ELECTRODE LIFETIME INFLUENCES ON ELECTROCOAGULATION EFFICIENCY IN WASTEWATER TREATMENT: MODELING AND OPTIMIZATION

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ABSTRACT

In this work, reactive red 198 dye removal by electrocoagulation were studied focusing electrode lifetime influences on dye removal efficiency. Other seven empirical parameters including initial dye concentration, initial pH, initial conductivity, initial salt concentration, applied voltage, electric current intensity, and process time were investigated experimentally and statistically. Achieved experimental dye removal data of 0-100% gained under different experimental conditions. The data was used for model building by employing reduced quadratic multiple regression model. Further statistical tests were applied to exhibit models goodness and significant variables. Experiments show the surprising results regarding lifetime influences on dye removal efficiency as well as other investigated parameters. The constructed model was shown the acceptable reliability and predictability. Finally, optimization of dye removal efficiency was carried out using Taguchi algorithm.

Keywords: Electrode lifetime; Taguchi Design of Experiments; reactive red 98; Reduced multiple linear regression; Dye removal; Electrocoagulation

1. Introduction

In electrocoagulation process, some factors including initial pH (pH_0), initial dye concentration (C_0), applied voltage (V_{EC}), applied electric current (I_{EC}), initial electrolyte concentration (C_S), initial conductivity (C) and treatment time (t_{EC}) influence the process efficiency [1, 2].

The influences of electrode lifetime on electrocoagulation efficiency have been neglected in electrocoagulation related researches. Application of fresh electrode and refresh the electrode surface were applied to deactivate the influences of electrode lifetime. But, there are no research to probe the electrode lifetime influences. The electrocoagulation efficiency is increased by the optimization of these factors [3].

Then, the goal of this work is to study the electrode lifetime influences as well as other empirical parameters including initial dye concentration, initial pH, initial conductivity, initial salt concentration, applied voltage, electrical current intensity and process time.

2. Materials and methods

2.1. Materials

The reactive red 198 dye (figure 1) with $C_{27}H_{18}ClN_7Na_4O_{15}S_5$ chemical formula and 968.21 molecular weight was supplied from Alvan-Sabet Co., Iran [4]. Stock solution of reactive red 198 was prepared by dissolving 1 gr dye powder per litter of distilled water solution of reactive red 198. The pH_0 of the solutions was adjusted using NaOH (3M) and H_2SO_4 (3M) (Merck, Germany). The C_S was adjusted to desire value using NaCl (Merck, Germany) [5].

Schematic of applied glass reactor is presented in figure 2. The reactor equipped with 100 rpm mechanical mixer, DC power supply (RXN-303D-II, Zhaoxin Electronic Tech. Co.), and two Aluminum sheets electrodes. The distance between electrodes was fixed at 1cm distance.

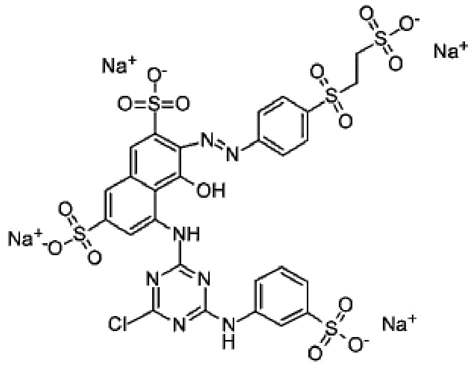

Figure 1: Structure formula of RR198

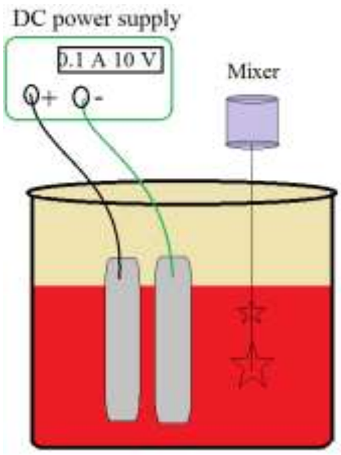

Figure 2: Schematic of EC unit

2.2. Experimental design

L_{16} (4×4) Taguchi design were applied for investigation of C_0, pH_0, V_{EC}, and C_S using Minitab 14. Whole parameters were investigated in four levels. Details of Taguchi design presented in table 1. Influences of reaction time was investigated by sampling during 120 min. Initial and final conductivity and pH, as well as electrical current intensity and electrode lifetime were empirically determined and monitored in whole runs.

