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Research

TREATABILITY STUDY OF CHEMICAL DEFLUORIDATION TECHNIQUE BY BORIC ACID: BATCH ANALYSIS AND OPTIMIZATION USING RESPONSE SURFACE METHODOLOGY

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Key words: Fluoride removal, Boric acid, Response surface methodology, Experimental design, Desirability function

ABSTRACT

This study is highlighted on evaluation of the feasibility of chemical de-fluoridation technique using boric acid. The remediation of fluoride containing solution was performed using boric acid in acidic medium. Batch continuous process was carried out using four process parameters such as dosage of boric acid (g), volume of acid (ml), reaction time (min) and reaction temperature (K). Central composite design (CCD) with the help of design expert software is applied to study the interactions of different process variables responsible for fluoride removal. Desirability function was achieved using numerical optimization which was further used to identify the optimum conditions. A confirmatory experiment was performed to evaluate the accuracy of the optimized procedure. By statistical approach, the optimized condition was achieved such as dose of boric acid 3 g/L; volume of phosphoric acid 12 ml; temperature 333 K; contact time 55 minutes. Under the optimized condition, fluoride removal efficiency was obtained as 97.3% and experimentally in batch process it is obtained as 97.8%. So, it can be concluded that boric acid can be chemically used for de-fluoridation study in waste water as it is economically and environmentally safe.

INTRODUCTION

Fluoride is the most common constituent of natural water samples. Its concentration varies depending on the water source. The major contribution of fluoride comes from geological resources though both geological and man-made sources (Hem, 1959; Abu and Alsokhny, 2004).

Surface waters seldom have fluoride levels exceeding 0.3 mg/l. There are several fluoride bearing minerals in the earth's crust. They occur in sedimentary (limestone and sandstone) and igneous (granite) rocks. The major contribution of higher fluoride levels in ground water comes from weathering of these minerals along with volcanic and fumarolic processes (Zeni, *et al.*, 2005). Dissolution of these barely soluble minerals is dependent on the water composition and the time of contact between the source minerals and the water. For many years ground water, has been considered to be a protected and safe source of water, fit for drinking without treatment. There is little consideration which is used to be given to the risks of chemical pollution, mainly to the presence of elevated levels of fluoride, arsenic and nitrate in groundwater. Fluorosis (Tamer, *et al.*, 2007) is caused by consumption of water having excess fluoride over a prolonged period. It has been reported that 23 nations around the globe have been affected by high-fluoride contaminated groundwater. It has led to endemic fluorosis, which has become a major geo-environmental health issue in many developing countries. The incidence of low dental caries demonstrate that fluoride concentrations of up to 1.0 mg/l in potable water are beneficial to the oral health of children. In several developed countries fluoridation of water supplies is used if the natural concentration is below the desired level. Skeletal fluorosis affects both adults and children. It is generally occurred after consumption of water with fluoride levels exceeding 3 mg/l. Typical symptoms of skeletal fluorosis are pain in the joints and backbone. After the result of recent research

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studies, it is shown that excess intake of fluoride can affect different non-skeletal health impacts such as gastro-intestinal problems, allergies, anemia and urinary tract problems.

According to the World Health Organization (WHO, 2006), the maximum permissible level of fluoride in drinking waters is 1.5 mg/l. As the fluoride intake determines health effects, standards are bound to be different for countries with temperate climates and for tropical countries, where significantly more water is consumed. Although water is generally the major route of fluoride intake, exposure from diet and air may become important in some situations. However, in many cases, the required data on different routes of exposure may be lacking. Data obtained by monitoring fluoride levels in local water supplies and the incidence of fluorosis in the local population can be used to arrive at the appropriate standards.

The main objective of the present study is to determine the optimum conditions for de-fluoridation by chemical reaction in aqueous system. Usually for de-fluoridation in water adsorption, precipitation, electro-dialysis, ion-exchange, reverse osmosis various methods are utilized (Antoniou, *et al.*, 2014). But in this study, chemo de-fluoridation technique is used which is not very common method at all. Batch study and statistical approach (Block and Rajagopalan, 2009) are represented here to confirm about optimized condition (Banihabib, *et al.*, 2012) of de-fluoridation. Response surface methodology (Design Expert Software) is utilized for statistical optimization for de-fluoridation in water.

