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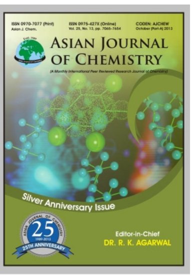

Removal of Silica from Cooling Water by Electrocoagulation: A Comprehensive and Systematic Study Using Response Surface Methodology

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To effectively recycle and reuse water, the prevention of silica deposits become an urgent requirement in many industries. The laboratory scale electrocoagulation system was investigated to remove silica. And the interactions between all the influencing factors were analyzed not only for silica removal efficiency, but also for energy consumption, electrode consumption and sludge production. The values of adj R^2 of the models were 0.9315 for silica removal efficiency, 0.9768 for energy consumption, 0.7633 for electrode consumption and 0.9698 for sludge production, respectively, which indicated that all the models were significant. The optimal conditions were 60 mg/L initial silica concentration, 2.50 cm interelectrode distance, 40.47 min reaction time, 7.50 pH value, 3.89 mA/cm^2 current density and 4 cm submerged height of electrode, under which 85.147 % silica removal efficiency, 1.455 KWh/m^3 energy consumption, 0.0389 kg/m^3 electrode consumption and 0.242 kg/m^3 sludge production were obtained and the desirability was 0.831 for the optimal conditions. By performing the optimum conditions, 83.243 % silica removal efficiency, 1.466 KWh/m^3 energy consumption, 0.0400 kg/m^3 electrode consumption and 0.245 kg/m^3 sludge production were obtained.

Key Words: Electrocoagulation, Silica, Response surface methodology, Optimization.

INTRODUCTION

With the increasing water pollution, the serious scarcity of water and increasing pressure for water conservation, the recycle and reuse of water have attracted more and more attention by the operators in different industries[1]. Surprisingly, 70 % of industrial water is used as cooling water. However, cooling water recycling can lead to concentration of dissolved species to a critical point of precipitation of insoluble mineral salts[2]. Silica scale is a big technical challenge and a big financial burden for industrial operators for it is a dense and rigid form. It can reduce boiler power output by 10-20 % and thermal efficiency by 10 %[3-6], plug the pipeline[7-9] and make more shutdowns[2]. Thus, it is urgent to remove silica from cooling water in order to recycle and reuse water.

Prevention of silica scale commonly follows two distinct approaches. The first one is adding anti-scalant in water to inhibit scale formation and the other is to install a pretreatment process to reduce the silica concentration before application[10]. The choice of anti-scalant is highly dependent on the processes and congurations, which makes this approach a little bit precarious. The pretreatment of cooling water include chemical precipitation[11,12] and silica-gel seeding[13]. The optimum dosage of precipitant depends on the silica concentration and the background matrix of the solution, as well as the required extent of silica reduction. Pretreatment by silica-gel seeding intends to provide a seeding surface on which silica monomers deposit, thereby removing dissolved silica from solution. However, this method was ineffective when silica polymers formed in the solution under highly supersaturated conditions.

Recently, electrocoagulation technology is investigated for water purification and wastewater treatment as a promising technology[14-17]. And sporadic studies have reported that it can be used as a pretreatment method for silica removal. Walter Den and Chia-Jung Wang[11] used electrocoagulation as a pretreatment process to treat the source brackish water containing 100 mg/L silica for reverse osmosis process and the extent of flux decline was markedly improved. Gelover-Santiago *et al.*[18] tested three electrochemical systems to remove silica in makeup water for cooling tower and the most advantageous system is the one with both aluminium electrodes working with direct current. Liao *et al.*[19] reported the effectiveness of electrocoagulation using iron and aluminium electrodes for treating cooling tower blowdown water containing dissolved silica, Ca^{2+} and Mg^{2+} with different initial pH values and additives. However, no one studied the effect of design and operation factors of the electrocoagulation process to reduce power consumption, electrode consumption and sludge production in the field of silica removal from cooling water. And few study reported the interrelation of the influencing factors simultaneously to recycle cooling water.