Table 1: Whole 16 Taguchi runs designed for experiments.

Run	C0	CS	pH	V	Run	C0	CS	pH	V
1	10	0	2	5	**9**	70	40	2	30
2	10	20	5	10	**10**	70	60	5	15
3	10	40	7	15	**11**	70	0	7	10
4	10	60	9	30	**12**	70	20	9	5
5	40	20	2	15	**13**	100	60	2	10
6	40	0	5	30	**14**	100	40	5	5
7	40	60	7	5	**15**	100	20	7	30
8	40	40	9	10	**16**	100	0	9	15

2.3. EC process and data collection

In each run, 500 mL solution containing RR198 was decanted into the EC reactor, then operational parameters were adjusted to the desired value according to table 1, process was started by applying the desired voltage on the electrodes, and the parameters including initial conductivity, electric current and electrode lifetime were determined. In each eight determined t_{EC}, 10 ml sample was extracted and centrifuged. The DR was determined for the decanted solution. The concentration of solution was evaluated using PG T80$^+$ spectrophotometer in the UV-VIS range and using calibration curve method. The DR was calculated for samples using equation 1:

$$DR = (1-C/C_0) \tag{1}$$

where C_0 and C are concentration of solution before and after process, respectively [5].

2.4. Methodology of modeling

After removing the missing data, 98 remaining DR data together with corresponding experimental conditions were used as a data set for modeling and optimization. The significant operational parameters were distinct using analysis of variances and stepwise multiple linear regression. The significant parameters were applied to generate linear model. Finally, the consistency of the models was revealed by tests quantified with statistical parameters [6, 7].

3. Results and discussions

3.1. EC process

The whole 98 obtained experimental data are given graphically in figure 3 and summarized in table 2. The influences of operational parameter especially is visible in figure 3 that cause to high variation in DR (table 2). As can be seen from figure 3, DR vary from 0 to 1 dependent to experimental condition.

Table 2: Qualitative statistic parameters of investigated variables

	Mean	Std. Deviation
DR	0.34	0.34
Conductivity	2800	6900
Initial pH	5.7	2.6
Voltage	14.9	9.7
Initial concentration	47	32
Reaction time	47.6	43.2
Salt concentration	0.014	0.01
Electric current intensity	0.07	0.19
Lifetime	460	270

In order to statistical study of variable parameters, some statistical treatment were done based on experimental data. Stepwise multiple linear regression based on analysis of variance (ANOVA) was used to identify the significant variables as well as constructing best linear model. The results of stepwise multiple linear regression were presented in table 3.

Among eight investigated variables, based on regression coefficient presented in table 3, five variables have significant influences on DR including initial concentration, initial conductivity, applied voltage, reaction time and electrode lifetime. Among these five significant parameters, initial concentration and conductivity have negative influences on dye removal efficiency. Negative influences of conductivity may rise from competition between salt ions and dye molecules to adsorb on coagulant. Negative influences of initial concentration may rise from improper dependent variable when relative removal selected instead the absolute removal.

Applied voltage, reaction time and electrode lifetime have positive influences on DR. More voltage cause to more electric current and coagulant production then more dye removal. More reaction time in one hand cause to more coagulant production and in other hand cause to more contact time between dye molecule and produced coagulants. Positive influences of electrode life time is surprising results that is against with our primary imagines. It may rise from surface alternation during lifetime of electrode.