A response surface methodology (RSM) is a set of design of experiments (DOE) which helps to optimize the response. This methodology is used to refine models after determining important factors using factorial designs. Response surface methods mainly involves 3 steps: (1) Calculating the coefficients by statistically conducting experiments. (2) To achieve the best operating conditions from maximum or minimum response. (3) Number of responses to be optimized at the same time and desirability function also used in the final step.

This investigation deals with boric acid in acidic medium, used as chemo de-fluoridating agent in batch and statistical method which is described vividly.

MATERIALS AND METHODS

Preparation of synthetic fluoride solution

Sodium fluoride (Merck, Germany) is used in this study. Stock fluoride ion solution (1000 mg L^{-1}) is prepared by dissolving accurately weighed quantity in double-distilled water. 50 mgL^{-1} fluoride solution was prepared by diluting stock fluoride solution, used in the experimental study.

EXPERIMENTAL SETUP

Batch Chemical Studies

For batch experiments 100 ml fluoride solutions of concentration 50 mgL^{-1} were taken in 250 mL PTFE(Polytetrafluoroethylene) conical flasks. The weighed amount of boric acid was added to each solution. Then the flasks were agitated at 150 rpm in an temperature controlled incubator shaker (INNOVA 4430, New Brunswick Scientific, Canada) at different temperatures. After shaking for particular time intervals samples were collected from the flasks for analysis of fluoride concentration in the solution. The residual amount of fluoride in each conical flask was estimated by using ion-meter (Thermo Scientific Orion ion-meter, USA). The effect of boric acid dose (g), volume of acid (ml), reaction time (min) and reaction temperature(K) were evaluated in batch study.

Response surface methodology for optimization of process parameters using central composite design(CCD)

RSM is an empirical statistical technique employed for multiple regression analysis by using quantitative data obtained from properly designed experiments to solve multivariate equations

In this work, RSM (Moravejalahkami, *et al.*, 2009) is used to assign the relationship between the response and the independent variables, as well as to optimize the relevant variables in order to predict the best value for the response. The central composite rotatable design (CCD) is the most widely used RSM approach,

and CCD has been employed to determine the effect of operational variables [boric acid dose(g), volume of acid(ml), reaction time (min) and reaction temperature(K)] on the de-fluoridation efficiency of boric acid in acidic medium (Owen and King, 1943).

The determination of optimum conditions for de-fluoridation by boric acid depends on the four process variables, which are dose of boric acid, volume of acid, reaction temperature and reaction time. The experimental ranges along with the levels of variables are given in Table 1. The percent removal of fluoride is the response of the system. Statistically the prediction of the optimum condition is obtained following the quadratic equation model given below (Eq. 1).

Table 1. Experimental range and levels of independent variables

Serial no	Variable	Unit	Notation	Range and levels(coded)				
				$-\alpha$	-1	0	+1	$+\alpha$
(1)	Amount of boric acid	g/L	A	0.3	1.2	2.1	3.0	3.9
(2)	Volume of acid	ml	B	6	8	10	12	14
(3)	Temperature	K	C	295.5	308	320.5	333	345.5
(4)	Reaction time	min	D	2.5	20	37.5	55	72.5

$$Y = \beta_0+\sum^{k}_{i=1} \beta ixi + \sum^{k}_{i=1} \beta iixi2 +\sum_{i=1}\sum_{j=i+1} \beta ijxixj+ \varepsilon \quad (1)$$

Here, Y=response (i.e. dependent variable), $\beta 0$=constant coefficient, $\beta i=\beta ii=\beta ij$=coefficients of linear, quadratic and interaction effect, xi and xj=factors (independent variables)

and ε =error.

Percentage of fluoride removal efficiency is calculated with a standard RSM design (CCD) and 30 experiments are performed. The percent removal (%) of fluoride is determined by using the following equation (2):

$$R(\%) =(Ci-C0)/Cix100 \quad (2)$$

Where Ci is the initial fluoride concentration (mg L^{-1}) and C0 is the final fluoride concentration in solution (mg L^{-1}).

