent variables (the four control factors and time) on COD removal. To determine the reaction order of COD removal, 1st, 2nd or 3rd reaction orders were considered; 1st order kinetics showed the highest average r^2 value. The backward elimination regression method was used to determine the 1st order k_{COD} equation, and main effects and 2-way interaction effects on the 1st order equation were investigated. Using this equation, k_{COD} values for the 16 experimental conditions were predicted and COD values were calculated with respect to time. Finally, we tried to determine optimal operating conditions using color and COD removal as endpoints using the multiple response surface method.

Key words: Factorial Design, Reaction Rate Constant, COD Removal, Electrochemical Treatment, Multiple Response Surface Methodology

INTRODUCTION

Efficiency of wastewater treatment by electrochemical oxidation is controlled by several parameters, such as the potential applied, the natures of electrodes used, and pH. Conventional methods of studying electrochemical processes based on maintaining other factors at constant levels cannot represent combinatorial and interaction effects between the factors involved. Furthermore, these methods require several experiments to determine optimum levels, and are thus unreliable and time consuming [1]. The statistical technique commonly called response surface methodology (RSM) is a powerful experimental design tool to optimize and understand the performance of complex systems. RSM is a collection of mathematical and statistical techniques that is used to develop, improve, and optimize processes. Furthermore, it can be used to evaluate the relative significances of several influencing factors in the presence of complex interactions. The main objective of RSM is to suggest an optimum model and to determine the optimum operational conditions for a system or to identify suitable operating parameters [2]. The adequacy of models proposed can be verified using diagnostic tests based on analysis of variances (ANOVA).

RSM has been recently applied to several wastewater treatment systems. The central composite design (CCD) technique has been used to study the effect of Fenton's peroxidation on the removal of organic pollutants [3], and the optimization of the treatment of azo dye wastewater has been performed successfully using CCD [4]. A four-level Box-Behnken factorial design was employed to optimize the medium composition for the degradation of phenol by *Pseudomonas putida* [5].

In previous studies that used RSM [3-5], the removal efficiencies were well predicted using the experimental data at the proposed time. However, it was difficult to predict the removal efficiencies at different times. Furthermore, no study has been previously undertaken to determine the optimal operating conditions required to maximize color and COD removal simultaneously. However, in the present study, the k regression model was used to predict color and COD removal efficiencies even at different several times. In addition, multiple response surface methodology, which has not been previously applied to dye treatment, was used to determine the optimized operating conditions to maximize color and COD removal simultaneously.

This paper concerns the study of four operation variables (current density, electrolyte concentration, air flow rate, pH), which are the main parameters that influence the performance of textile wastewater treatment by electrochemical oxidation. The independent and combined effects of theses four variables on COD removal were optimized. ANOVA was used to test the combined effects of independent variables (the four control factor and time) on COD removal. In addition, the reaction rate constant was optimized by using a 2^4 factorial design, which is useful for estimating main effects and interactions between variables. Furthermore, using multiple response surface methodology, we attempted to derive the optimized operating conditions to maximize the color and COD removal using the predicted reaction rate constant.

EXPERIMENTAL

1. Electrochemical Experiments

The electrochemical reactor used in this study consisted of Ru-Sn-Sb oxide coated titanium mesh electrodes (11 cm×6.3 cm). The

†To whom correspondence should be addressed.
E-mail: ysparkk@daegu.ac.kr

mesh anode and cathode were positioned vertically and parallel to each other with an inter electrode gap of 2 mm. The experiments were done using 1.0 L of Rhodamine B (RhB) dye solution with constant stirring at 150-200 rpm using a magnetic stirrer to maintain uniformity throughout the system. The area of each electrode exposed for the electrolysis was fixed at 69.3 cm^2.

The chemical oxygen demands (CODs) of the RhB sample before and after electrolysis were measured using standard methods [6].

2. Reaction Rate Constant

Reaction rate constant was calculated using the integral method [7]. The integral method is most commonly used when the reaction order is known and specific reaction rate constants are required. The equation for reaction rate (r_A) is in (1).

$$\frac{dC_A}{dt} = r_A = kC_A^n \tag{1}$$

Where, t, C_A, k, and n are time, concentration, the reaction rate constant, and reaction order, respectively.

