Multiparameter optimization of bromate sorption on anion exchange resin by a two-step statistical strategy: Plackett–Burman and Box–Behnken experimental design

Yu Zhong[a,b], Qi Yang[a,b,*], Xin Li[a,b], Hongbo Chen[a,b], Xiaoming Li[a,b,*], Ren Chen[a,b], Yang Liu[a,b], Guangming Zeng[a,b]

[a]*College of Environmental Science and Engineering, Hunan University, Changsha 410082, China, email: zhongyu@hnu.edu.cn (Y. Zhong), Tel./Fax: +86 731 88822829; emails: yangqi@hnu.edu.cn (Q. Yang), hgxlixin@hnu.edu.cn (X. Li), chenhongbo@hnu.edu.cn (H. Chen), xmli@hnu.edu.cn (X. Li), qxxu@hnu.edu.cn (R. Chen), yangqi1974@163.com (Y. Liu), zgming@hnu.edu.cn (G. Zeng)*
[b]*Key Laboratory of Environmental Biology and Pollution Control, Hunan University, Ministry of Education, Changsha 410082, China*

ABSTRACT

Bromate, an anion with carcinogenic properties, may be formed during ozonation or chlorination of water containing bromide. Previous research has shown that the trace levels of bromate in solution could be effectively removed by macroporous Cl-type anion exchange (D201-Cl) resin. In the present study, this multiparameter sorption process was optimized by a two-step statistical experiment design. Firstly, the Plackett–Burman (P–B) design was employed to evaluate the effect of sorption parameters. Sorbent dosage, coexisting anion concentration, and initial pH had a significant impact on bromate removal, and these variables were further optimized using a Box–Behnken design (BBD). The maximum removal efficiency of bromate (90.67%) was achieved under the optimal conditions of sorbent dosage 1.08 g/L, coexisting anion conc. 15.55 mg/L, and pH 8.93. The experimental results were in good agreement with the predicted values, indicating that the integrated P–B and BBD design is a powerful and effective approach for the statistical optimization of bromate sorption process.

Keywords: Bromate; Anion exchange resin; Plackett–Burman design; Box–Behnken design; Optimization

1. Introduction

Bromate is a disinfection byproduct generally formed during ozonation or chlorination of bromide-containing water [1,2]. Even very low level of bromate is harmful to aquatic life and human health due to its toxicological, potential carcinogenic, and neurological effects [3]. So bromate is classified as a Group 2B or "possible human" carcinogen by the International Agency for Research on Cancer (IARC) [4]. Research showed that bromate concentrations in pilot and full-scale drinking water were as high as 60 μg/L [5], which was much higher than the maximum contaminant level of bromate in drinking water of 10 μg/L

*Corresponding authors.

[3,6]. Therefore, it is urgent to develop an efficient and low-cost technology for bromate removal from drinking water.

In recent decades, several methods including chemical reduction, biological reduction, and adsorption were used to remove bromate [7–9]. Among them, ion exchange has been considered to be an economical alternative [10]. Compared with other adsorption materials, anion exchange resin has many advantages, such as commercial availability, convenient regeneration, and more environmental friendly. A highly selective anion exchange resin was used to remove low levels of perchlorate (ClO_4^-). Meanwhile, ClO_4^--loaded resins were regenerated effectively using tetrachloroferrate ($FeCl_4^-$) displacement technique [11]. Humbert and his co-workers found that strong anion exchange resin could quickly remove nitrate and sulfate from a high DOC content surface water [12]. In our previous study, macroporous Cl-type strong base anion exchange resin (D201-Cl) was found to exhibit an excellent bromate removal efficiency, and the maximum bromate adsorption capacity reached to 105.5mg/g at 298K [13]. Bromate sorption on anion exchange resin is strongly influenced by various variables, such as temperature, contact time, sorbent dosage, initial pH, and coexistent anions [13,14]. The comprehensive investigation of their impact is laborious and all-consuming. So, it is necessary to select the most significant variables from them and evaluate their possible interactions.

The Plackett–Burman (P–B) design, a two-level multifactor design based on the well-known balanced incomplete block rationale [15], is usually used to screen the important factors among a large number of variables. After determining the critical factors, it is necessary to further optimize the values of these process factors. Conventional single dimensional approach involves changing one independent variable at a time with the others at a constant level, which gives unreliable results and inaccurate conclusion [16]. Response surface methodology (RSM), including central composite design and Box–Behnken design (BBD), is an efficient strategic experimental technique to determine optimal conditions for a multivariable system [17,18]. To our knowledge, P–B and RSM design, the two-step statistical experiment design has been widely employed in the optimization of various processes, such as biochemistry [19], food chemistry [20],and water treatment [21]. However, there are few reports concerning the optimization of bromate sorption on anion exchange resin.