The paper presents a systematic study of the factors which affect the electrocoagulation process using response surface methodology. The optimal condition was obtained to maximize silica reduction efficiency and minimize energy consumption, electrode consumption and sludge production. The study was proposed as an economical and efficient technology for silica removal from cooling water.

EXPERIMENTAL

Setup and procedure: Fig. 1 shows an overview of experimental set-up. The electrocoagulation reactor is made of plexiglass with a dimension of 8 cm × 20 cm × 16 cm (W × L × H) and the effective volume is 1.5 L. The two aluminium electrodes were prepared with identical size of 18 cm × 18 cm and the effective area is adjusted by changing the submerged height of the electrode. Before using, the impurities on the surfaces of aluminium were removed with sand paper, then dipped for 5 min in a solution which was mixed HCl aqueous solution (35 %, 50 mL) with aqueous solution of hexamethylenetetramine (2.8 %, 100 mL) together[20]. Electrical current is provided by a manually digital DC power supply. Artificial water containing different concentrations of silica is treated for different time, under different operating conditions. During the experiments, the initial pH of solution is adjusted to desirable values using diluted NaOH or H_2SO_4 solution. After each run, the samples were filtered with 0.45 μm membrane to separate the solids and then they were dried at 30 ºC for 48 h to determine the sludge production[21].

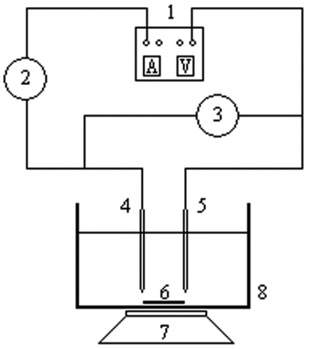


Fig. 1. Schematic diagram of experimental set-up. 1. Digital DC power supply; 2. Digital amperemeter; 3. Digital voltmeter; 4,5 Electrodes; 6. Magnetic bar; 7. Magnetic stirring controller; 8. Water tank

Calculation method: The removal efficiency of silica is calculated as

$$\eta(\%) = \frac{C_i - C_f}{C_i} \times 100 \tag{1}$$

where C_i is the initial silica concentration (mg/L) and C_f is the final silica concentration (mg/L).

The energy consumption ENC (KWh/m^3) is calculated from

$$ENC = \frac{\sum (U \times I \times t')}{V} \tag{2}$$

where U is the voltage (V), I is the electrolysis current (A), t' is the time interval (h) and V is the volume of the solution treated (it is 1.5 L).

The electrode consumption ELC (kg/m^3) was obtained by weighing the quality before and after the experiment and expressed as follows:

$$ELC = \frac{(m - m')}{V} \tag{3}$$

where m and m' are the weight of electrode before and after the experiment (g) and V is the volume of the solution treated.

The sludge production Ws (kg/m^3) is calculated from

$$Ws = \frac{M}{V} \tag{4}$$

where M is the weight of sludge produced from the experiments (g) and V is the volume of the solution treated.

Design and optimization: The electrolysis experiments were carried out according to a five-level, six-factor central composite rotatable design (CCD) using Design-Expert 7.0. Each independent variable was coded at five levels between -2 and +2 at the ranges according to the preliminary experiments, where the independent variable were initial silica concentration C_0 (x_1 = 20-100 mg/L), interelectrode distance d (x_2 = 0.5-2.5 cm), reaction time t (x_3 = 15-75 min), initial pH (x_4 = 4.5-10.5), current density i (x_5 = 2-6 mA/cm^2) and submerged height h (x_6 = 4-8 cm). Table-1 shows run number and experimental conditions of the runs arranged by the CCD. Performance of the process was evaluated by analyzing the responses: silica removal efficiency, energy consumption, electrode consumption and sludge production.