The electrical current intensity and salt concentration variables are insignificant variables with insignificant Pearson correlation coefficient (table 4). It may rise because of high co-linearity between these parameters and parameters including voltage and conductivity. However initial pH have significant correlation with DR but it have negligible effect to improve the model goodness then it doesn’t cooperate in linear model (table 4).

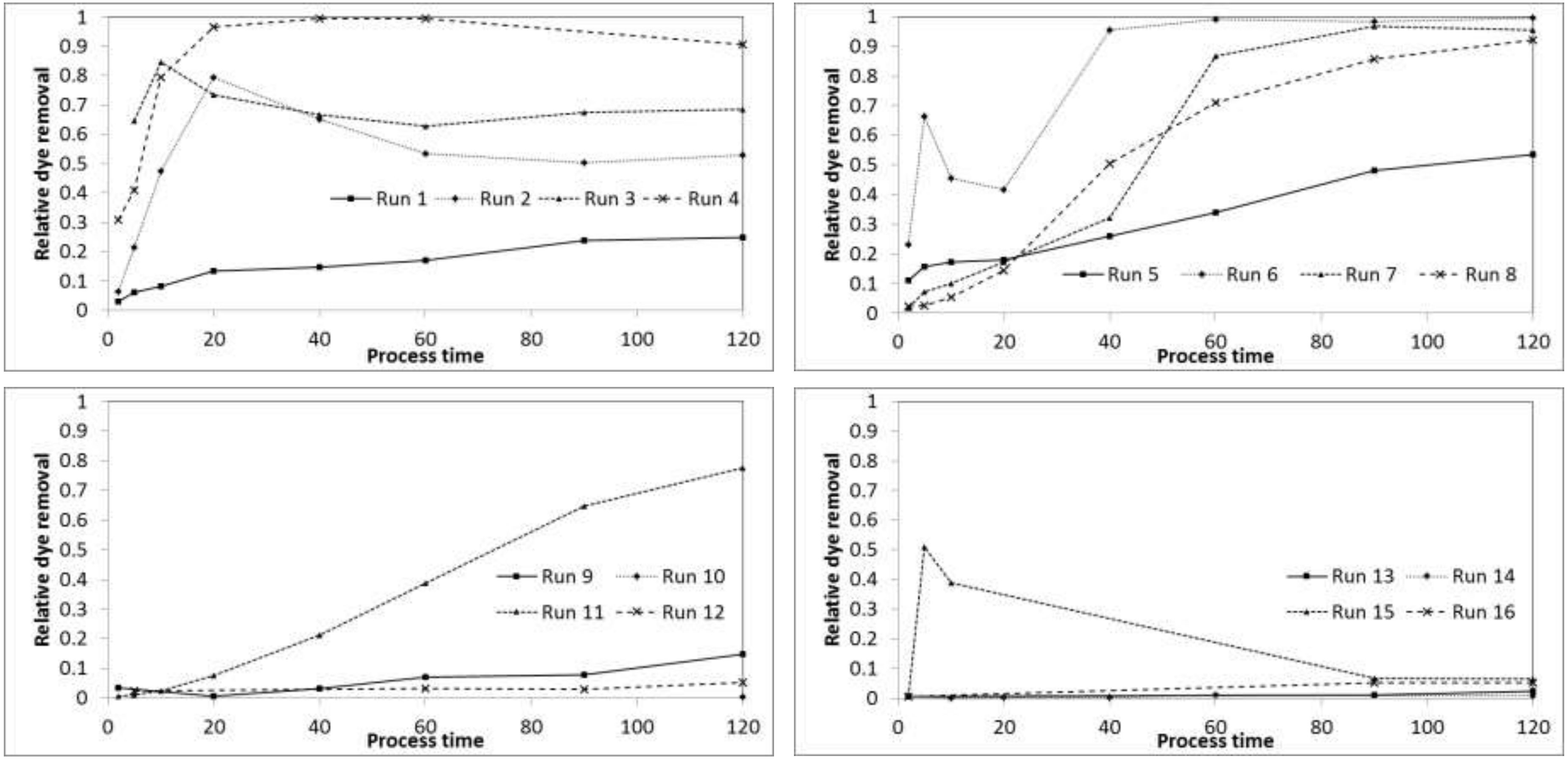

Figure 3: The whole obtained 98 dye removal data of 16 runs

Table 3: Statistic details of linear and reduced quadratic model				
	Reduced quadratic model		**Linear model**	
variable	**Regression coefficient**	**p-value**	**Regression coefficient**	**p-value**
Constant	0.2033	0.02	0.3624	0
Initial concentration	-4.715	0	-7.8×10^{-3}	0
Initial conductivity	-1×10^{-5}	0	-1.3×10^{-5}	0
Reaction time	6.62×10^{-3}	0	2.5×10^{-3}	0
Voltage	8.72×10^{-3}	0	8.5×10^{-3}	0
Lifetime	6.6×10^{-3}	0.02	3×10^{-4}	0
(Reaction time)^2	-4×10^{-5}	0.0	-	-
(Lifetime)^2	0	0.01	-	-
Initial concentration × Reaction time	-5.891×10^{-2}	0	-	-
Reaction time × Lifetime	1×10^{-5}	0	-	-

Table 4: Correlation coefficient of independent and dependent variables										
		DR	**Conductivity**	**Initial pH**	**Voltage**	**Initial concentration**	**Reaction time**	**Salt concentration**	**Electric current intensity**	**Lifetime**
Pearson Correlation	**DR**	1.00	-0.20	0.28	0.30	-0.51	0.27	0.12	-0.10	0.11
	Conductivity		1.00	-0.58	-0.29	-0.35	-0.04	-0.41	0.42	-0.41
	Initial pH			1.00	0.01	0.02	-0.01	0.25	-0.43	0.49
	Voltage				1.00	0.03	-0.02	0.01	0.00	0.01
	Initial concentration					1.00	0.11	-0.03	-0.15	0.40
	reaction time						1.00	0.02	-0.18	0.14
	Salt concentration							1.00	-0.14	0.23