Design Expert Version 7.1.6 (Stat Ease, USA) was utilized for regression and graphical analysis of the experimentally obtained data. The optimum values of the independent variables were obtained by solving the regression equation and by analyzing the response surface contour plots (Hamsaveni, *et al.*, 2001). The variability in response was statistically analyzed using multiple coefficient of determination. The correlation of coefficient and the quadratic model equation was used to predict the optimum value from which the interaction effect of different factors within the specified range may be described.

Determination of fluoride (F^-) concentration

The fluoride (F^-) concentration is determined using ion-meter (Thermo Scientific Orion ion-meter, USA). According to Standard Methods 10 ml sample is placed in 100 ml beaker and 1 ml of TISAB III solution is added the beaker to maintain pH 5-5.5 and to eliminate the interference effect of complex ions. The total volume should be sufficient to immerse the electrode and permit the use of the stirrer. Fluoride concentrations of the samples are taken directly from the ion-meter.

RESULTS AND DISCUSSION

Batch studies

Effect of reaction time: It was observed from Fig. 1 the experimental results that with increasing reaction time at particular temperature, de-fluoridation efficiency increased. As the reaction time increased from 5 min to 60 min, higher number of fluoride ions reacted with boric acid molecules. The reason behind the phenomenon is that higher the number of fluoride ions react with the boric acid molecules, increasing the number of fluoroborate ions (insoluble) formed, which can be replaced in the acidic medium, that may increase the percent removal of fluoride in solution.

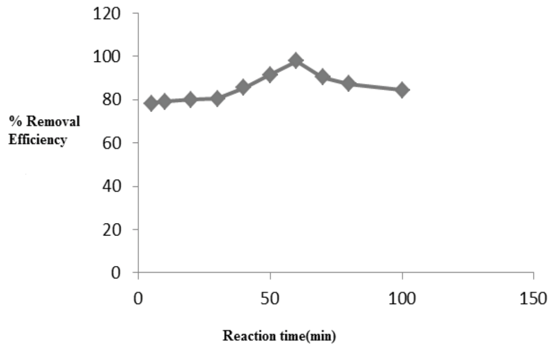

Fig. 1 Effect of reaction time on chemo de-fluoridation.

Effect of boric acid dose: Within the experimental range of dose of boric acid in between 1 g/L to 4 g/L, percent removal of fluoride firstly increased, then decreased. The dose of boric acid within the range of 1 g/L to 3 g/L, de-fluoridation efficiency increased because more the number of fluoride ions reacted with the boric acid molecules, increasing the number of fluoroborate ions (insoluble) which can be removed in the acidic medium. When the boric acid dose increased higher than 3 g/L, then removal decreased due to the saturation of boric acid by fluoride ions, and so increasing the number of fluoride ions (soluble) along with the fluoroborate ions (insoluble). As a result, de-fluoridation efficiency decreased (Fig. 2).

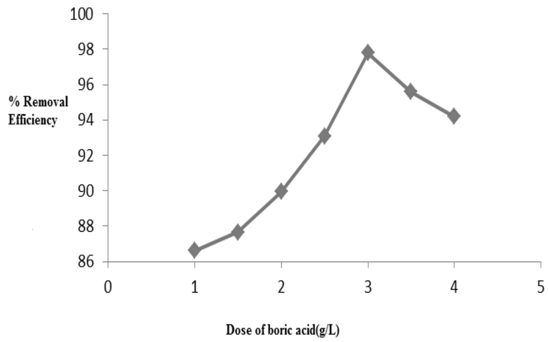

Fig. 2 Effect of boric acid dose on chemo de-fluoridation.

The chemical reaction of the phenomena may be: Sodium fluroride (NaF) is used in this experiment. To remove the undesirable insoluble fluoride deposits boric acid and hydrochloric acid can be used. But due to the poor solubility of boric acid in hydrochloric acid, the mixture of boric acid and hydrochloric acid is not particularly effective in dissolving the unwanted insoluble fluoride compounds. The reaction is occurred as follows:

$$H_3PO_4 + NaF + H_3BO_3 \rightarrow BF_4^- + Na_3PO_4 + H^+ + H_2O$$

Boron and Fluoride form complexes as fluoborate ions (BF^-_4). The BF^-_4 ion hydrolyzes rapidly to $BF_3(OH)^-$ and then to $BF_2(OH)_2^-$, and then very slowly after this. As most of these fluoborate solutions can be replaced in acidic medium (Huang and Jackson, 1996), so in this way fluoride treatment can be possible.