The quadratic model which also includes the linear model for predicting the optimal point is expressed as:

$$y_i = b_0 + \sum_i b_i x_i + \sum_i b_{ii} x_i^2 + \sum_i \sum_j b_{ij} x_i x_j \tag{5}$$

where y_i is the predicted response, b_0 is the interception coefficient, b_i is the linear term, b_{ii} is the quadratic term and b_{ij} is the interaction term.

Validation of the model: In the case of a composite design, the validation of the model is carried out by an appropriate analysis of variance (ANOVA). The model is considered adequate if the variance due to regression is significantly different from the total variance. 'Statistica' software is performed for regression and graphical analysis of data obtained. The optimum conditions is studied by analyzing the response surface plots.

RESULTS AND DISCUSSION

Regression coefficients estimating and interaction of variables analyzing: The regression coefficients and corresponding F and P values for silica removal efficiency, energy consumption, electrode consumption and sludge production are given in Tables 2-5. For silica removal efficiency, reaction time, initial pH, current density, the interaction between reaction time and initial pH, the interaction of initial silica concentration, the interaction of reaction time, the interaction of initial pH and the interaction of current density are significant model terms. For energy consumption, interelectrode

TABLE-1
ACTUAL DESIGN OF EXPERIMENTS AND RESPONSES FOR ELECTROCOAGULATION PROCESS

Run No.	x_1	x_2	x_3	x_4	x_5	x_6	y_1: η	y_2: ENC	y_3: ELC	y_4: Ws
1	60	1.5	45	10.5	4	6	100	2.30	0.10	0.51
2	60	1.5	45	7.5	2	6	79.30	0.67	0.05	0.21
3	80	2.0	60	6	5	5	100	3.66	0.12	0.54
4	60	0.5	45	7.5	4	6	95.06	0.96	0.09	0.40
5	40	1.0	60	9	5	7	100	2.88	0.11	0.56
6	60	1.5	45	7.5	4	8	97.09	3.13	0.12	0.53
7	80	1.0	60	9	3	7	100	1.28	0.11	0.49
8	60	1.5	45	7.5	4	6	99.23	2.26	0.09	0.39
9	60	1.5	45	7.5	4	6	99.06	2.21	0.09	0.40
10	60	2.5	45	7.5	4	6	100	3.26	0.11	0.39
11	60	1.5	45	4.5	4	6	73.89	1.99	5.45	0.37
12	40	1.0	30	9	5	5	100	1.44	0.06	0.30
13	80	1.0	60	6	5	7	100	3.03	0.17	0.77
14	60	1.5	45	7.5	4	4	95.12	1.47	0.05	0.27
15	40	2.0	30	9	5	7	100	3.66	0.08	0.41
16	80	2.0	30	6	5	7	77.78	2.86	0.08	0.37
17	60	1.5	45	7.5	4	6	99.15	2.27	0.10	0.39
18	60	1.5	45	7.5	4	6	99.20	2.24	0.08	0.39
19	80	2.0	60	9	3	5	95.81	0.91	0.07	0.38
20	40	1.0	30	6	3	5	59.30	0.55	0.05	0.15
21	40	2.0	60	9	5	5	100	3.92	0.09	0.45
22	20	1.5	45	7.5	4	6	92.08	2.12	0.09	0.39
23	60	1.5	15	7.5	4	6	62.04	0.70	0.03	0.14
24	80	1.0	30	6	5	5	70.42	1.08	0.05	0.28
25	40	2.0	60	6	3	5	84.84	1.85	0.06	0.30
26	60	1.5	75	7.5	4	6	100	3.62	0.15	0.64
27	100	1.5	45	7.5	4	6	85.82	1.73	0.08	0.42
28	80	2.0	30	9	3	7	86.39	1.58	0.05	0.28
29	40	2.0	30	6	3	7	69.32	1.35	0.05	0.21
30	60	1.5	45	7.5	6	6	97.63	4.67	0.15	0.59
31	40	1.0	60	6	3	7	95.26	1.59	0.09	0.43
32	60	1.5	45	7.5	4	6	99.19	2.25	0.09	0.39
33	80	1.0	30	9	3	5	77.03	0.46	0.05	0.22

