Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.937765

56 (2015) 736–743
October

Arsenic removal from aqueous solutions by ultrafiltration assisted with polyacrylamide: an application of response surface methodology

Bekir Varol[a], Nigmet Uzal[b,*]

[a]*Ahiler Development Agency, Atatürk Bulv, No. 57, Nevsehir, Turkey, Tel. +90 384 214 36 66; email: bvarol@yahoo.com*
[b]*Faculty of Engineering and Natural Sciences, Department of Environmental Engineering, Abdullah Gul University, 38380 Kayseri, Turkey, Tel. +90 352 2248800/3019; Fax: +90 352 2242483; email: nigmet.uzal@agu.edu.tr*

Received 14 February 2014; Accepted 16 June 2014

ABSTRACT

The present work deals with removal of arsenic from aqueous solutions by ultrafiltration assisted with polyacrylamide as an environmental friendly complexing polymer. The system performance was evaluated in relation to quality of permeate in terms of operating variables as feed concentration of arsenic ions (C_o, μg L^{-1}), ratio of polymer to arsenic (r, w/w), and pH of feed solution. The effect of the operating variables and maximum arsenic removal efficiency was determined by adopting design of experiments and response surface methodology under different conditions for this polymer. The experimental data were analyzed with a second order polynomial model validated by statistical analysis. Based on the response model developed, the maximum removal efficiency, close to 100%, of arsenic ions has been obtained at optimum operating parameters as C_o: 150 μg L^{-1}, r: 2, and pH 10.

Keywords: Arsenic removal; Ultrafiltration; Polyacrylamide; Complexation; Response surface model

1. Introduction

Arsenic (As) in natural water is a worldwide problem since long-term exposure to elevated As concentrations threaten human health [1,2]. About 70 million people are suffering from an As problem in drinking water especially in South Asia, including India, Bangladesh, and also China [3]. Because of lethality to human health, The World Health Organization has set a provisional guideline value for As, at 10 μg L^{-1} as the maximum contaminant level in drinking water [4] and this standard level has also been implemented by the European Union and USEPA [5].

Several treatment methods, especially oxidation/precipitation, coagulation, adsorption, ion-exchange, nanofiltration, and reverse osmosis have been applied to comply with this As standard level [6–10]. These processes have some significant drawbacks, which are for instance incomplete removal of As, production of toxic sludges that can be a further potential source for secondary As pollution, and a high energy requirement [1,11–13]. However, integrated systems and hybrid technologies coupled with membrane filtration have recently drawn more attention because of success in reducing As levels to desired standard levels [14].

---

*Corresponding author.

At the time of the study, the authors Nigmet Uzal and Bekir Varol were affiliated with the Department of Environmental Engineering, Nigde University, 51240 Nigde, Turkey

Complexation and ultrafiltration (UF) is a hybrid pressure-driven membrane technology based on the complexation of metal ions with water soluble polymers in combination with UF. This technology has demonstrated its effectiveness in removing heavy metals from aqueous solutions [11,15,16]. This process is also defined as polymer enhanced ultrafiltration (PEUF) or polymer assisted UF process. Since the pore size of UF membranes are not appropriate to separate heavy metals, a water soluble polymer is used to complex with metallic ions and form a macromolecular structure, having a higher molecular weight than the molecular weight cut-off (MWCO) the UF membrane [15,16]. The advantages of complexation and UF process over the other treatment technologies include high separation selectivity [17,18] and a low energy requirement [11,18,19].

Although successful applications of heavy metal removal by complexation and UF have been reported in the literature [11,16,20], studies on As removal with complexation and UF are quite limited [16,21,22]. Sánchez and Rivas [23] studied arsenate retention by P(ClAETA) polymer, and high As removal was obtained for low feed As concentrations. In another study, cationic polymer poly (diallyldimethyl ammonium chloride) polyelectrolyte has been used to investigate the removal of As (V) from dilute aqueous solutions. It has been reported in the study that 99.5% removal efficiencies were achieved and the efficiency increases with increasing polymer concentration [21]. The published literature lacks any report on the performance of complexation and UF on As removal using polyacrylamide (PAM) as an environmental friendly complexation agent.

The main parameters that affect the complexation and UF process are pH, polymer loading ratio, existence of other metal ions, temperature, membrane type, trans-membrane pressure, and polymer concentration [24]. The conventional experimental approach for evaluating the process variables in complexation and UF experiments requires a large number of trials that are very time consuming. However, using design of experiments with response surface methodology (RSM) approach allows the evaluation of the relative significance of several variables together with a small number of experiments. RSM is an effective tool for the investigation, modeling, and optimization of complexation and UF processes [25]. RSM obtains the statistical design of experiments in which all factors are varied simultaneously over a set of experimental runs, provides estimation of coefficients in the mathematical model based on experimental design, checks the adequacy of the developed model, and predicts the response and optimization of experimental conditions using the valid model [26].