Effect of volume of phosphoric acid: It was observed (Fig. 3) from the experimental results that with increasing the volume of nitric acid at particular temperature, de-fluoridation efficiency increased. As the volume increased, more the number of H+ ions reacted with fluoride ions and boric acid molecules. As a consequence, increasing the number of fluoroborate ions. The feasibility of de-fluoridation (Lounici, *et al.*, 1997) process in acidic medium enhanced as the number of H^+ increases. So, de-fluoridation efficiency increased.

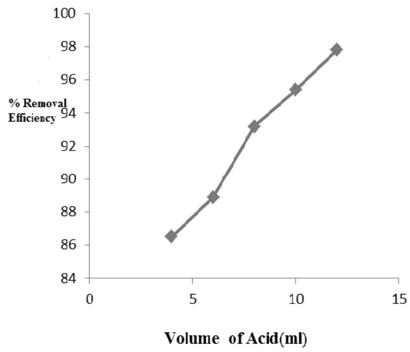

Fig. 3 Effect of volume of acid on chemo de-fluoridation.

Effect of temperature: It was illustrated in Fig. 4 that with increasing temperature, the percent removal of fluoride first increased from 308 K to 333 K. After 333 K, defluoridation efficiency (Jackson, 1996; Meenakshi and HazardMater, 2006; Mohapatra, *et al.*, 2009) decreased. As temperature increased, the higher number of fluoride ions reacted with boric acid molecules. But as temperature increased beyond 333 K, as higher number of fluoride ions (soluble) along with the fluoroborate ion s(insoluble) present in the reaction system due to higher activation energy (according to the Arrhenius Equation), as a result, the de-fluoridation efficiency firstly increased, then decreased.

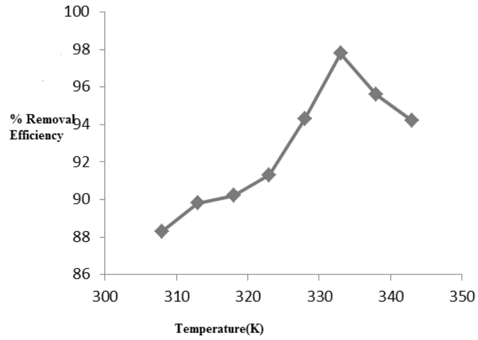

Fig. 4 Effect of reaction temperature on chemo de-fluoridation.

Estimation of response surface for maximum fluoride removal: The results of the 30 experiments performed as per CCD analysis were in Table 2. A maximum fluoride removal was obtained 97.3% at 333 K, 55 min reaction time, and 3 g/L 100 ml of boric acid and 12 ml phosphoric acid.

In this case response surface quadratic model showed that the interaction effects of dose of boric acid, reaction time and temperature. In order to study the interaction among the different independent variables and their corresponding effect on the response, contour plots were drawn (Figs. 5-11). A contour response variable which is expressed as a

Table 2. Central composite design (CCD) used for optimizing four independent variables along with the obtained response

Serial No.	A: Boric acid (g/L)	B: Volume of phosphoric acid (ml)	C: Temperature (K)	D: Reaction time (min)	Percent removal (%) of fluoride (R1)
1	**3.00**	**12.00**	**333**	**55.00**	**97.3**
2	3.00	12.00	333	20.00	90.3
3	3.00	8.00	308	20.00	83.4
4	2.10	10.00	320.5	37.50	77.6
5	2.10	10.00	320.5	37.50	77.6
6	1.20	12.00	308	20.00	70.3
7	3.00	8.00	333	55.00	93.2
8	2.10	10.00	320.5	2.50	73.4
9	2.10	6.00	320.5	37.50	83.3
10	2.10	6.00	320.5	37.50	83.3