distance, reaction time, current density, submerged height of electrode, the interaction between initial silica concentration and initial pH, the interaction between interelectrode distance and current density, the interaction between interelectrode distance and submerged height of electrode, the interaction between reaction time and initial pH, the interaction between reaction time and current density and the interaction of initial silica concentration are significant model terms. For electrode consumption, reaction time, current density and submerged height of electrode are significant model terms. For sludge production, reaction time, initial pH, current density, submerged height of electrode, the interaction between initial silica concentration and reaction time, the interaction between reaction time and current density, the interaction between reaction time and initial pH and the interaction between initial pH and current density are significant model terms. It can be seen from the above analysis that reaction time and current density are the two main factors affecting the four responses severly.

According to the experimental results, the regression equations relating to the four responses are developed and given in eqns. 6-9. It is noteworthy that the predicting model is a linear one for electrode consumption.

$$\begin{aligned}
y_1 &= 97.85 - 1.56x_1 + 1.23x_2 + 9.49x_3 + 6.53x_4 + 4.58x_5 \\
&+ 1.74x_6 + 0.81x_1x_2 + 2.04x_1x_3 - 0.43x_1x_4 + 0.13x_1x_5 \\
&+ 0.030x_1x_6 - 0.84x_2x_3 - 0.11x_2x_4 + 0.16x_2x_5 + 1.01x_2x_6 \\
&- 4.43x_3x_4 - 2.00x_3x_5 + 0.48x_3x_6 - 1.48x_4x_5 - 0.89x_4x_6 \\
&1.66x_5x_6 - 1.81x_1^2 + 0.33x_2^2 - 3.79x_3^2 - 2.31x_4^2 \\
&- 1.93x_5^2 - 0.65x_6^2 \qquad (6)
\end{aligned}$$

$$\begin{aligned}
y_2 &= 2.31 - 0.098x_1 + 0.57x_2 + 0.73x_3 + 0.078x_4 \\
&+ 1.00x_5 + 0.41x_6 - 0.073x_1x_2 - 0.021x_1x_3 + 0.19x_1x_4 \\
&+ 0.068x_1x_5 + 0.057x_1x_6 + 0.14x_2x_3 + 0.035x_2x_4 \\
&+ 0.24x_2x_5 + 0.35x_2x_6 - 0.15x_3x_4 + 0.17x_3x_5 \\
&+ 0.11x_3x_6 + 0.051x_4x_5 + 0.061x_4x_6 + 0.019x_5x_6 - 0.12x_1^2 \\
&- 0.071x_2^2 - 0.059x_3^2 - 0.063x_4^2 + 0.069x_5^2 - 0.024x_6^2 \qquad (7)
\end{aligned}$$

$$\begin{aligned}
y_3 &= 0.087 + 0.0035x_1 - 0.00233x_2 + 0.026x_3 - 0.00325x_4 \\
&+ 0.017x_5 + 0.013x_6 \qquad (8)
\end{aligned}$$

$$\begin{aligned}
y_4 &= 0.39 + 0.007x_1 - 0.004x_2 + 0.12x_3 + 0.036x_4 \\
&+ 0.097x_5 + 0.065x_6 - 0.00769x_1x_2 + 0.021x_1x_3 \\
&+ 0.021x_1x_4 + 0.033x_1x_5 + 0.0068x_1x_6 + 0.00819x_2x_3 \\
&+ 0.00944x_2x_4 - 0.00194x_2x_5 + 0.018x_2x_6 - 0.022x_3x_4 \\
&+ 0.014x_3x_5 + 0.011x_3x_6 - 0.026x_4x_5 \\
&- 0.0073x_4x_6 + 0.011x_5x_6 \qquad (9)
\end{aligned}$$

