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Response Surface Methodology for Radioactive Strontium Adsorption on Molecular Sieves

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Abstract

In the nuclear industry, the disposal of radioactive waste is a crucial issue. Strontium is one of the most dangerous radionuclides to human health. In this study, strontium 85 adsorption on molecular sieves was investigated. The factors affecting the adsorption on molecular sieves were examined. In order to increase adsorption, the sorbent (molecular sieves) was activated before contact with radioactive strontium. The response surface methodology was used to evolve the predictive regression model for adsorption of radioactive strontium on molecular sieves. The experimental and predicted maximum decontamination factor 14.23 and 12.93 was obtained, respectively. Molecular sieves were found to be useful for the removal of radioactive strontium from water solution.

Keywords: Response surface methodology, adsorption, radioactive strontium, molecular sieves

1. Introduction

Radioisotopes are delivered into the environment from nuclear facilities. During the normal functioning of nuclear power plants, radioactive liquid waste is produced besides nuclear accidents [1-3]. Due to its toxicological effect on the human health radioactive wastewater is a serious environmental concern. The sample of these health problems are cancer and carcinogenic diseases. Radioactive strontium is a main contaminant in low-level radioactive wastewaters from nuclear power plants. There are different types of cations including nonradioactive strontium in the environment [2-4].

The atomic number of elemental strontium is 38. Stable strontium is a soft, silver-gray metal. Strontium occurs naturally in the earth as a mixture of four stable isotopes. ^{85}Sr (half-life = 64.84 d) is radioactive strontium isotope which occur in the environment from routine discharges from nuclear installations and from nuclear accidents [5].

Because of its relatively long half-life and high toxicity the removal of strontium from wastewaters is of great interest. Strontium is one of the most harmful radionuclides in radioactive liquid waste. A half-life of strontium-90 28.8 years and it is a pure beta emitter of 0.546 MeV. It is generally found in many groundwater systems coming from historic nuclear waste storage sites [3, 4, 6, 7]. The contamination by the radioactive strontium increases environmental interest on the healthy water [2, 8].

In order to decrease the volume of radioactive waste, many techniques including adsorption, ion-exchange processes, evaporation have been reported. Among the methods, adsorption method is examined to be economic and effective, due to its radiation stability [2, 3, 4, 9]. Although a number of investigations have focused on the application of sorbents for the removal of radionuclide from waste solutions, however few have reviewed adsorption modelling with response surface methodology (RSM) [3, 10-12]. This study aimed to investigate molecular sieves application for the removal of strontium from waste solutions.

2. Materials and Methods

Molecular sieve (Merck, 105705) is used in this study. ^{85}Sr was obtained from Institute of Nuclear Chemistry and Technology, Warsaw, Poland. The radionuclidic purity of ^{85}Sr was determined by gamma spectrometry. Molecular sieve was activated for two hours at 873 K before contact with strontium ions. Subsequently molecular sieve was added to the strontium 85. We shook the mixtures for 4 h. Thereafter the mixtures were filtered with a syringe filter. The activities of strontium 85 were measured with Polon Warszawa Analyzer (A-22p HT Power supply ZW N-21M HT Control 0/2000V). For all liquid radioactivity measurements, a volume of 10 mL solution was used. Each measurement was repeated for 10 times. The experimental decontamination factor was described as below:

$$DF = A_0 / A_f \quad \textbf{(1)}$$

where A_0 was the initial activity of Strontium 85 feed solution (Bq/L) and A_f was the final activity (Bq/L).
More details regarding experiment can be found in previous studies [12, 13].

We regulated the experimental analysis and modeling using RSM as a useful mathematical and statistical technique as described previously in details [12-16]. The response decontamination factor can be determined which is influenced by various input variables in RSM model.