Therefore, the primary objective of this study is to select the significant parameters from eight process variables of bromate sorption using P–B design. Then,

the standard RSM design of BBD was employed to optimize the P–B design identified parameters regarding the bromate removal efficiency. Finally, the validity of the proposed model for bromate removal was verified.

2. Materials and methods

2.1. Materials and water samples

The macroporous anion exchange (D201-Cl) resin (Dayu Chemical Co. Ltd, Changsha, China) was used as sorbent in this study. D201-Cl is a macroporous and strong base anion exchange resin with polystyrene matrix, quaternary amine functional groups, and chloride. Prior to use, the resin was pretreated with hydrochloric acid and ethanol to remove impurities.

According to our previous studies [13], the impact of coexisting anions on bromate removal was in the following order: nitrate (NO_3^-) > sulfate (SO_4^{2-}) > chloridion (Cl^-). Therefore, nitrate was selected as the representative to study the effect of coexisting anions on bromate removal. Standard stock solutions of bromate and nitrate were prepared by dissolving analytical pure grade chemicals $NaBrO_3$ and $NaNO_3$ (Kermel Chemical Reagent Co. Ltd, Tianjin, China) in deionized water (DW), respectively. The stock solution for each inorganic salt was diluted with DW to obtain desired anion concentration.

2.2. Batch sorption experiments

Batch sorption experiments were performed in 250-ml Erlenmeyer flasks with 200 mL working volume on a thermostated rotary shaker, and temperature maintained at 25℃ or 50℃ according to the P–B design. The pH of solution was adjusted by 0.1 M HCl or NaOH and measured using a pH meter (pHS-3C model, Leici, China). The concentrations of bromate and nitrate were determined using a Dionex ion exchange chromatography (ICS-900, USA) consisting of a DS-5 electrochemical detector and AS19 analytical columns. The mobile phase (flow rate 1 mL/min) was a 9.4 mM solution of Na_2CO_3 and 1.8 mM solution of $NaHCO_3$. The bromate removal efficiency (Y) was calculated using the following equation:

$$Y = \frac{\text{initial conc. of bromate} - \text{final conc. of bromate}}{\text{initial conc. of bromate}}$$

(1)

Each sample was analyzed in a triplicate and the results were presented as average.

2.3. Experimental designs

2.3.1. Plackett–Burman design

The P–B design is an efficient way to determine the significant factors among a large number of variables [22]. In this study, a total of 12 runs P–B design were applied to evaluate 11 factors (including three dummy variables). Each variable was tested at low (−1) and high (+1) levels. Table 1 illustrates the variables and their corresponding levels used in the experimental design.

The P–B design was arranged by Design Expert software version 8.0 (STAT-EASE, Inc., USA) and the response values of bromate removal (Y) are shown in Table 2. The effect of individual variable on bromate removal was calculated as follows [23]:

$$E(X_i) = 2\left(\sum M_H - M_L\right)/N \tag{2}$$

where $E(X_i)$ is the effect of the variable (X_i); M_H and M_L represent bromate removal efficiency obtained at the high and low levels for each variable, respectively; N is the total number of trials ($N = 12$).

The standard error (SE) was estimated according to Eq. (3):

$$\text{SE} = \sqrt{\sum (E_d)^2/n} \tag{3}$$

in which, E_d represents the effect of dummy variables and n is the number of dummy variables. The significance (p-value) of each variable was determined via Studen's t-test.

2.3.2. Box–Behnken design

The optimal levels of the significant variables identified by P–B design and the interactions of these variables on bromate removal were analyzed by a standard RSM design called BBD. The three-level, three-factorial BBD method was employed to fit the polynomial model and the experimental errors were calculated based on the standard deviation of the center point with five runs [24]. The three independent variables are investigated at three different levels (−1, 0, +1), while the center (zero) levels of each variable in BBD were sorbent dosage 0.67 g/L, coexisting anion concentration 28 mg/L, and initial pH 7.75. The experimental levels of variable are listed in Table 3.