TABLE-2
ESTIMATED REGRESSION COEFFICIENTS AND CORRESPONDING F AND P VALUES FOR SILICA REMOVAL EFFICIENCY

Term	Coefficient	Sum of squares	F	Prob > F
Intercept	97.85	–	–	–
x_1	-1.56	19.59	1.87	0.2298
x_2	1.23	12.18	1.16	0.3303
x_3	9.49	720.67	68.77	0.0004
x_4	6.53	340.92	32.53	0.0023
x_5	4.58	167.99	16.03	0.0103
x_6	1.74	24.35	2.32	0.1880
x_1x_2	0.81	10.46	1.00	0.3637
x_1x_3	2.04	66.87	6.38	0.0528
x_1x_4	-0.43	1.00	0.096	0.7696
x_1x_5	0.13	0.094	0.008977	0.9282
x_1x_6	0.030	0.015	0.00139	0.9717
x_2x_3	-0.84	3.76	0.36	0.5751
x_2x_4	-0.11	0.20	0.019	0.8955
x_2x_5	0.16	0.42	0.040	0.8493
x_2x_6	1.01	5.45	0.52	0.5030
x_3x_4	-4.43	314.20	29.98	0.0028
x_3x_5	-2.00	64.27	6.13	0.0561
x_3x_6	0.48	1.21	0.12	0.7483
x_4x_5	-1.48	11.75	1.12	0.3381
x_4x_6	-0.89	12.68	1.21	0.3215
x_5x_6	-1.66	44.32	4.23	0.0949
x_1^2	-1.81	98.76	9.42	0.0278
x_2^2	0.33	3.37	0.32	0.5950
x_3^2	-3.79	433.55	41.37	0.0013
x_4^2	-2.31	161.06	15.37	0.0112
x_5^2	-1.93	112.53	10.74	0.0220
x_6^2	-0.65	12.63	1.20	0.3224

TABLE-3
ESTIMATED REGRESSION COEFFICIENTS AND CORRESPONDING F AND P VALUES FOR ENERGY CONSUMPTION

Term	Coefficient	Sum of squares	F	Prob > F
Intercept	2.31	–	–	–
x_1	-0.098	0.077	2.83	0.1531
x_2	0.57	2.64	97.49	0.0002
x_3	0.73	4.28	157.92	<0.0001
x_4	0.078	0.049	1.82	0.2353
x_5	1.00	8.00	295.13	<0.0001
x_6	0.41	1.37	50.46	0.0009
x_1x_2	-0.073	0.086	3.16	0.1355
x_1x_3	-0.021	0.0069	0.26	0.6346
x_1x_4	0.19	0.19	7.10	0.0447
x_1x_5	0.068	0.025	0.92	0.3812
x_1x_6	0.057	0.052	1.91	0.2252
x_2x_3	0.14	0.11	3.94	0.1041
x_2x_4	0.035	0.020	0.72	0.4347
x_2x_5	0.24	0.94	34.84	0.0020
x_2x_6	0.35	0.65	23.85	0.0045
x_3x_4	-0.15	0.38	14.03	0.0133
x_3x_5	0.17	0.48	17.73	0.0084
x_3x_6	0.11	0.062	2.30	0.1900
x_4x_5	0.051	0.014	0.51	0.5059
x_4x_6	0.061	0.059	2.18	0.1996
x_5x_6	0.019	0.00597	0.22	0.6587
x_1^2	-0.12	0.42	15.47	0.0110
x_2^2	-0.017	0.15	5.66	0.0632
x_3^2	-0.059	0.10	3.87	0.1064
x_4^2	-0.063	0.12	4.48	0.0879
x_5^2	0.069	0.14	5.33	0.0690
x_6^2	-0.024	0.018	0.65	0.4557