The present work deals with removal of As from aqueous solutions by complexation using PAM as a water soluble polymer and UF process. PAM is usually used as a flocculation agent in water and wastewater treatment and offers no systemic toxicity to aquatic life [27]. The main objective of this study is to fulfill the need in the literature for As removal with PEUF using PAM polymer. In this study, PAM was used to generate the macro-molecular metal complexes as an environmental friendly alternative polymer. The effect of operating variables and maximum As removal efficiency were determined by adopting design of experiments and RSM to analyze the effectiveness of the system under different conditions for this polymer. The experimental data were analyzed with a second order polynomial model validated by the analysis of variance (ANOVA) statistical analysis. The system performance was evaluated in relation to quality of permeate, in terms of operating variables as feed concentration of As ions (C_o, $\mu g\,L^{-1}$), ratio of polymer (PAM) to As (r, w/w), and pH of feed solution.

2. Materials and methods

2.1. Materials and membranes

All chemicals used in experiments were of analytical reagent grade, and all feed solutions were prepared using ultra-pure water. $Na_2HAsO_4{\cdot}7H_2O$ salt (Mw: 312.01 g/mol, Sigma–Aldrich) was used to prepare the synthetic aqueous solutions of As. For the initial pH arrangement of feed solutions, 0.2 M and 0.02 M nitric acid and sodium hydroxide solutions were used. In the experiments, PAM polymer with an average molecular weight of 10,000 (PAM, 50 wt% in H_2O, Sigma–Aldrich) was used as complexing agent. The UF membranes used in experiments were made of regenerated cellulose with a MWCO of 5 kDa. The diameter of the UF membrane was 63.5 mm with an effective area of 31.6 cm^2 for dead-end filtration.

2.2. Complexation and UF experiments

Complexation and UF experiments were designed to observe the effects of operating conditions on As retention. The operating variables were feed concentration of As ions (C_o, $\mu g\,L^{-1}$) between the ranges of 150–500 $\mu g\,L^{-1}$, ratio of polymer (PAM) to As (r, w/w) was between 2 and 5, and pH of feed solution was in the range 4–10. As levels were selected based on the groundwater As concentrations in Turkey, which are usually lower than 500 μg L^{-1} [28]. UF experiments were performed in a batch-stirred filtration cell (Amicon 8200, Millipore). The complexing polymer was added to the feed As solution at the determined concentration before filtration and then pH was fixed at the required pH level. The volume of the As solution used in UF experiments was 250 mL. In each experiment, the solution was stirred gently for about 1 h to achieve complex formation. After complexation, 150 mL feed solution was introduced to the dead-end filtration cell. Dead-end cell was stirred slowly to prevent the formation of concentration polarization. Dead-end UF experiments were carried out at 300 kPa pressure and 25 ± 2˚C temperature conditions. After each experiment, the membrane was thoroughly washed, *in situ*, by ultra-pure water. The UF cell was dismantled thereafter, and the membrane was washed in ultra-pure water between experiments, and the membranes were used repeatedly till the flux decline was over 5%. The experiments ended when the 30 mL volume of permeate was collected. The feed and permeate samples were then analyzed for their As content. The removal efficiency (denoted by Y) of As was determined in experiments as follows:

$$Y(\%) = \left(1 - C_p/C_o\right) \times 100$$

where C_p: permeate concentration As ions (μg L^{-1})
C_o: feed concentration of As ions (μg L^{-1})

2.3. Analytical methods

The concentrations of As ions in feed and permeate solutions were measured by Agilent 7500ce Inductively Coupled Plasma-mass Spectrophotometer (ICP-MS) which is an instrument combining a high-temperature ICP source with a mass spectrometer. A pH meter (Thermo Scientific, Orion 3 Star) was used for pH measurements of feed and permeate solutions throughout the study.

3. Results and discussion

The removal of As with UF process was carried out using the central composite experimental design (CCD). In the design of the experiments, Design Expert (Trial Version 6.0.7, Stat Ease, Inc., USA) was used for regression and graphical analyses of the data obtained. The experimental factors taken into account were as follows; feed concentration of As ions (C_o, μg L^{-1}), ratio of polymer (PAM) to As (r, w/w), and pH of feed solution. The ranges of operating parameters and the levels of the independent and controllable variables examined in this study are given in Table 1.

The data obtained from the experimental runs as mentioned above were fitted to a historical design for the application of RSM. A CCD of orthogonal type, which is widely used in statistical modeling to obtain response surface models that set the mathematical relationships between response and design variables, was also employed in this study for planning the experiments. The experimental plan is shown in Table 2, where the operating ranges and the levels of the considered variables are given in actual and coded values. The CCD experimental design consists of 16 experiments with 8 orthogonal design points, 6 star points with $\alpha = 1.215$ and for the replication 2 center points.