The data obtained from BBD method were fitted using a second-order polynomial equation Eq. (4):

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i \leq j}^{k} \sum_{j}^{k} \beta_{ij} X_i X_j + \cdots + e \tag{4}$$

where Y is the predicted response, X_i and X_j are both the independent variables. k is the number of variables and e is the residual error. β_0, β_i, β_{ii}, and β_{ij} are the constant coefficient, the linear coefficient, the quadratic coefficient, and the interaction coefficient, respectively.

The Design-Expert 7.1 was used for the regression analysis of the experimental data, and also for plotting

Table 1
Variables and their levels employed in P–B design for the determination of significant factors affecting bromate removal using D201-Cl resin

Variables	Symbol	Experimental values	
		−1 level	+1 level
Temperature (℃)	A	25	50
Sorbent dosage (g/L)	B	0.15	1.20
Contact time (min)	C	10	600
Oscillation frequency (r/min)	D	100	180
Particle size (mm)	E	0.5	1.0
Initial pH	F	2.5	13.0
Coexisting anion conc. (mg/L)	G	0	56
Initial bromate conc. (μg/L)	H	200	600
Dummy 1	I [a]	0	0
Dummy 2	J [a]	0	0
Dummy 3	K[a]	0	0

[a]The symbol of I, J, K represent three dummy variables.

Table 2
P–B design of variables in coded levels with bromate removal efficiency as response

Run	Coded variable level											Y (%)
	A	B	C	D	E	F	G	H	I	J	K	
1	−1	1	−1	1	1	−1	1	1	1	−1	−1	76.37
2	−1	−1	1	−1	1	1	−1	1	1	1	−1	28.44
3	1	1	−1	1	1	1	−1	−1	−1	1	−1	57.97
4	−1	1	1	1	−1	−1	−1	1	−1	1	1	37.07
5	1	1	−1	−1	−1	1	−1	1	1	−1	1	34.14
6	−1	−1	−1	1	−1	1	1	−1	1	1	1	59.67
7	1	−1	1	1	−1	1	1	1	−1	−1	−1	10.15
8	−1	1	1	−1	1	1	1	−1	−1	−1	1	97.04
9	1	−1	−1	−1	1	−1	1	1	−1	1	1	2.61
10	1	1	1	−1	−1	−1	1	−1	1	1	−1	35.67
11	1	−1	1	1	1	−1	−1	−1	1	−1	1	17.04
12	−1	−1	−1	−1	−1	−1	−1	−1	−1	−1	−1	12.81

Table 3
Experimental range and levels of the significant variables in BBD

Variable	Symbol	Range and levels		
		Low level (−1)	Center level (0)	High level (+1)
Sorbent dosage (g/L)	X_1	0.15	0.67	1.20
Coexisting anion conc. (mg/L)	X_2	0	28.00	56.00
Initial pH	X_3	2.50	7.75	13.00

the response surface graphs. The significance of variables is determined using analysis of variance (ANOVA) and judged statistically by computing the F-value at a probability (p-value) of 0.001, 0.01, or 0.05; details of this procedure can be found elsewhere [25,26]. The multiple coefficients of correlation (R^2) and the adjusted determination coefficient (R^2_{adj}) were calculated to assess the validity of the model.

3. Results and discussion

3.1. Screening of significant variables using Plackett–Burman design

As shown in Table 2, the bromate removal efficiency fluctuated from 8.5 to 97.7% in 12 runs, suggesting that process optimization is important to improve the removal efficiency. The variable evidencing statistically significant effects were screened via Student's t-test for ANOVA and the results are listed in Table 4. The variable with confidence level above 95% (p-value < 0.05) is considered as a significant parameter [27]. The sorbent dosage has obvious impact on the sorption process since its value determines the amount of sorbent surface area and active sites [6,13]. Therefore, sorbent dosage with probability value of 0.0054 was found to be the most influencing variable, followed by initial pH (0.0351) and coexisting anion concentration (0.0472). Similar to our results, Ding and his co-workers also observed that adsorbent dosage, initial pH of solution, and coexistent anions had significant effect on the phosphate removal by resin [14].