TABLE-4
ESTIMATED REGRESSION COEFFICIENTS AND CORRESPONDING F AND P VALUES FOR ELECTRODE CONSUMPTION

Term	Coefficient	Sum of squares	F	Prob > F
Intercept	0.087	–	–	–
x_1	0.0035	0.000294	1.15	0.2943
x_2	-0.00233	0.000131	0.51	0.4819
x_3	0.026	0.016	62.41	<0.0001
x_4	-0.00325	0.000254	0.99	0.3295
x_5	0.017	0.00728	28.37	<0.0001
x_6	0.013	0.00406	15.80	0.0005

TABLE-5
ESTIMATED REGRESSION COEFFICIENTS AND CORRESPONDING F AND P VALUES FOR SLUDGE PRODUCTION

Term	Coefficient	Sum of squares	F	Prob > F
Intercept	0.39	–	–	–
x_1	0.007	0.000392	0.67	0.4319
x_2	-0.004	0.000128	0.22	0.6502
x_3	0.12	0.12	211.40	<0.0001
x_4	0.036	0.010	17.36	0.0016
x_5	0.097	0.075	127.15	<0.0001
x_6	0.065	0.034	56.95	<0.0001
x_1x_2	-0.007688	0.000946	1.61	0.2313
x_1x_3	0.021	0.00735	12.49	0.0047
x_1x_4	0.021	0.00228	3.88	0.0747
x_1x_5	0.033	0.00592	10.05	0.0089
x_1x_6	0.0068	0.000743	1.26	0.2854
x_2x_3	0.00819	0.000358	0.61	0.4523
x_2x_4	0.00944	0.00142	2.42	0.1481
x_2x_5	-0.00194	0.00006	0.10	0.7554
x_2x_6	0.018	0.00164	2.79	0.1228
x_3x_4	-0.022	0.0077	13.07	0.0041
x_3x_5	0.014	0.00322	5.47	0.0393
x_3x_6	0.011	0.00061	1.03	0.3310
x_4x_5	-0.026	0.00369	6.27	0.0293
x_4x_6	-0.0073	0.000856	1.45	0.2534
x_5x_6	0.011	0.00196	3.32	0.0955

Models analyzing: Table-6 shows the F-value of the models for silica removal efficiency, energy consumption, electrode consumption and sludge production are 17.13, 50.89, 18.20 and 49.87, respectively, which implied the models are significant. For all models, the values of Prob > F are less than 0.0500, indicating that terms are significant in all models. In this study, the values of adj R^2 are 0.9315 for silica removal efficiency, 0.9768 for energy consumption, 0.7633 for electrode consumption and 0.9698 for sludge production which ensures a satisfactory adjustment of the model to the experimental data. The coefficient of variance (CV) as the ratio of the standard error of estimate to the mean value of the observed response is a measure of reproducibility of the model. As shown in Table-6, the values of coefficient of variance for the four responses are 3.58, 7.77, 18.32 and 6.18 %, respectively. A ratio of AP > 4 indicates that adequate model is desirable. Then the AP values in the study are 14.480, 26.929, 16.196 and 32.132 for silica removal efficiency, energy consumption, electrode consumption and sludge production, respectively.

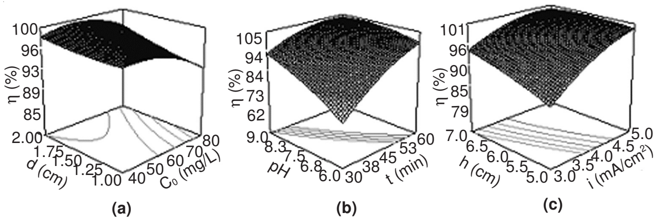
3D response surfaces of responses: The 3D response surfaces are generally the graphical representation of the regression equation. This representation shows the relative effects of any two variables when the remaining variables are