Multiple Linear Regression has been used to compute the regression coefficients of the response model. The estimation of regression coefficients can be calculated by using the following Eq. (1) [29–31] :

$$\bar{b} = (\bar{\bar{X}}^T \bar{\bar{X}})^{-1} \bar{\bar{X}}^T \bar{y} \tag{1}$$

where is a (L×1) vector of regression coefficients, is a (N×L) extended design matrix of the coded levels of input variables, is a (N×1) column vector of response determined experimentally according to the arrangement points into CCD, N is the number of experimental runs, and L is the number of regression coefficients within the response surface model. Therefore, the quadratic regression model with coded variables has been constructed and may be written as follows (Eq. (2)):

$$\begin{aligned}\acute{Y} = {} & 39.11 - 2.08\,x_1 - 0.83\,x_2 + 18.98\,x_3 + 0.76\,x_1x_2 \\ & - 9.31\,x_1x_3 - 1.74\,x_2x_3 + 18.56\,x_1^2 + 4.20\,x_2^2 \\ & + 12.70\,x_3^2\end{aligned} \tag{2}$$

Subject to: $-\alpha \le x_j \le +\alpha$; $j = 1, 2, 3$;
where x_1, x_2, and x_3 are the coded levels of process variables and α equals to 1.215, is the "star" point in central composite design that gives the limits of the valid region. The empirical model with actual factor values is as follows (Eq. (3)) where the empirical coefficients depend on the operating range of each input variable:

$$\begin{aligned}\acute{Y} = {} & 110.162 - 0.292C_o - 11.864r - 6.315\mathrm{pH} + 0.003C_or \\ & - 0.018C_o\mathrm{pH} - 0.386r\mathrm{pH} + 0.006{C_o}^2 + 1.867r^2 \\ & + 1.411\mathrm{pH}^2\end{aligned} \tag{3}$$

Subject to: $112.4 \le C_o \le 537.6$ (μg L^{-1}); $1.68 \le r \le 5.32$ (w/w); $3.35 \le \mathrm{pH} \le 10.65$.

Table 1
Experimental design variables of the As removal process by UF assisted by PAM complexation

		Actual values of coded levels				
Factors	Symbol	$-\alpha$*	−1	0	1	$+\alpha$
Feed concentration of As ions (C_o, μg L^{-1}),	x_1	112.4	150	325	500	537.6
Ratio of polymer (PAM) to As (r, w/w)	x_2	1.7	2	3.5	5	5.3
pH of feed solution	x_3	3.4	4	7	10	10.7

Note: $\alpha = 1.215$.

Table 2
Central composite orthogonal design applied for UF experiments for the As complexation with PAM.

		Factors (controllable input variables)						
		Feed concentration of As ions		Ratio of polymer to metal: PAM/As		pH of feed solution		
Run number (N)	Type[a]	C_o (μg L^{-1})	Level[b] (x_1)	r (w/w)	Level[b] (x_2)	pH	Level[b] (x_3)	Response[c]: Y (%)
1	O1	150	−1	2	−1	4	−1	41.7
2	O2	150	−1	2	−1	10	1	94.4
3	O3	150	−1	5	1	10	1	91.1
4	O4	500	1	5	1	10	1	65.9
5	O5	500	1	2	−1	4	−1	50.7
6	O6	500	1	2	−1	10	1	73
7	O7	150	−1	5	1	4	−1	38.5
8	O8	500	1	5	1	4	−1	57.4
9	C1	325	0	3.5	0	7	0	28.1
10	C2	325	0	3.5	0	7	0	50.6
11	S1	112.4	−α	3.5	0	7	0	78.6
12	S2	325	0	3.5	0	3.4	−α	38.7
13	S3	325	0	3.5	0	10.7	+α	97.8
14	S4	325	0	1.7	−α	7	0	56.6
15	S5	537.6	+α	3.5	0	7	0	75.2
16	S6	325	0	5.3	+α	7	0	54.8

[a] O: orthogonal design points, C: center points, S: star or axial points.

[b] −1 = low value, 0 = center value, +1 = high value, +/−α = star point value.

[c] As removal coefficient.

In order to test the adequacy of the response surface model and the results for the As removal coefficients, ANOVA was performed and the results are summarized in Table 3. According to ANOVA table, the R^2 value for As removal by UF assisted with PAM is 0.979, close to 1, which is acceptable. Moreover, the predicted R^2 was in agreement with the adjusted coefficient of determination R^2_{adj}, which is 0.941. Also, the F_{value} is significantly far from unity. In addition to this, the P_{value} is very low, determined as 0.0012 and smaller than 0.05 that shows the response surface model is statistically validated. All these statistical estimations indicate that response model used for the As removal is admissible from the statistical point of view for the prediction of the response in the considered range of factors (valid region).