3. Factorial Design

To investigate the combined effects of the four factors (current density, C; NaCl concentration, N; air flow rate, A; pH, P), the 2^4 full factorial design by Design-Expert software was used in this study. COD measurements were conducted in random order under the 16 experimental conditions (Table 1) over 420 minutes. ANOVA (analysis of variance) was conducted to test the statistical significances of effects (main effects and interaction effects) of the four independent factors on COD removal. During ANOVA, time (2 levels: 120 and 360 minutes) was included as an independent factor.

4. Optimization Using the k-Regression Model

Using the regression models based on the data of factorial design experiment, there are two approaches to determine the optimal conditions of the independent variables. The first involves building a regression model that predicts COD or color removal using the experimental factors as independent variables [8]. Using this method, the time variable must be included as an independent variable in the regression model because the dependent variables (COD or color removal) change with time. However, this method assumes that the relationships between time and the values of dependent variables are linear. The second approach involves building a k-regression model using the reaction rate constant (k) as a dependent variable in the regression model. Furthermore, this approach allows the incorporation of the non-linear relationships between time and the dependent variables [9].

The first step for building the k-regression model was to determine the order of the reaction rate constant using experimental data. In this study, 1st, 2nd, and 3rd order reaction rate constants were calculated using experimental data, and the appropriate reaction order was determined using coefficient of determination (r^2). The k prediction equation was then derived using the backward elimination regression method [10], and this allowed COD values to be predicted using the reaction rate equation such as Eq. (1).

Table 1. Experimental conditions for the 2^4 full factorial design study

Run	Current density, C (mA/cm^2)	NaCl concentration, N (mg/L)	Air flow rate, A (L/min)	pH, P (−)
No. 1	14.43	0.5	0	3
No. 2	14.43	0.5	0	9
No. 3	14.43	0.5	2	3
No. 4	14.43	0.5	2	9
No. 5	14.43	1.75	0	3
No. 6	14.43	1.75	0	9
No. 7	14.43	1.75	2	3
No. 8	14.43	1.75	2	9
No. 9	43.29	0.5	0	3
No. 10	43.29	0.5	0	9
No. 11	43.29	0.5	2	3
No. 12	43.29	0.5	2	9
No. 13	43.29	1.75	0	3
No. 14	43.29	1.75	0	9
No. 15	43.29	1.75	2	3
No. 16	43.29	1.75	2	9

RESULTS AND DISCUSSION

1. Results of the 2^4 Factorial Experiment

The relation between experimental COD results and electrolysis time is shown in Fig. 1. ANOVA was conducted to test the combined effects of the independent variables (four control factors and a time factor) on COD removal, and Table 2 summarizes results. The effects of all five main variables were statistically significant ($p<0.01$).

To examine the effects of variables on COD removal, the results at two points were compared (current density, 14.43 and 43.29 mA/cm^2; NaCl concentration, 0.5 and 1.75 g/L; air flow rate, 0 and 2 L/min; pH, 3 and 9; time, 120 and 360 min) (Fig. 2). The COD values were lower for a current density of 43.29 mA/cm^2, an NaCl concentration of 1.75 g/L, an air flow rate of 2 L/min, a pH of 3 and time of 360 min.

As was expected, interactions were found between the five variables, and five 2-way interaction effects (C*N, C*P, N*P, C*T, and