TABLE-6
ANOVA RESULTS OF THE MODELS FOR THE FOUR RESPONSES

Source	Degree of freedom	Sum of squares	Mean square	F value	Prob > F
Silica removal efficiency (%)[a]					
Model	27	4845.80	179.47	17.13	0.0025
Residual	5	52.40	10.48		
Lack of fit	1	52.38	52.38	12233.63	<0.0001
Pure error	4	0.017	0.004282		
Energy consumption (KWh/m^3)[b]					
Model	27	37.25	1.38	50.89	0.0002
Residual	0.14	5	0.027		
Lack of fit	0.13	1	0.13	260.18	<0.0001
Pure error	0.00205	4	0.000513		
Electrode consumption (kg/m^3)[c]					
Model	6	0.028	0.00467	18.20	<0.0001
Residual	26	0.00667	0.000257		
Lack of fit	22	0.00654	0.000297	8.61	0.0246
Pure error	4	0.000138	0.0000345		
Sludge production (kg/m^3)[d]					
Model	21	0.62	0.029	49.87	<0.0001
Residual	11	0.00648	0.000589		
Lack of fit	7	0.00643	0.000919	81.33	0.0004
Pure error	4	0.0000452	0.0000113		

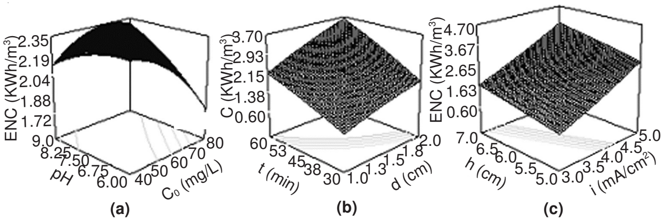
[a]R^2 = 0.9893, adjR^2 = 0.9315, CV = 3.58 %, AP = 14.480. [b]R^2 = 0.9964, adjR^2 = 0.9768, CV = 7.77 %, AP = 26.929. [c]R^2 = 0.8077, adjR^2 = 0.7633, CV = 18.32 %, AP = 16.196. [d]R^2 = 0.9896, adjR^2 = 0.9698, CV = 6.18 %, AP = 32.132.

kept constant. The response surface plots are given in Fig. 2 for the silica removal efficiency, Fig. 3 for energy consumption and Fig. 4 for sludge production. For the linear relationship between electrode consumption and the variables, here presents no 3D response surfaces for electrode consumption.



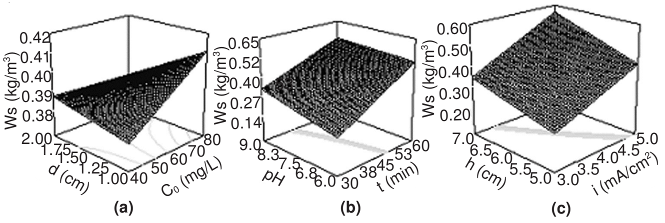
(a) (b) (c)

Fig. 2. 3D response surface graphs for interaction of variables on silica removal efficiency



(a) (b) (c)

Fig. 3. 3D response surface graphs for interaction of variables on energy consumption



(a) (b) (c)

Fig. 4. 3D response surface graphs for interaction of variables on sludge production

Fig. 2(a) shows the effect of initial silica concentration and interelectrode distance on the silica removal efficiency when fixing the remaining variables at constant level. The response has little change with interelectrode distance, indicating that the other variables sufficient to ensure high removal efficiency. When interelectrode distance is about 1.4 cm and initial silica concentration is between 40 and 70 mg/L, a region of more than 98 % silica removal efficiency is obtained. A neutral value of pH and a long reaction time enable higher removal efficiency, according to Fig. 2(b). The 98 % removal efficiency is attained in the range of pH from 9 to 6 and reaction time from 38 to 60 min. It may because that larger mass of electrode was dissolved in the solution to combine with silica for a longer reaction time. The pH value plays an important role in electrocoagulation process, for the types and the formation rates of silica and Al species mainly depend on the pH value. The monomeric anion of $Al(OH)_4^-$ will form when the pH is high and the soluble cation of Al^{3+} will form when pH is low[22]. As shown in Fig. 2(c), the removal efficiency increases with the increase of current density and submerged height of electrode. The region of submerged height from 7 to 5 and current density from 3.9 to 4.5 mA/cm^2 ensures silica removal efficiency higher than 98 %. At higher current density and larger effective area, higher dissolution rates of electrode material and higher formation rates of aluminum hydroxides result in higher removal efficiency of silica.