Table 3
ANOVA of response surface model

	DF[a]	SS[b]	MS[c]	F_{value}	P_{value}	R^2	R^2_{adj}
Model	10	6638.5	809.3	25.68	0.0012	0.979	0.941
Residual	5	141.9	28.4				
Total	15	6780.4					

[a] Degree of freedom.

[b] Sum of squares.

[c] Mean square.

In addition to ANOVA tables, the model adequacy can easily be analyzed by examination of the residuals. The experimental data plotted against the response surface model and the internally studentized residuals are shown in Fig. 1. As can be seen, response surface model give good predictions for the experimental and predicted responses (Fig. 1(a)). Internally studentized residuals measures the number of standard deviations separating the actual and predicted values and the residual divided by the estimated standard deviation of that residual and used to validate the ANOVA. For the As removal, studentized residuals were obtained within the three sigma limits for the response model and the experimental values, and it is desirable (Fig. 1(b)).

In the next figures, (Figs. 2–4) the response surfaces plots and contour-lines maps are presented to investigate the effects of three different process variables, i.e. feed concentration of As ions, pH of feed solution, and ratio of polymer (PAM) to As ions. The effects of feed concentration of As ions (C_o, $\mu g\,L^{-1}$) and pH of feed solution, when the third factor (ratio of polymer (PAM) to As (r, w/w) is held at the central level r: 3.5, are shown in Fig. 2. As can be seen, at low pH of feed solution, the increase in the feed concentration of As ions does not considerably affect the As removal. However, with the increment in pH of feed solution, the rejection efficiency increases considerably. It is known that hydrolysis of the acrylamide pendant chains ($\pm CONH_2$), into acrylic acid groups ($\pm COOH$) takes place at basic pH values [31] and the dominant As species in aquatic systems at higher pH's (>10) are AsO_4^{3-} [13]. In considering the polymer As bonding under these basic conditions, the most probable situation is where the electrostatic repulsion between the hydrolyzed polymer and the negative surface charge of As species increases As removal efficiency [32,33].

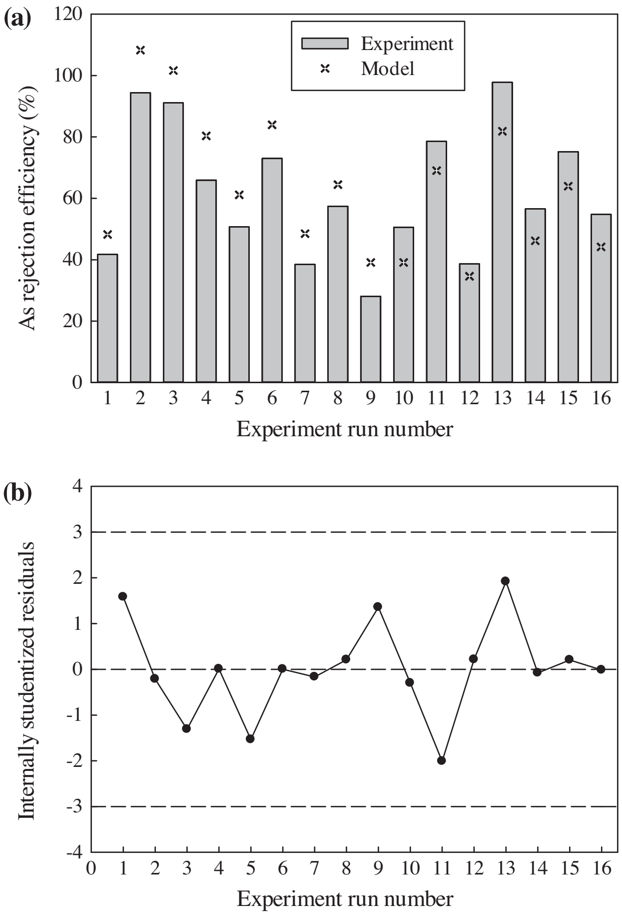

Fig. 1. Experimental data plotted against response model for the As removal efficiency (a) and internally studentized residuals (b).