11	1.20	8.00	308	20.00	83.2
12	3.00	12.00	308	20.00	85.6
13	2.10	10.00	295.5	37.50	78.9
14	0.30	10.00	295.5	37.50	70.3
15	1.20	12.00	333	20.00	72.3
16	3.00	8.00	308	55.00	75.4
17	3.00	12.00	308	55.00	73.2
18	2.10	10.00	345.5	37.50	85.5
19	2.10	10.00	320.5	37.50	77.6
20	2.10	10.00	320.5	37.50	77.6
21	1.20	8.00	308	55.00	85.4
22	3.00	8.00	333	20.00	89.8
23	1.20	12.00	333	55.00	82.2
24	2.10	14.00	320.5	37.50	88.3
25	1.20	8.00	333	55.00	94.6
26	3.90	10.00	320.5	37.50	77.6
27	1.20	8.00	333	20.00	72.3
28	2.10	10.00	320.5	72.50	95.2
29	2.10	10.00	320.5	37.50	77.6
30	1.20	12.00	308	20.00	70.3

function of independent variables defined in multiple regression model, developed by the software was expressed in the form of different numerical factors in equation (3) given below:

% Removal (R1)

$$+77.92 + 2.82 * A - 0.89 * B + 3.08 * C + 3.02 * D + 2.53 * A * B + 2.83 * A * C - 3.06 * A * D + 0.98 * B * C - 0.68 * B * D + 3.52 * C * D - 0.80 * A2 + 2.16 * B2 + 1.26 * C2 + 1.79 * D2 \quad (3)$$

In this case A, C, D, AB, AC, AD, CD, B2, D2 are significant model terms (described in Table 1). In presence of many insignificant model terms, model reduction was required which can improve the model. The goodness of fit of the model is verified by the correlation coefficient (R2) between the experimental and model predicted values of the response variable (Fig. 5). Statistically R2 value of 0.9021 indicated that the model was statistically significant. The predicted correlation coefficient (pred. R2 0.8907) also showed good agreement with the adjusted correlation coefficient. Overall, the applicability of the model was used to predict the percentage removal of fluoride in solution within the limits of the experimental factors

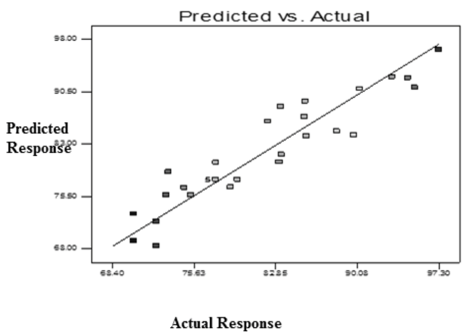

Fig. 5 The graph showing predicted removal % Vs actual removal % of fluoride.

Interaction Effect of Process Variables

Effect of variation in volume of acid and boric acid dose: The contour plot of Fig. 6 demonstrated the interaction effects of the independent variables (volume of phosphoric acid and boric acid dose). According to the contour plot of Fig. 6, de-fluoridation efficiency increased as volume of nitric acid and dose of boric acid increased. The maximal removal efficiency of 97.3% was achieved using 12 ml of acid and dose of boric acid 3 g/L, while the other process variables were constant at the middle value. Due to increase of dose of boric acid in acidic medium higher number of fluoroborate ions are produced, which are insoluble, in this way fluoride removal can be obtained by filtration.

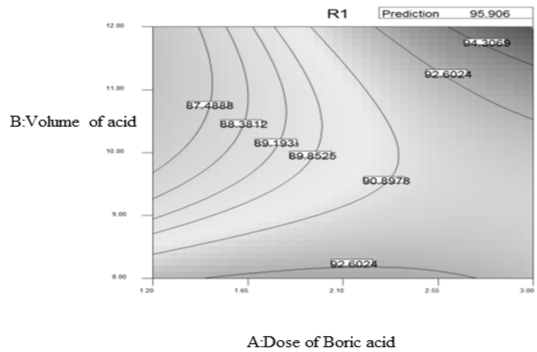

Fig. 6 Contour plot showing interaction effect of volume of acid and boric acid dose.