3.2. Optimization of significant variables using BBD

After sorbent dosage, coexisting anion concentration and initial pH being determined to be the significant variables based on the P–B design, the optimum levels of these variables were further optimized by BBD. The design matrix, the corresponding results of BBD, and the predicted values are shown in Table 5. From Table 5, it can be found that the removal efficiency of bromate ranged from 16.12 to 90.12%. According to the multiple regression analysis on

Table 4
Effects of the variables and statistical analysis of the P–B design

Factor	Effect	Coefficient	Standard error	t-value	p-value
A-temperature	−6.79	−7.07	2.40	−2.83	0.0529
B-sorbent dosage	17.29	17.30	2.40	7.21	0.0054[a]
C-contact time	−1.07	−1.52	2.40	−0.45	0.5728
D-oscillation frequency	4.41	3.97	2.40	1.84	0.1974
E-particle size	7.48	7.49	2.40	3.12	0.0531
F-initial pH	9.26	8.82	2.40	3.86	0.0351[a]
G-coexisting anion conc.	8.27	7.83	2.40	3.45	0.0472[a]
H-Initial bromate conc.	−7.17	−7.61	2.40	−2.98	0.0511

[a]p-values < 0.05 indicate model terms and variables are significant.

Table 5
Experimental design and results of BBD

Run	Coded levels			Response Y (%)	
	X_1	X_2	X_3	Actual	Predicted
1	−1	0	−1	16.12	13.68
2	−1	−1	0	60.55	60.64
3	0	+1	−1	21.32	15.31
4	−1	+1	0	21.78	29.95
5	0	−1	+1	67.18	73.19
6	0	0	0	81.81	81.41
7	+1	−1	0	90.12	85.45
8	0	0	0	81.87	81.41
9	0	−1	−1	41.46	43.93
10	+1	0	+1	71.03	73.30
11	+1	+1	0	61.27	61.58
12	0	+1	+1	50.05	47.58
13	0	0	0	78.69	81.41
14	0	0	0	83.99	81.41
15	+1	0	−1	49.27	55.26
16	0	0	0	80.67	81.41
17	−1	0	+1	64.26	58.44

experimental data, the following quadratic model was established as follows:

$$Y_1 = -32.445 + 80.426X_1 + 0.365X_2 + 17.316X_3$$
$$+ 0.116X_1X_2 - 2.423X_1X_3 - 5.119E - 003X_2X_3 \quad (5)$$
$$- 28.163X_1^2 - 0.017X_2^2 - 0.828X_3^2$$

The adequacy of the model was checked using ANOVA, as shown in Table 6. The model F-value of 19.84 and Prob. > F of 0.0004 revealed that the model was significant as well. Furthermore, the F-value (19.84) was found to be greater than the tabulated F-value ($F_{\alpha(\mathrm{df}, n-\mathrm{df}+1)} = F_{0.05,9,7} = 3.68$) at 5% level, indicating that the computed Fisher's variance ratio at

this level is large enough to justify a appropriate of the model, as similarly reported by Yetilmezsoy and his co-workers [26]. The values of Prob. > F less than 0.05 indicated that the model terms were significant. In this case, the linear terms of X_1 (sorbent dosage), X_2 (coexisting anion conc.), X_3 (initial pH), and quadratic terms of X_2^2, X_3^2 were significant for bromate removal. A higher value of the model correlation coefficient ($R^2 = 0.9774$) means that 97.74% of the total variation on bromate removal data could be explained by the selected model. Simultaneously, a relatively low value of the coefficient of variation (C.V. = 9.76%) indicated good precision and reliability of the experiments.

Diagnostic plots of the predicted values versus the actual values can also be used to judge the suitability of the model [28]. Fig. 1 shows the relationship between the actual and predicted values of bromate removal efficiency. It could be seen that the observed points were distributed relatively near to the straight line with the higher correlation coefficient (R^2) 0.966, indicating that the regression model is able to predict these removal efficiencies.

3.3. Analysis of response surface

Three dimensional (3D) response surface plots as a function of two variables, maintaining all other variables at fixed levels, are more helpful in understanding both the main and the interaction effects of these two variables [26]. Three 3D response surface plots for the measured responses and three corresponding contour plots were formed based on the model equation (Eq. (5)) and showed in Fig. 2.