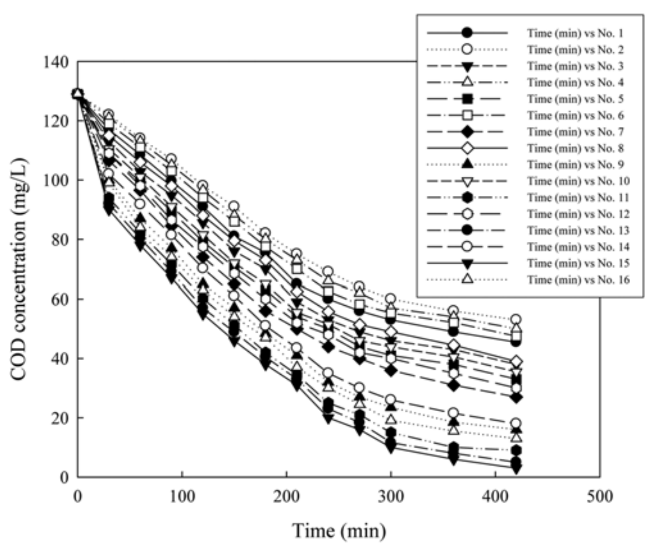

Fig. 1. Experimental results of COD concentration versus electrolysis time.

Table 2. ANOVA results for main and interaction effects

Source	df	SS	MS	F	p
Current density (C)	1	4740.9	4740.9	125380.4	0.0018**
NaCl concentration (N)	1	843.6	843.6	22309.5	0.0043**
Air flow rate (A)	1	203.5	203.5	5382.2	0.0087**
pH (P)	1	1475.6	1475.6	39024.2	0.0032**
Time (T)	1	15891.0	15891.0	420257.5	0.0010**
C*N	1	12.1	12.1	320.7	0.0355*
C*P	1	33.4	33.4	883.7	0.0214*
C*T	1	39.8	39.8	1053.3	0.0196*
N*P	1	8.1	8.1	214.2	0.0434*
N*T	1	6.6	6.6	173.8	0.0482*
C*N*A	1	8.5	8.5	225.0	0.0424*
C*N*P	1	101.9	101.9	2694.6	0.0123*
C*P*T	1	8.1	8.1	214.2	0.0434*
N*A*P	1	15.5	15.5	411.0	0.0314*

Effects that are not statistically significant (p>0.05) were omitted (the five-way interaction effect was used as an error source)
** p<0.01, * p<0.05

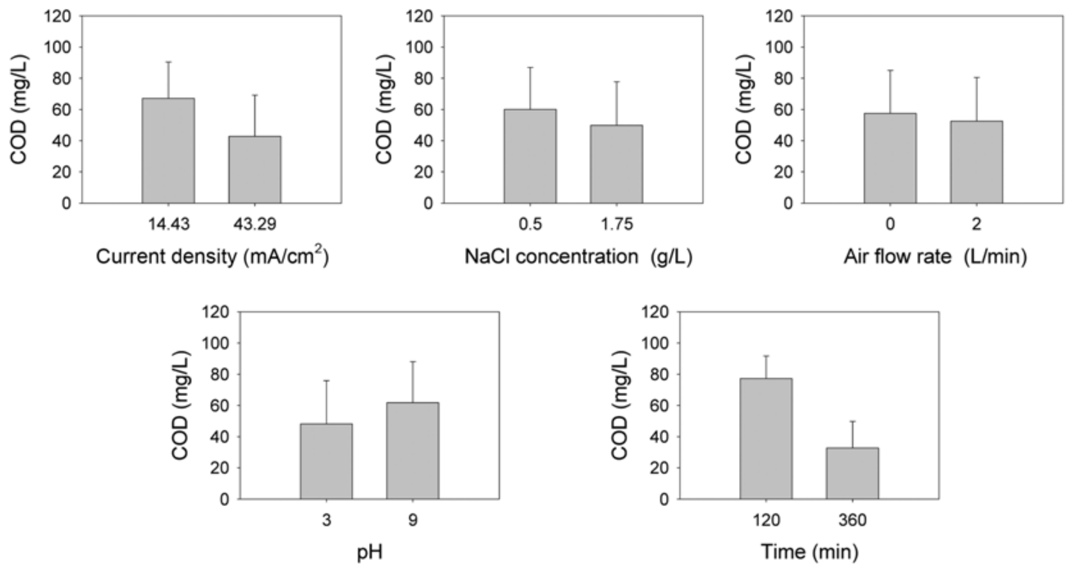

Fig. 2. The main effects of the five independent variables.