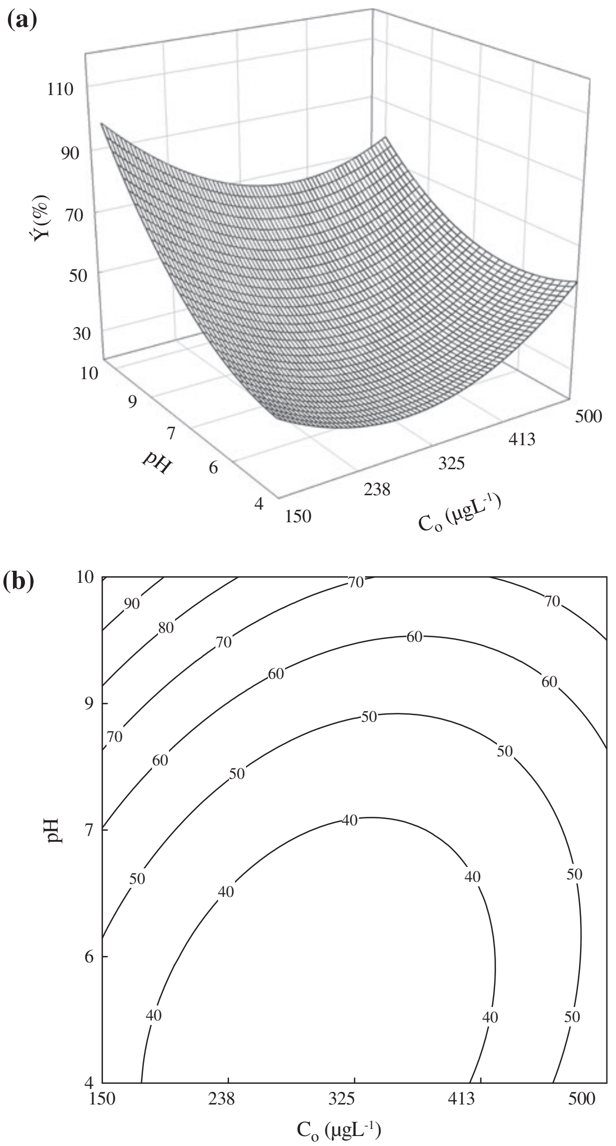

Fig. 2. (a) Response surface plot and (b) Contour plot indicating the effect of C_o and pH upon rejection coefficient for $r = 3.5$ (w/w).

In Fig. 3, the effects of feed concentration of As ions and ratio of polymer (PAM) to As are illustrated by holding at center level the third factor, i.e. pH 7. According to the figure, at fixed pH 7, the lowest As removal efficiency was obtained for *r*: 3.5 and 325 $\mu g\,L^{-1}$ initial As concentration at around 40%. It was observed that neither feed concentration of As ions nor ratio of polymer (PAM) to As have any significant effects on As removal efficiency. The maximum removal efficiency, close to 100%, of As ions was obtained at optimum operating parameters of C_o: 150 $\mu g\,L^{-1}$, *r*: 2, and pH 10. Lin et al. [34] has investigated the coupled chitosan/UF process for the removal of dissolved organic matter and As. For the feed As concentration of 1,000 $\mu g\,L^{-1}$ and the pH of 7.6, the authors reported 65% As rejection by this combined process.

Fig. 4 represents the effects of two factors, pH of feed solution and ratio of polymer (PAM) to As in combinations when the third factor (feed concentration of As ions, C_o) is held at the center level 325 $\mu g\,L^{-1}$. As one can see, the influence of pH of feed solution on the As removal efficiency is much more than the ratio of polymer (PAM) to As. With increasing pH, the rejection efficiency increases and the highest rejection values are attained under high pH conditions.