Fig. 2(a) shows the effect of sorbent dosage and coexisting anion concentration on bromate removal at a fixed initial pH 7.75. More than 80% removal efficiency was achieved when the dosage was greater than 0.6 g/L. Similar to the most of the sorbents, higher

Table 6
ANOVA of the response surface quadratic model for bromate removal

Source	Sum of squares	Degree of freedom	Mean sum of squares	F-value	p-value (Prob. > F)
Model	8,774.64	9	974.96	21.29	0.0004
X_1	1,592.74	1	1,592.74	34.77	0.0006
X_2	1,486.58	1	1,486.58	32.46	0.0007
X_3	1,970.39	1	1,970.39	43.02	0.0003
X_1X_2	11.67	1	11.67	0.25	0.6283
X_1X_3	178.66	1	178.66	3.94	0.0601
X_2X_3	2.27	1	2.27	0.049	0.8304
X_1^2	252.26	1	252.26	5.51	0.0529
X_2^2	776.98	1	776.98	16.96	0.0045
X_3^2	2,192.50	1	2,192.50	47.87	0.0002
Residual	320.61	7	45.80	–	–
Lack of fit	305.63	3	101.88	19.84	0.0041
Pure error	14.91	4	2.64	–	–

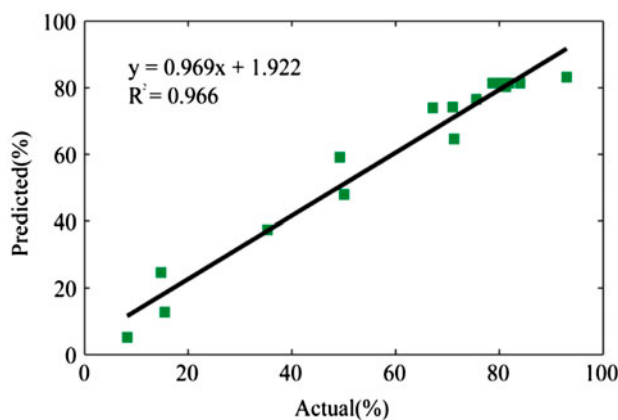Notes: S.D. = 6.05, C.V.% = 9.76, R^2 = 0.9774, R^2_{adj} = 0.9483.

Fig. 1. Plot of actual and predicted bromate removal efficiency.

dosage of D201-Cl resin increased the number of active sites and overall surface area of the sorbent particles for bromate removal.

The effect of interaction between sorbent dosage and initial pH at the center level of coexisting anion concentration can be inferred from Fig. 2(b). The effect of sorbent dosage on bromate removal was the same as that in Fig. 2(a). The bromate removal efficiency increased with the increase in initial pH value from 4.0 to 8.9, but decreased when the initial pH value continuously increased from 8.9 to 12.0. In the low pH medium (pH < 4), bromate was protonated to molecules form with H^+, which weaken the ion exchange between bromate and resin. In the higher pH medium (pH > 9), the high concentration of OH^- was present in the solution, which competed with bromate for the adsorption active site on the resin. The sorbent dosage impacts the surface area of adsorption process and the pH is responsible for surface charge, therefore their combined effect is significant in the combined variables (p-value = 0.0601).

The combined effect of coexisting anion concentration and initial pH on bromate removal is displayed in Fig. 2(c). As seen from Fig. 2(c), lower coexisting anion concentration and alkaline condition were more beneficial for bromate removal by D201-Cl resin. Only when the coexisting anion concentration was beyond 30 mg/L, it will bring negative effect on bromate removal.