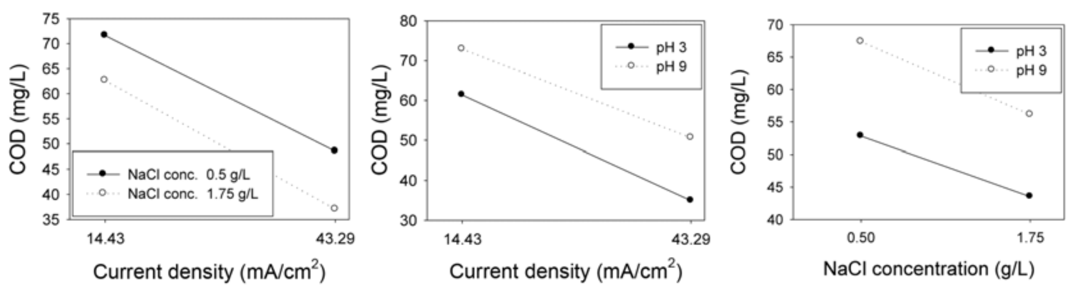

Fig. 3. Two-way interaction effects of C*N (current density and NaCl concentration), C*P (current density and pH), and N*P (NaCl concentration and pH).

N*T) and four 3-way interaction effects (C*N*A, C*N*P, N*A*P, and C*P*T) were statistically significant (p<0.05) (Table 2). Fig. 3 shows the typical 2-way interaction effects of C*N, C*P, N*P. COD decreased as current density was increased from 14.43 mA/cm^2 to 43.29 mA/cm^2. However, the difference between COD values at NaCl concentrations of 0.5 g/L and 1.75 g/L (and at pH values of

Table 3. Reaction rate constants (k) calculated from experimental data for 1st, 2nd, and 3rd orders reactions

Run number	Current density (mA/cm^2)	NaCl conc. (g/L)	Air flow rate (L/min)	pH, (−)	1st Order		2nd Order		3rd Order	
					k	r^2	k	r^2	k	r^2
1	14.43	0.5	0	3	0.00284	0.966	0.000035	0.983	4.6×10^{-7}	0.946
2	14.43	0.5	0	9	0.00238	0.977	0.000027	0.977	3.3×10^{-7}	0.942
3	14.43	0.5	2	3	0.00328	0.967	0.000044	0.980	6.4×10^{-7}	0.922
4	14.43	0.5	2	9	0.00252	0.978	0.000029	0.978	3.7×10^{-7}	0.937
5	14.43	1.75	0	3	0.00366	0.965	0.000052	0.984	8.3×10^{-7}	0.907
6	14.43	1.75	0	9	0.00267	0.973	0.000032	0.979	4.1×10^{-7}	0.939
7	14.43	1.75	2	3	0.00415	0.974	0.000065	0.977	11.9×10^{-7}	0.861
8	14.43	1.75	2	9	0.00312	0.979	0.000041	0.978	5.8×10^{-7}	0.908
9	43.29	0.5	0	3	0.00543	0.986	0.000111	0.912	30.6×10^{-7}	0.736
10	43.29	0.5	0	9	0.00349	0.961	0.000048	0.986	7.3×10^{-7}	0.924
11	43.29	0.5	2	3	0.00669	0.988	0.00019	0.804	87.1×10^{-7}	0.596
12	43.29	0.5	2	9	0.00386	0.972	0.000057	0.981	9.7×10^{-7}	0.881
13	43.29	1.75	0	3	0.00745	0.985	0.000275	0.686	200.7×10^{-7}	0.442
14	43.29	1.75	0	9	0.00507	0.988	0.000097	0.921	24.0×10^{-7}	0.752
15	43.29	1.75	2	3	0.00818	0.975	0.000398	0.580	472.5×10^{-7}	0.346
16	43.29	1.75	2	9	0.00589	0.988	0.000135	0.880	43.8×10^{-7}	0.688
Average						0.976		0.912		0.795
S.D.						0.009		0.121		0.189

S.D.: standard deviation

3 and 9) was larger at a current density of 43.29 mA/cm^2 than at a current density of 14.43 mA/cm^2, indicating that the effects of NaCl concentration and pH on COD values increased with current density. In addition, the COD difference between pH values 3 and 9 was smaller at an NaCl concentration of 1.75 g/L than at 0.50 g/L. However, the difference between COD values at NaCl concentration of 0.5 g/L and 1.75 g/L and at pH values of 3 and 9 was larger at a current density of 43.29 mA/cm^2 than at a current density of 14.43 mA/cm^2.