The general form of RSM can be present as below:

$$DF = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{12} X_1 X_2 \quad \textbf{(2)}$$

$$\beta = [\beta_0\ \beta_1\ \beta_2\ \beta_{11}\ \beta_{22}\ \beta_{12}]^T \quad \textbf{(3)}$$

$$\beta = (X^T \cdot X)^{-1} \cdot X^T \cdot DF \quad \textbf{(4)}$$

where: β – shows regression coefficients;
X –input values;
DF – decontamination factor (response).
Adsorption of strontium ions solutions onto molecular sieves;
- regression model with actual variables:

$$\widehat{DF} = 7.66 - 123.1SD - 0.00042A_0 + 1257SD^2 - 0.00711SDA_0 \quad \textbf{(5)}$$

valid for the range:

$0.05 \leq SD \leq 0.15$ (%w/v); $4037 \leq A_0 \leq 8992$ (Bq/L)

The calculation details can be found in previous studies [12- 19]. All calculations were done by means of Minitab 19 software.

3. Results and Discussion

The ANOVA was employed to test the accuracy of the calculated model. According to the results the model was compatible where the probability value was 0.016. F value was 11.96 it was indicating that the experimental decontamination factor obtained by changing the factor levels were statistically meaningful at the 94% confidence limit. R^2 value should be close to 1 for a better statistical model. The mathematical model is adequate for the prediction radioactive strontium 85 removal by molecular sieves sorption since $R^2 = 0.94 > (0.75)$. Lack of fit F and p values are 32.17 and 0.129 ($P > 0.05$), respectively. Lack of fit was not significant and this means that the model is adequate [19, 20].

Table 1. Strontium 85 experimental design for molecular sieves.

Run number (N)	Factors (input values)					Response	
	Amount of sorbent (g/100ml)		Initial activity of Strontium 85 Bq/L		Final activity Bq/L	Decontamination Factor (DF)	
						Experimental	Predicted
	SD (% w/v)	level [a] x_1	C_0 (Bq/L)	level [a] x_2		*DF*	*DF*
1	0.15	1	8992	1	762.13	11.80	11.35
2	0.05	-1	8992	1	2141.12	4.20	4.91
3	0.15	1	4037	-1	283.68	14.23	12.93
4	0.05	-1	4037	-1	1298.78	3.11	2.96
5	0.15	1	6514.5	0	661.71	9.84	11.59
6	0.05	-1	6514.5	0	1651.13	3.95	3.38
7	0.1	0	8992	1	1714.54	5.24	4.99
8	0.1	0	4037	-1	1200.13	3.36	4.81
9	0.1	0	6514.5	0	1379.29	4.72	4.34
10	0.1	0	6514.5	0	1266.07	5.15	4.34

a −1 = low value. 0 = center value. +1 = high value.

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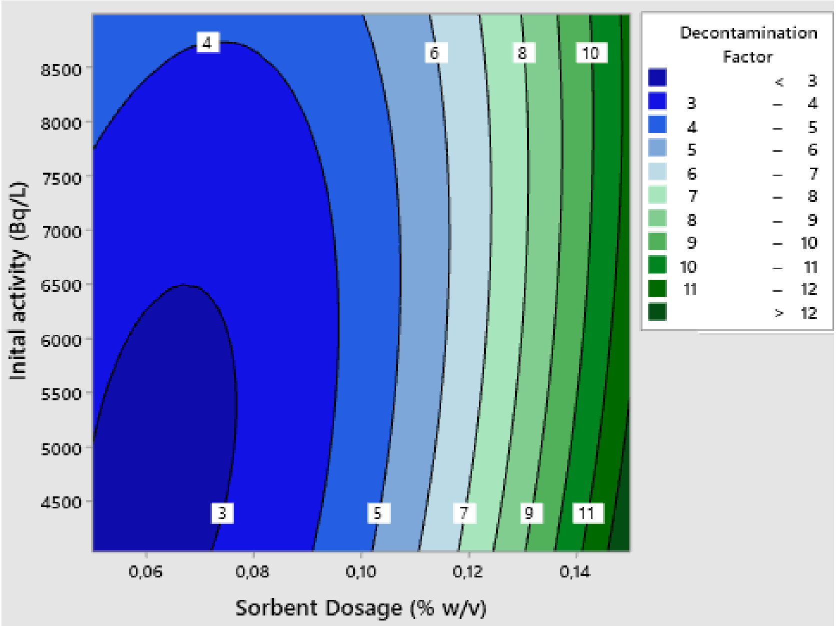

Figure 1. Response surface plot of decontamination factor response depending on for molecular sieves- Radioactive strontium.