3.4. Confirmation results

From the proposed model Eq. (5), the results of mathematical calculations indicated that the optimal coded value of the three most significant variables were $X_1 = 0.77$, $X_2 = 0.45$, and $X_3 = 0.25$. Correspondingly, the values of sorbent dosage, coexisting anion concentration, and initial pH were 1.08 g/L, 15.55 mg/L, and 8.93, respectively. The maximal bromate removal efficiency was predicted to be 91.11% under optimal condition. In order to confirm the optimized experimental condition, three additional experiments were performed using the predicted conditions and the average results are shown in Table 7. The average of obtained experimental value was 90.67%, which agreed well with the predicted value. These results demonstrated that the statistical model is effective and reliable.
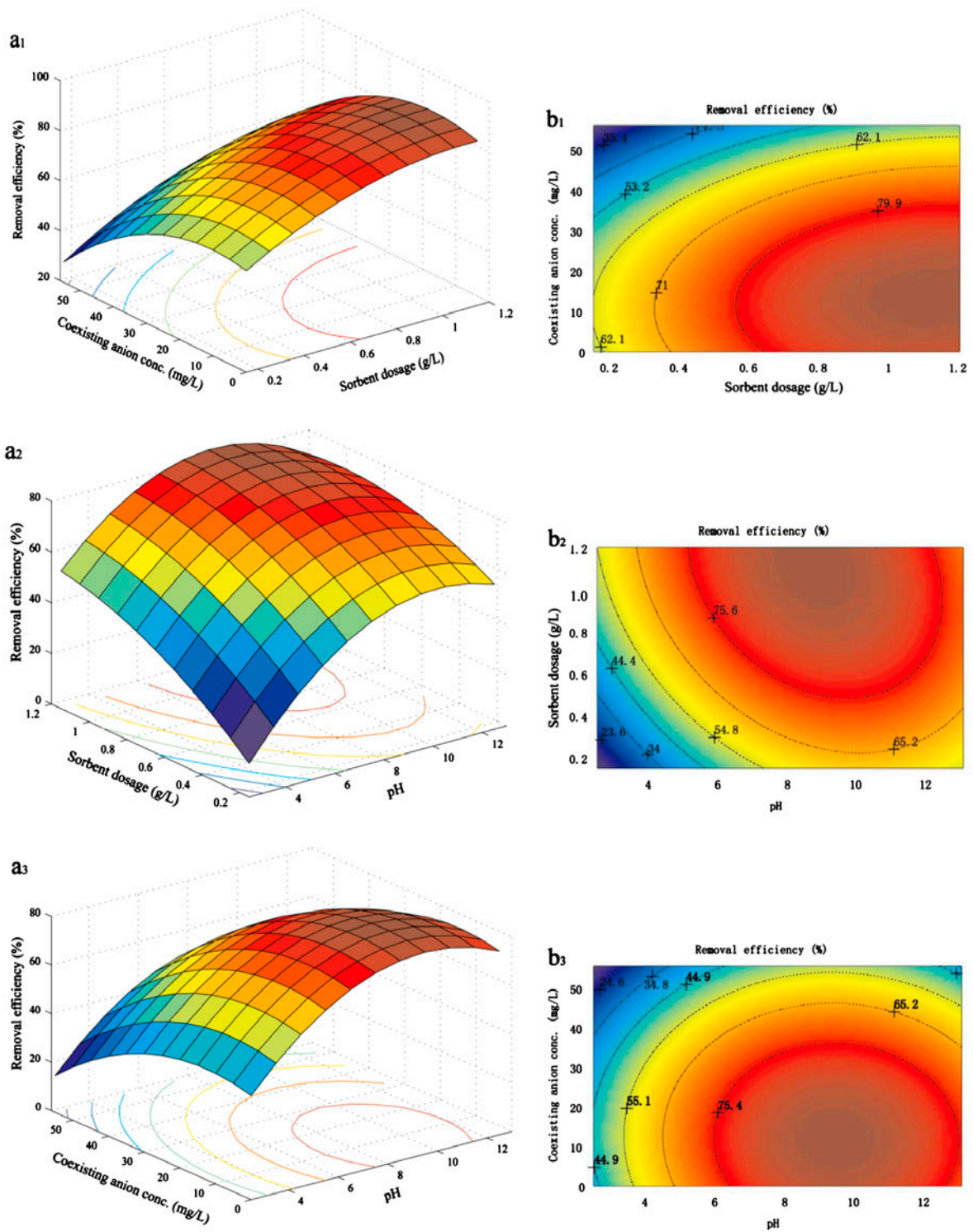
Fig. 2. Three-dimensional response surface plots and two-dimensional contour plots for bromate removal by D201-Cl resin showing variable interactions of: (a$_1$ and b$_1$) sorbent dosage and coexisting anion conc.; (a$_2$ and b$_2$) sorbent dosage and pH; (a$_3$ and b$_3$) coexisting anion conc. and pH.

Table 7
Predicted and experimental values of response at optimum conditions

Sorbent dosage (g/L)	Coexisting anion conc. (mg/L)	pH	Predicted (%)	Experimental (%)
1.08	15.55	8.93	91.11	90.67 ± 0.8

4. Conclusions

In this study, a two-step statistical experiment design, P–B design, and BBD method was used to optimize process variables for bromate removal using macroporous Cl-type anion exchange resin. Three variables (sorbent dosage, coexisting anion concentration, and initial pH) were screened among a large number of process parameters as the significant variables by P–B design. The effect and interaction of the three variables were further optimized using BBD method. The maximal bromate removal efficiency (91.11%) predicted under optimal conditions of sorbent dosage 1.08 g/L, coexisting anion concentration 15.55 mg/L, and initial pH 8.93, was close to the experimental value (90.67%). Results suggested that the P–B design followed by BBD method is an effective and powerful approach for the optimization of bromate sorption process.

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