Effect of variation in temperature and boric acid dose: The effect of different temperature and dose of boric acid were required to remediate fluoride in solution. It was observed from Fig. 7 that both the independent process variables are the significant factors on the % removal of fluoride. As the temperature increased in between 303 K to 333 K, the fluoride uptake capacity increased at particular reaction time which indicated that at higher temperature with higher boric acid dose, due to number of fluoride ions reacted with the boric acid molecules increases which resulted higher number of fluoroborate ions (insoluble) formation, as a consequence % removal of fluoride increased.

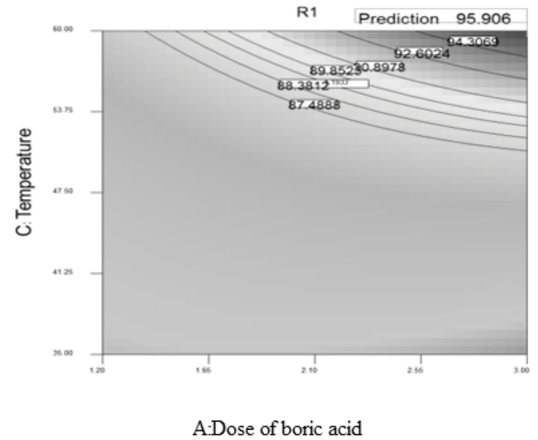

Fig. 7 Contour plot showing interaction effect of volume of acid and boric acid dose.

Effect of variation in volume of acid and temperature: The combined effect of volume of acid and temperature for de-fluoridation was depicted in the contour plot of Fig. 8. It was observed that percentage of fluoride removal increased with increasing the temperature and volume of acid. So, it was inferred that higher values of fluoride removal may be obtained by increasing temperature and volume of acid. As temperature increased, % removal of fluoride increased which supported the formation of fluoroborate ion (insoluble) as a result of chemical reaction between fluoride ion and boric acid molecules. As the volume of acid increased, increasing the number of H^+ ions, resulting in more number of fluoroborate ion formation. From this contour plot, a maximal removal efficiency of 97.3% is achieved by using 12 ml acid, at 333 K and 55 min of reaction time.

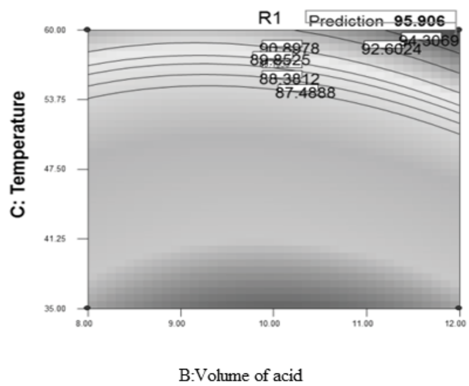

Fig. 8 Contour plot showing interaction effect of temperature and volume of acid.

Effect of variation in reaction time and boric acid dose: The effect of different reaction time and dose of boric acid were required in de-fluoridation technique using boric acid. It was observed from the contour plot (Fig. 9) that both the independent process variables play important role on the % fluoride remediation process. From this contour plot, a maximal removal efficiency of 97.3% was obtained at 55 min reaction time and 3 g/L of boric acid, while the other variable is set at the middle value. As the reaction time increased the fluoride uptake capacity increased at particular temperature, which indicated that at higher reaction time with higher boric acid dose, due to higher rate of chemical reaction of fluoride ions with the boric acid surfaces which results chemical de-fluoridation and as a result % removal efficiency

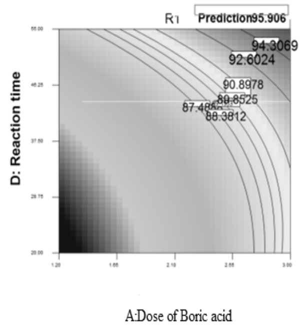

Fig. 9 Contour plot showing interaction effect of reaction time and boric acid dose.

Effect of variation in volume of acid and reaction time: It was observed from Fig. 10 that both the independent process variables play significant role in % fluoride removal process. From this contour plot, a maximal removal efficiency of 97.3% is obtained at 55 min reaction time and 12 ml acid. As the reaction time increased within the experimental limit, the fluoride uptake capacity increased at particular reaction temperature which indicated that at higher volume of nitric acid with higher reaction time, increasing the number of fluoride ions reacted with the boric acid molecules which resulted chemical de-fluoridation and as a result % removal efficiency increased.