The practical implications of 3-way interaction effects were difficult to understand in this experiment. Nevertheless, the significances of interaction effects suggest that combinations of factors should be considered when investigating COD removal efficiency and optimizing set-up conditions.

2. Determination of the Reaction Rate Constant for COD Removal

To determine the reaction order of COD removal, reaction orders of 1st, 2nd or 3rd were considered. Table 3 shows the reaction rate constants (k_{COD}) for 1st, 2nd, and 3rd orders kinetics calculated by Eq. (1) using experimental data. The 1st order kinetics had the highest average r^2 value of 0.976, and thus, a 1st order reaction was considered appropriate.

3. Optimization Using Single Response Surface Methodology

The optimal condition of the current density, NaCl concentration, air flow rate, and pH is the combination of the particular values of the four variables in which condition COD removal is achieved most efficiently. In terms of the reaction rate constant k_{COD}, COD removal becomes more efficient as k_{COD} increases. Thus, we tried to find values for these four factors that maximize the reaction rate constant k_{COD}. First-order k_{COD} was selected for the optimization because it had the largest r^2 value, as described above.

The 1st order k_{COD} prediction equation (response surface) was derived using backward the elimination regression method [10] with an alpha out value of 0.1. The initial terms used were the four main effects, six 2-way interaction effects, four 3-way interaction effects, and one 4-way interaction effect. The 1st order k_{COD} prediction equation derived, shown below, and was found to be significant by ANOVA ($p<0.001$) with r^2 value of 0.994 (adjusted $r^2=0.988$).

$$k_{COD}=0.0016165+0.000103985*C+0.000062*N+0.00008625*A -0.00000583333*P+0.0000314622*C*N +0.00000718988*C*A-0.000008.95126*C*P \quad (2)$$

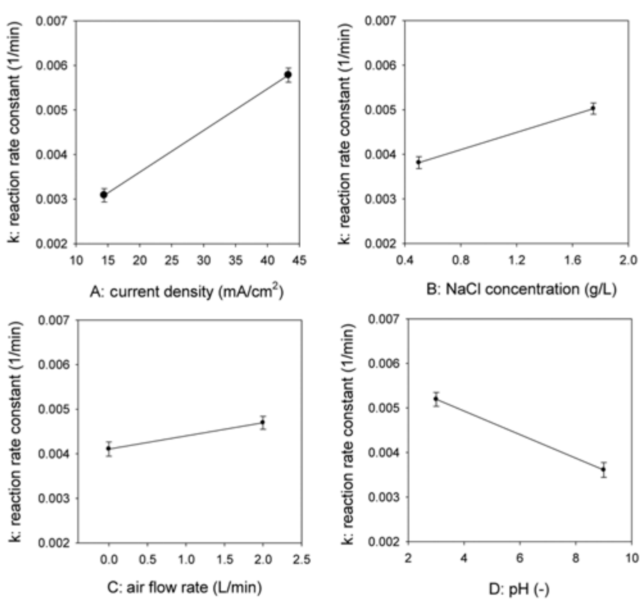

Fig. 4. Main effects of the four independent factors on the 1st order reaction rate constant k.