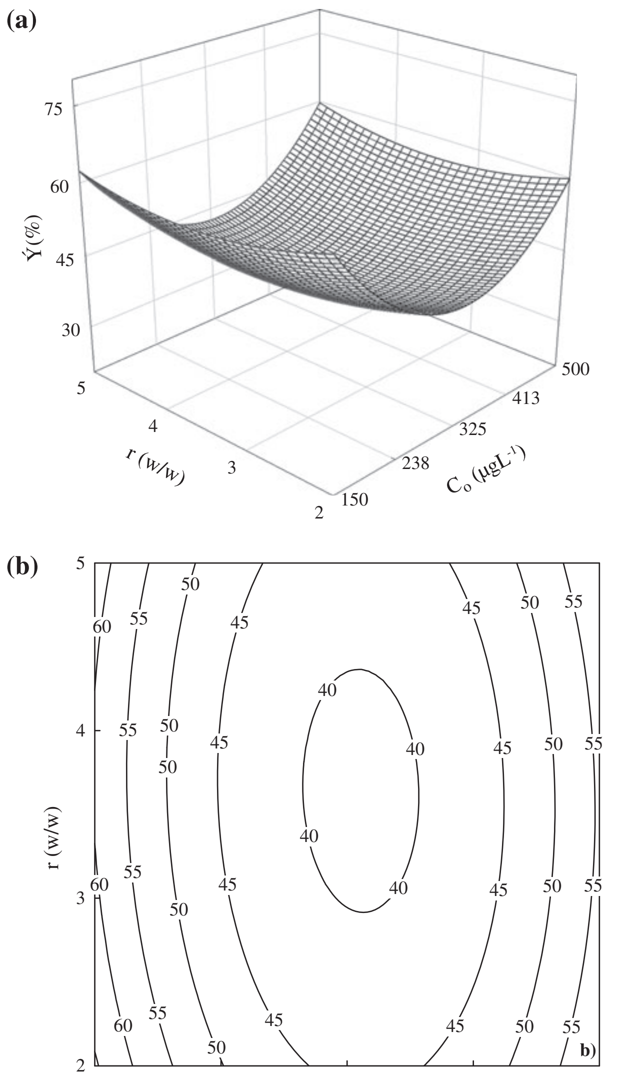

Fig. 3. (a) Response surface plot and (b) Contour plot indicating the effect of C_o and r upon rejection coefficient for pH 7.

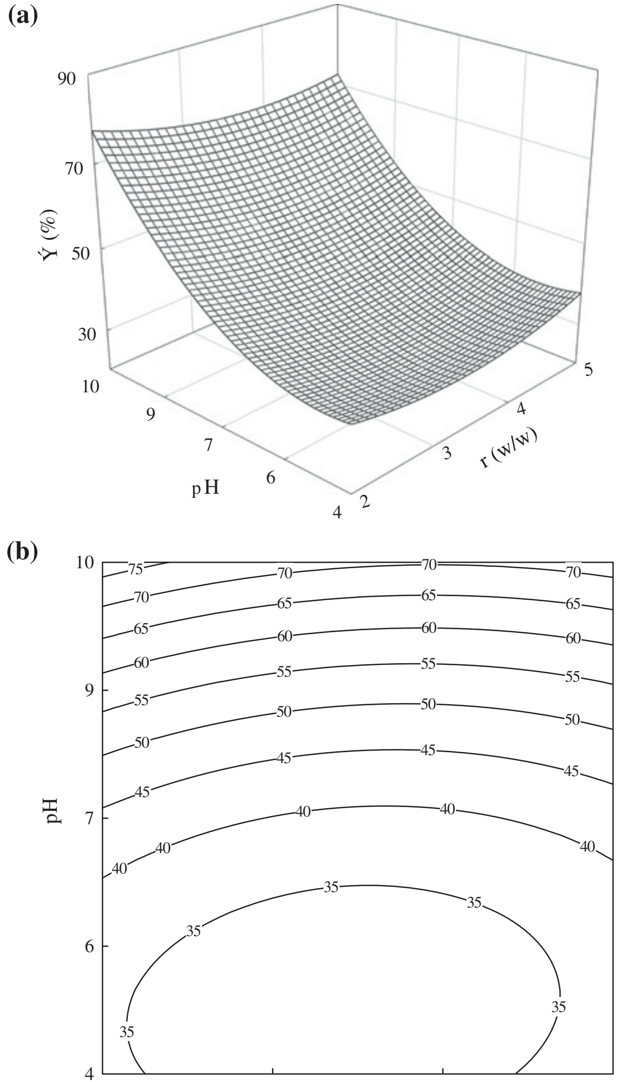

Fig. 4. (a) Response surface plot and (b) Contour plot indicating the effect of pH and *r* upon rejection coefficient for $C_o = 325\,\mu g\,L^{-1}$.

This fact is also consistent with the literature, and as the pH of the PAM solution becomes higher, the association of PAM cations with negatively charged As species can more easily take place [35]. In the As removal, the effect of the ratio of polymer (PAM) to As is negligibly small when compared to the effect of pH of the feed solution. Gecol et al. [36] and Gallo et al. [19] investigated the removal of As (V) from water using different polymers and surfactants with UF. The results of these studies showed that, the highest As removals are achieved at higher pH (over pH 8) which is in accordance with our study results.

Thus, the response surfaces illustrated in Figs. 2–4 for As removal efficiency (*Y*-response) indicates that the increasing of pH will lead to enhancing of As removal efficiency. The main effect of initial As concentration and polymer/metal ratio factor is negligible (Figs. 3 and 4) while the pH of the feed solution factor has the highest and the main effect (Fig. 2). The quadratic effects are similar for all factors giving the contribution to the response surface curvature. The relatively high interaction effect between the feed concentration of As ions and pH factors can be detected from Fig. 2 and there are no significant interaction effects between the feed concentration of As ions and *r* factors (Fig. 3). For instance, the effect of pH of the feed solution on As removal efficiency becomes more important at lower values of feed As ion concentration while the effect of r variable has no effect at higher pH values (Fig. 4).