	electric current intensity								1.00	-0.05
	Lifetime									1.00
Sig. (1-tailed)	**DR**	.	0.03	0.00	0.00	0.00	0.00	0.13	0.16	0.13
	Conductivity		.	0.00	0.00	0.00	0.35	0.00	0.00	0.00
	Initial pH			.	0.45	0.42	0.46	0.01	0.00	0.00
	Voltage				.	0.40	0.42	0.45	0.48	0.47
	Initial concentration					.	0.14	0.37	0.08	0.00
	reaction time						.	0.43	0.04	0.09
	Salt concentration							.	0.09	0.01
	electric current intensity								.	0.32
	Lifetime									.

The obtained linear model with R^2 equal 0.62 and adjusted R^2 equal 0.60 shows acceptable linear model for this complex process. Improve the model by using quadratic and interaction parameters were done in following. The best improved model shows R^2 equal 0.77 and adjusted R^2 equal 0.74. The statistical details of both linear and quadratic model presented in table 3.

Based on table 3, four additional quadratic and interaction parameters related to reaction time, lifetime, and initial concentration have been used in reduced quadratic model that indicate the importance of these parameters.

4. Conclusion

In this study, successful application of electrocoagulation for dye removal is reported with maximum removal efficiency. As a novel idea, electrode life time and its influences on process efficiency was investigated. Statistical treatments clearly certify the significant direct influences of electrode life time. However, the anode electrode scarify during electrode life time but it help to make the electrode more efficient. Surface alteration and refreshing by electrochemical activity may interpret this phenomenon. Then, future studies based on surface probing during process can be helpful and strongly recommended.

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