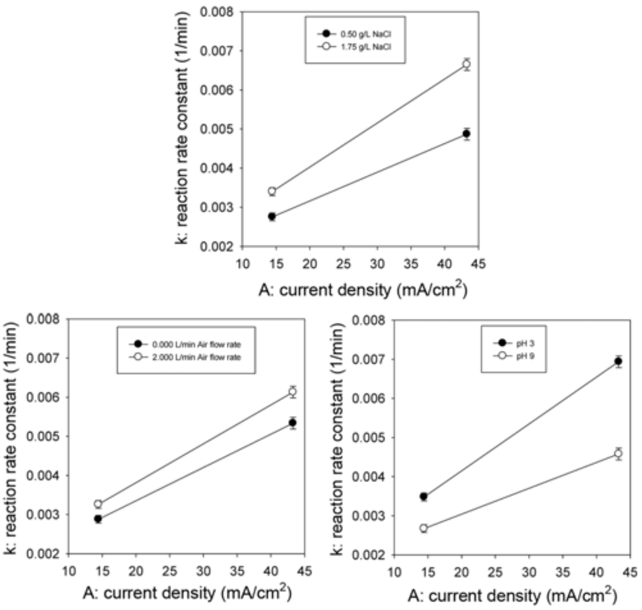

Fig. 5. Two-way interaction effects of the four independent factors on the 1st order reaction rate constant k.

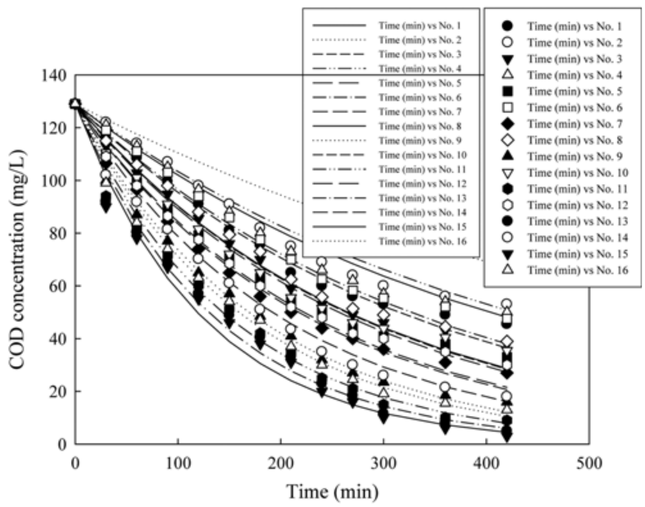

Fig. 6. Predicted results of COD concentration by k-regression model analysis (symbols and lines represent experimental and predicted data, respectively).

Fig. 4 and Fig. 5 show the main effects and 2-way interaction effects on the prediction equation, respectively. At the main effects of the four independent variables (Fig. 4), k_{COD} values increased on increasing current density, NaCl concentration and air flow rate, but k_{COD} decreased on increasing pH.

k_{COD} increased by increasing the current density because the increment of current density increases the generation of several oxidants (O_3, free chlorine, ClO_2, H_2O_2, OH radical and others) [11]. Increasing the NaCl concentration increased k_{COD} due to the increased mass transport of chloride ions to the anode surface and to the increased diffusion in the anode diffusion layer. Thus, more oxidants are generated by increasing NaCl concentration [11], which increases COD removal efficiency. The effect of air flow rate on k_{COD} in the present study agreed with that found by Chen and Liang [12] and Sudoh et al. [13], who found that the production of electro-generated H_2O_2 was proportional to the mass transfer rate of dissolved oxygen to the cathode surface. Chen and Liang [12] found that TOC removal efficiencies in electrochemical reactions without oxygen dosing were slightly lower than those with oxygen dosing. k_{COD} decreased drastically when the initial pH was 11.0. Although chlorine/hypochlorite generation is stable at given current density, but at higher pH values hypochlorite acid is converted into chlorate or hypochlorate, which reduces the availability of hypochlorite and reduces COD removal. Another reason may be that under acid conditions chlorine is present in solution as hypochlorous acid, which has higher oxidation potential than hypochlorite [8]. On the other hand, hypochlorite predominates under alkaline conditions [14].

The slope of current density on k_{COD} was greatest followed, in descending order, by pH, NaCl concentration, and air flow rate. Therefore, our study shows that current density has the greatest effect on COD removal rate, and that the air flow rate has least effect.

In view of 2-way interaction effect (Fig. 5), the slope of current density on k_{COD} increased on increasing NaCl concentration and air flow rate, but decreased on increasing pH. Based on these slope differences, pH was found to have the effect on COD removal followed by NaCl concentration and air flow rate.