4. Conclusions

In the present work As ions were successfully removed from synthetic aqueous solutions by UF assisted with PAM, an environmental friendly complexing polymer. The constructed response surface models were statistically validated by ANOVA and used for the prediction of removal efficiency over the valid region. The following conclusions can be drawn from the experimental and response model results:

- The pH of feed solution was found to have a prominent and direct effect on the As removal efficiency.
- As removal efficiency increased with the increase in pH of feed solution.
- The influence of feed concentration of As ions (C_o, $\mu g\,L^{-1}$) and ratio of polymer (PAM) to As (*r*, w/w) is much smaller than the effect of pH of feed solution.
- Based on the response model developed, the maximum removal efficiency, close to 100%, of As ions was obtained at optimum operating parameters at C_o: 150 $\mu g\,L^{-1}$, *r*: 2, and pH 10.
- It should be noted that there is still a necessity to validate the optimized operating parameters using real drinking water samples as a further study, before potential practical applications.

Acknowledgment

The authors gratefully acknowledge the financial support provided by the Nigde University Scientific Research Projects Unit with a research grant of BAP-FEB2010/22.

References

[1] M.C. Shih, An overview of arsenic removal by pressure-driven membrane processes, Desalination 172 (2005) 85–97.
[2] P.L. Smedley, D.G. Kinniburgh, A review of the source, behaviour and distribution of arsenic in natural waters, Appl. Geochem. 17 (2002) 517–568.
[3] P. Roy, N.K. Mondal, K. Das, Modeling of the adsorptive removal of arsenic: A statistical approach, J. Environ. Chem. Eng. 2 (2014) 585–597.
[4] WHO, Guidelines for Drinking-water Quality, fourth ed., World Health Organization, Geneva, 2011.
[5] T.S.Y. Choong, T.G. Chuah, Y. Robiah, F.L.G. Koay, I. Azni, Arsenic toxicity, health hazards and removal techniques from water: An overview, Desalination 217 (2007) 139–166.
[6] S. Bordoloi, S.K. Nath, S. Gogoi, R.K. Dutta, Arsenic and iron removal from groundwater by oxidation–coagulation at optimized pH: Laboratory and field studies, J. Hazard. Mater. 260 (2013) 618–626.
[7] E. Mohora, S. Rončević, B. Dalmacija, J. Agbaba, M. Watson, E. Karlović, M. Dalmacija, Removal of natural organic matter and arsenic from water by electrocoagulation/flotation continuous flow reactor, J. Hazard. Mater. 235–236 (2012) 257–264.
[8] M. Sen, A. Manna, P. Pal, Removal of arsenic from contaminated groundwater by membrane-integrated hybrid treatment system, J. Membr. Sci. 354 (2010) 108–113.
[9] Y. Yu, C.W. Zhao, Y.G. Wang, W.H. Fan, Z.K. Luan, Effects of ion concentration and natural organic matter on arsenic(V) removal by nanofiltration under different transmembrane pressures, J. Environ. Sci. 25 (2013) 302–307.
[10] Y. Zhang, M. Yang, X.M. Dou, H. He, D.S. Wang, Arsenate adsorption on an Fe−Ce bimetal oxide adsorbent: Role of surface properties, Environ. Sci. Technol. 39 (2005) 7246–7253.
[11] M.A. Barakat, E. Schmidt, Polymer-enhanced ultrafiltration process for heavy metals removal from industrial wastewater, Desalination 256 (2010) 90–93.
[12] A.H. Malik, Z.M. Khan, Q. Mahmood, S. Nasreen, Z.A. Bhatti, Perspectives of low cost arsenic remediation of drinking water in Pakistan and other countries, J. Hazard. Mater. 168 (2009) 1–12.

[13] D. Mohan, C.U. Pittman, Arsenic removal from water/wastewater using adsorbents—A critical review, J. Hazard. Mater. 142 (2007) 1–53.

[14] P. Mondal, S. Bhowmick, D. Chatterjee, A. Figoli, B. Van der Bruggen, Remediation of inorganic arsenic in groundwater for safe water supply: A critical assessment of technological solutions, Chemosphere 92 (2013) 157–170.

[15] C. Cojocaru, G. Zakrzewska-Trznadel, A. Jaworska, Removal of cobalt ions from aqueous solutions by polymer assisted ultrafiltration using experimental design approach. Part 1: Optimization of complexation conditions, J. Hazard. Mater. 169 (2009) 599–609.

[16] M. Palencia, B.L. Rivas, E. Pereira, Metal ion recovery by polymer-enhanced ultrafiltration using poly(vinyl sulfonic acid): Fouling description and membrane–metal ion interaction, J. Membr. Sci. 345 (2009) 191–200.