Effect of variation in reaction temperature and reaction time: The contour plot of Fig. 11 demonstrated the interaction effects of the independent variables (reaction temperature and reaction time) in the above response. According to the contour plot of Fig. 11, de-fluoridation efficiency increased as temperature (higher activation energy) and reaction time increased. The optimum point is achieved as (333 K, 55 min). At that particular point the solution is saturated by fluoroborate ions (insoluble), beyond which the number of fluoroborate ions are decreased, which results in lower removal efficiency. From this study, the maximal removal efficiency of 97.3% was achieved at 333 K and volume of concentrated nitric acid 12 ml, while the other process variables were constant.

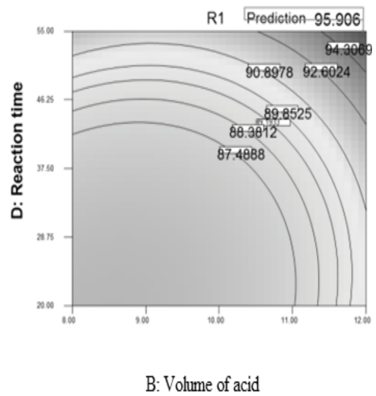

Fig. 10 Contour plot showing interaction effect of reaction time and volume of acid.

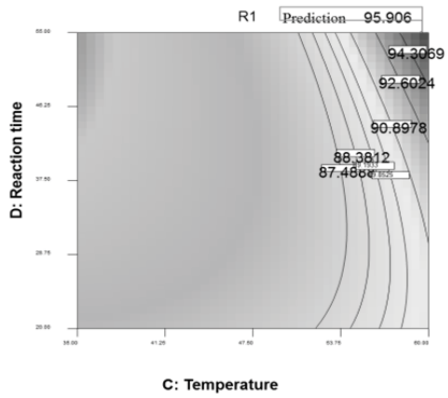

Fig. 11 Contour plot showing interaction effect of reaction time and reaction temperature.

Confirmational study: To support the optimized data given by numerical modeling (Fig. 12) confirmatory experiments were conducted with the parameters as suggested by the model (Dose of boric acid 3 g/L; volume of nitric acid 12 ml; temperature 333 K; contact time 55 minutes). The corresponding removal efficiency in optimum conditions is found 97.3% and experimentally it is 97.8% and the error observed is 0.005% which is very low. Here error is estimated by conducting experiment triplicate.

CONCLUSION

The present study was performed with the aim of de-fluoridation process using boric acid and to investigate the influence of various process parameters on fluoride removal using response surface methodology (Jain M, *et al.*, 2011). The fluoride removal efficiency was significantly affected by the dose of boric acid, volume of nitric acid, reaction time and temperature. By applying desirability function maximum fluoride removal efficiency was obtained in optimum conditions. The level of the four variables, dose of boric acid 3 g/L, reaction time 55 min, volume of nitric acid 12 ml, temperature 333 K were found to be optimum for maximum fluoride removal. The corresponding removal efficiency in optimum conditions was found to be 97.3%. As boric acid is low cost effective, so this process may be an effective material for the treatment (Ahmad, 2001) of wastewater treatment plant.

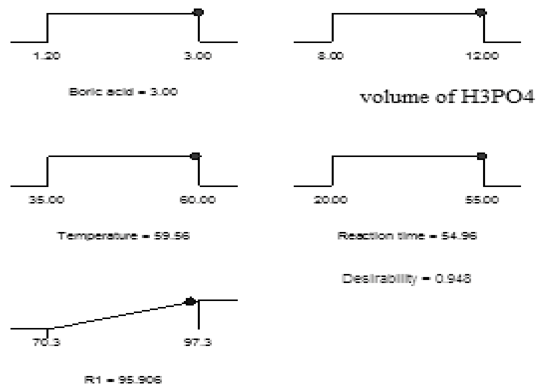

Fig. 12 Desirability function.

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