Using Eq. (2), k_{COD} values for the 16 experimental conditions were predicted and COD levels were calculated using a 1st order reaction rate equation in the time domain of 0-420 min (Fig. 6). Prediction errors were calculated for the 16 experimental conditions, and the total average error value was found t be only 4.5 mg/L (min= 2.7 mg/L and max=13.0 mg/L), and the total average error percentage to be 8.9% (min=6.2% and max=18.8%). Thus, the 1st order k_{COD} regression model appeared to be suitable as a response surface model for the optimization of COD removal.

Using the k_{COD} Eq. (2) as an objective function, the optimal operating condition that maximized the objective function (k_{COD}) was predicted by Design Expert software. The predicted optimal operating condition was as follows: current density 43.21 mA/cm^2, NaCl 1.75 g/L, air flow rate 1.99 L/min, and pH 3.06, at which k was predicted to be 0.00818 1/min.

Using the k_{COD} regression model and the linear approach model suggested by Prasad and Srivastava [8], we predicted the times required to accomplish different target COD removal percentages with three different time combinations (60-240 min, 120-360 min, 180-420 min). Table 4 shows the prediction results. Furthermore, the mean prediction error of the k_{COD} regression model was lowest at 19.7 min, which was lower than that obtained using the linear approach [8]. Thus, in the linear approach model, the two time values should be selected carefully, because its mean prediction error can be more than three times those of other models (the k_{COD} regression model or a model based on another linear approach model with different time values).

4. Optimization Using Multiple Response Surface Method

Above we described the optimal solution that maximizes COD removal rate or 1st order k_{COD}. However, the optimal solution may be dependent on the end-point used, for example, the color removal. Thus, we tried to find an optimal solution by considering two end-points (color removal, COD removal) using the multiple response surface method [15]. This method optimizes the desirability func-

Table 4. The prediction results of six target COD removal percentages using the optimal conditions. The initial COD level was assumed to be 129 mg/L

Target COD removal efficiency (%)	Target COD concentration (mg/L)	Experiment time (min)	Time predicted (min)			
			k-Regression model	Linear model [14]		
				T1=60, T2=240	T1=120, T2=360	T1=180, T2=420
50	64.5	96.0	82.9	96.0	51.6	0.0
60	51.6	132.0	109.6	136.0	117.7	97.8
70	38.7	180.0	143.9	176.1	183.9	195.6
80	25.8	222.0	192.4	216.2	250.0	293.4
90	12.9	288.0	275.3	256.3	316.2	391.2
95	6.45	354.0	358.2	276.6	349.2	440.4
Mean prediction error (min)			19.7	20.5	20.6	67.8

tion which combines multiple end-points. The k_{COD} (k value for COD removal) prediction equation was derived in the previous section, and the k_{Color} (k value for color removal) prediction equation was developed using published data [9]. The regression options for the k_{Color} were the same as those for k_{COD} and the derived prediction equation was as follows:

$$k_{Color}=-0.080516+0.017036*C+0.30228*N+0.079199*A -0.00835176*P-0.00624455*C*N-0.000603925*C*P -0.022886*N*P-0.00495092*A*P+0.000751481*C*N*P \quad (3)$$

Using the k_{COD} and k_{Color} prediction equations, the desirability function D was defined as Eq. (4). The $k_{COD\,min}$ ($k_{COD\,max}$) and $k_{Color\,min}$ ($k_{Color\,max}$) are the minimum (maximum) values of k_{COD} and k_{Color}, respectively.

$$D=\left(\left(\frac{k_{COD}-k_{COD\,min}}{k_{COD\,max}-k_{COD\,min}}\right)\times\left(\frac{k_{Color}-k_{Color\,min}}{k_{Color\,max}-k_{Color\,min}}\right)\right)^{1/2} \quad (4)$$

The simultaneous objective function D is a geometric mean of the two transformed responses. The ranges of the desirability function D are from 0 to 1 (least to most desirable, respectively). The optimal operating condition, which maximizes the desirability function, was found and is summarized in Table 5 with optimum solutions for single responses (k_{COD} and k_{Color}). The optimal operating condition using k_{COD} was similar to that for k_{Color} and the optimal operating condition using the two end-points (k_{COD} and k_{Color}) was similar to the individual end-points (k_{COD} or k_{Color}).