[17] F.L. Fu, Q. Wang, Removal of heavy metal ions from wastewaters: A review, J. Environ. Manage. 92 (2011) 407–418.

[18] C.W. Li, C.H. Cheng, K.H. Choo, W.S. Yen, Polyelectrolyte enhanced ultrafiltration (PEUF) for the removal of Cd(II): Effects of organic ligands and solution pH, Chemosphere 72 (2008) 630–635.

[19] D. Gallo, E.J. Acosta, J.F. Scamehorn, D.A. Sabatini, Pilot-scale study of polyelectrolyte-enhanced UF for arsenic removal, J. Am. Water Works Assn. 98 (2006) 106–116.

[20] F. Balaska, M. Bencheikh-Lehocine, M. Chikhi, A.H. Meniai, A. Bouledjouidja, Removal of chromium (III) ions from aqueous solutions by polymer assisted ultrafiltration using experimental and calculation approach. Part 1: Optimization of complexation conditions, in: C. Salame, M. Aillerie, G. Khoury (Eds.), Terragreen 2012: Clean Energy Solutions for Sustainable Environment, Beirut, 2012, pp. 622–631.

[21] P. Pookrod, K.J. Haller, J.F. Scamehorn, Removal of arsenic anions from water using polyelectrolyte-enhanced ultrafiltration, Sep. Sci. Technol. 39 (2004) 811–831.

[22] B.L. Rivas, M. del Carmen Aguirre, E. Pereira, Cationic water-soluble polymers with the ability to remove arsenate through an ultrafiltration technique, J. Appl. Polym. Sci. 106 (2007) 89–94.

[23] J. Sánchez, B.L. Rivas, Arsenate retention from aqueous solution by hydrophilic polymers through ultrafiltration membranes, Desalination 270 (2011) 57–63.

[24] P. Cañizares, A. Pérez, R. Camarillo, R. Mazarro, Simultaneous recovery of cadmium and lead from aqueous effluents by a semi-continuous laboratory-scale polymer enhanced ultrafiltration process, J. Membr. Sci. 320 (2008) 520–527.

[25] N. Uzal, A. Jaworska, A. Miśkiewicz, G. Zakrzewska-Trznadel, C. Cojocaru, Optimization of CO^{2+} ions removal from water solutions via polymer enhanced ultrafiltration with application of PVA and sulfonated PVA as complexing agents, J. Colloid Interface Sci. 362 (2011) 615–624.

[26] C. Cojocaru, G. Zakrzewska-Trznadel, Response surface modeling and optimization of copper removal from aqua solutions using polymer assisted ultrafiltration, J. Membr. Sci. 298 (2007) 56–70.

[27] K.R. Desai, Z.V.P. Murthy, Removal of silver from aqueous solutions by complexation–ultrafiltration using anionic polyacrylamide, Chem. Eng. J. 185–186 (2012) 187–192.

[28] O. Gunduz, C. Bakar, C. Simsek, A. Baba, A. Elci, H. Gurleyuk, M. Mutlu, A. Cakir, Statistical analysis of death causes (2005–2010) in villages with high arsenic levels in drinking water supplies of Simav plain, Turkey, Arch. Environ. Occup. Health. doi: 10.1080/19338244.2013.872076.

[29] S. Akhnazarova, V. Kafarov, Experiment Optimization in Chemistry and Chemical Engineering, Mir, Moscow, 1982.

[30] G.E.P. Box, N.R. Draper, Response Surfaces, Mixtures, and Ridge Analyses, second ed., Wiley, Hoboken, NJ, 2007.

[31] D.C. Montgomery, Design and Analysis of Experiments, fifth ed., Wiley, New York, 2001.

[32] L.T. Lee, P. Somasundaran, Adsorption of polyacrylamide on oxide minerals, Langmuir 5 (1989) 854–860.

[33] J.M. Gonzalez-Saiz, C. Pizarro, Polyacrylamide gels as support for enzyme immobilization by entrapment. Effect of polyelectrolyte carrier, pH and temperature on enzyme action and kinetics parameters, Eur. Polym. J. 37 (2001) 435–444.

[34] C.-F. Lin, C.-H. Wu, H.-T. Lai, Dissolved organic matter and arsenic removal with coupled chitosan/UF operation, Separ. Purif. Technol. 60 (2008) 292–298.

[35] N. Tekin, A. Dinçer, Ö. Demirbaş, M. Alkan, Adsorption of cationic polyacrylamide onto sepiolite, J. Hazard. Mater. 134 (2006) 211–219.

[36] H. Gecol, E. Ergican, A. Fuchs, Molecular level separation of arsenic(V) from water using cationic surfactant micelles and ultrafiltration membrane, J. Membr. Sci. 241 (2004) 105–119.