From Fig. 3(a), higher pH and lower initial silica concentration or lower pH and higher initial silica concentration result in lower energy consumption. However, relatively higher pH leads to higher silica removal efficiency from Fig. 2(b). Therefore, the region with higher pH and lower initial silica concentration is desirable in the experiment. Energy consumption has a linear relationship with reaction time, interelectrode distance, current density and submerged height of electrode, according to the Fig. 3(b) and (c). Because of ENC $\propto$ UIt, the energy consumption is proportional to reaction time and current density. Increased distance between anode and cathode electrodes leads to increased voltage and then results in higher energy consumption[23].

Amount of sludge produced during electrochemical treatment is of environment relevance. It points to the problem of solid waste generation and disposal costs associated with the process. Then the production of sludge is the less the better. Fig. 4(a) shows that higher initial silica concentration and smaller interelectrode distance result in higher sludge production. This may due to that more formation of hydroxyaluminosilicates[24] as eqn. 6 generated under these conditions.

$$nAl^{3+} \xleftrightarrow{OH^-} [Al(OH)_3]_n \xleftrightarrow{Si(OH)_4} (AlO)_n(SiO)_{n/2}(OH)_{2n} \qquad (6)$$

Fig. 4(b) and (c) indicate that sludge production increases with the increase of pH, reaction time, current density and submerged height of electrode in linear speed, for more formation of hydroxyaluminosilicates which mean that more silica was removed under these conditions.

Optimization of responses: The Design-Expert software can be used to optimize the experimental results, on the basis of the operator's requirements to achieve different project purposes. The software can give different solutions when different ranges of variables or different goals of responses are set. Since each kind of target water has established silicon concentration, here initial silica concentration is targeted at 60 mg/L as a representative. The pH value of target water is generally neutral and is targeted at 7.5. Then the other variables are set in range. For the responses, silica removal efficiency is to be maximized, energy consumption is to be minimized, electrode consumption is to be minimized and sludge production is to be minimized. For the purpose of silica removal, the importance of silica removal efficiency is set at III and the other three are set at II. Then one of the optimized conditions under these specified limits is 60 mg/L of initial silica concentration, 2.50 cm of interelectrode distance, 40.47 min of reaction time, 7.50 of pH value, 3.89 mA/cm^2 of current density and 4 cm of submerged height of electrode. At the optimum condition, 85.147 % of silica removal efficiency, 1.455 KWh/m^3 of energy consumption, 0.0389 kg/m^3 of electrode consumption and 0.242 kg/m^3 of sludge production are foreseen by the program optimization and the desirability is 0.831. The verification of these results using the optimized conditions was accomplished by performing the experiments. And the actual results were obtained as follow: 83.243 % of silica removal efficiency, 1.466 KWh/m^3 of energy consumption, 0.0400 kg/m^3 of electrode consumption and 0.245 kg/m^3 of sludge production.

Conclusion

The present study clearly demonstrated the applicability of electrocoagulation process using alminium electrode for silica removal from cooling water. It was showed that RSM is one of the suitable methods to optimize the operating factors to maximize silica removal efficiency and minimize energy consumption, electrode consumption and sludge production. The experimental results showed that satisfactory empirical model equations were developed for these variables. The interactions of the variables were demonstrated and reaction time and current density were the two main factors affecting the four responses seriously. The optimum conditions were also foreseen by the process software, which provided a better selection for practical application. And these were verified by preforming the optimum conditions.

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