CONCLUSIONS

The interactions among four factors (current density, electrolyte concentration, air flow rate, pH) were investigated with respect to COD removal using a 2^4 factorial experimental design. ANOVA was conducted to test the combined effects of independent variables (the four control factors and time) on COD removal. All five main effects were statistically significant ($p<0.01$) and combined effects of the independent factors were evident ($p<0.05$). In addition, we used the regression models as an alternative response surface model for the optimization of operating conditions using four factors and interaction terms. For COD removal, 1^{st} order k_{COD} achieved better prediction results than 2^{nd} or 3^{rd} order k_{COD}. Using the k_{COD} prediction equation as an objective function, the optimal operating condition was predicted as follows: current density 43.21 mA/cm^2, NaCl 1.75 g/L, air flow rate 1.99 L/min, and pH 3.06, and the k_{COD} value was predicted to be 0.00818 1/min under optimal operating condition. Using the 1^{st} order k_{COD}, the required times to accomplish several target COD removal percentages were well predicted within the mean prediction error time of 19.7 min. Multiple response surface method was used to optimize the desirability function which combined two objective functions (k_{COD} and k_{Color}). The optimal operating condition identified using both objective functions (k_{COD} and k_{Color}) was similar to that using individual objective functions (k_{COD} or k_{Color}).

ACKNOWLEDGEMENTS

This work was supported by research grants from the Catholic University of Daegu in 2010.

REFERENCES

1. K. Ravikumar, S. Ramalingam, S. Krishman and K. Balu, *Dyes Pigm*., **70**(1), 18 (2006).
2. D. C. Montgomery, *Design and analysis of experiments*, 6th Ed., Wiley, New York (1991).

Table 5. Optimization results for the single and multiple response surface models

Objective function	Optimal condition				Objective function value
	Current density, C (mA/cm^2)	NaCl concentration, N (mg/L)	Air flow rate, A (L/min)	pH, P (–)	
k_{COD} (single)	43.21	1.75	1.99	3.06	k_{COD}=0.00818
k_{Color} (single)	43.28	1.75	2.0	3.0	k_{Color}=0.788631
Desirability (multiple)	43.29	1.74	2.0	3.0	D=0.997

3. M. Ahmadi, F. Vahabzadeh, B. Bonakdarpour, E. Mofarrah and M. Mehranian, *J. Hazard. Mater*., **B123**, 187 (2005).
4. I. Arslan-Alaton, G. Tureli and T. Olmez-Hanci, *J. Photochem. Photobiol., A: Chem*., **202**, 142 (2009).
5. G. Annadurai, L. Y. Ling and J. H. Lee, *J. Hazard. Mater*., **151**, 171 (2008).
6. A. Eaton, L. Clesceri and A. Greenberg, *Standard methods for examination of water and wastewater*, 19th Ed., United Book Press, Maryland (1995).
7. H. Scott Fogler, *Elements of chemical reaction engineering*, Prentice-Hall, Englewood Cliffs, New Jersey (1986).
8. R. Krishna Prasad and S. N. Srivastava, *Chem. Eng. J*., **146**, 22 (2009).
9. Y. Song, D. Kim and Y. Park, *Korean J. Chem. Eng*., **28**(1), 156 (2011).
10. R. Myers, *Classical and modern regression with application*, PWS-KENT Publishing Company, Boston (1990).
11. D. S. Kim and Y. S. Park, *J. Env. Sci*., **18**(11), 1235 (2009).
12. W.-S. Cheng and J.-S. Liang, *J. Hazard. Mater*., **161**, 1017 (2009).
13. M. Sudoh, H. Kitaguchi and K. Koide, *J. Chem. Eng. Jpn*., **18**, 409 (1985).
14. L. Szpyrkowicz, C. Juzzolino, S. N. Kaul, S. Daniele and M. D. De Faveri, *Ind. Eng. Chem. Res*., **39**, 3241 (2000).
15. R. Myers and D. Montgomery, *Response surface methodology*, John Wiley & Sons (1995).