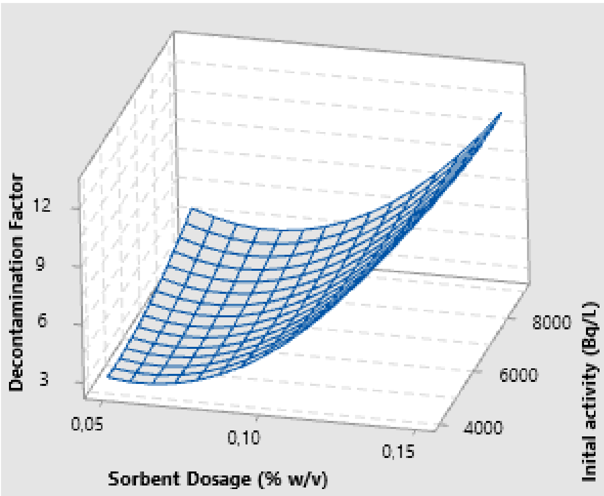

Figure 2. 3D-Response surface plot of decontamination factor for molecular sieves- Radioactive strontium.

The experimental and predicted minimum decontamination factor 3.11 and 2.96 was obtained. respectively. The experimental and predicted maximum decontamination factor is 14.23 and 12.93 respectively.

In a previous study, the optimal conditions for zeolite 3A were found to be of initial activity of ^{85}Sr feed solution =8992 Bq/L and sorbent dosage 0.1 %w/v. A maximal decontamination factor (experimentally) was 41.67 [12]. In this study, decontamination factor of 5.24 was found experimentally for molecular sieves in same conditions.

Strontium is one of the prominent radionuclides in radioactive wastewater. There are many techniques for the removal of radionuclides from wastewater.

Among these removal techniques. adsorption is simple and has tremendous efficiency. The adsorption capacities rise as the sorbent dosage augmentation. The diverse cations interfere with the strontium adsorption on molecular sieves through the purification. Radioactive wastewater such as strontium generated from the nuclear fuel cycle. Radioactive elements have huge potential of collection in plants causes important environmental and health problems [2- 4].

4. Conclusions

In this study. initial activity and the influence of sorbent amount on decontamination factor were investigated by means of RSM. It was found that experimental design and RSM are the efficient approaches for modeling of the removal efficiency of strontium ions.

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Author's Contributions

Ekrem ÇİÇEK: Drafted and wrote the manuscript, performed the experiment and result analysis.

Ethics

There are no ethical issues after the publication of this manuscript.

References

1. Honda, MC, Aono, T, Aoyama, M, Hamajima, Y, Kawakami, H, Kitamura, M, Masumoto, Y, Miyazawa, Y, Takigawa, M, Saino, T. 2012. Dispersion of artificial caesium-134 and -137 in the western North Pacific one month after the Fukushima accident. *Geochemical Journal;* 46: 1-9.

2. Munthali, MW, Johan, E, Aono, H, Matsue, N. 2015. Cs+ and Sr2+ adsorption selectivity of zeolites in relation to radioactive decontamination. *Journal of Asian Ceramic Societies;* 3: 245-250.

3. Zhang, L, Wei, J, Zhao, X, Li, F, Jiang, F, Zhang, M, Cheng, X. 2016. Competitive adsorption of strontium and cobalt onto tin antimonite. *Chemical Engineering Journal;* 285: 679-689.

4. Hasan, S, Iasir, ARM, Ghosh, TK, Gupta, BS, Prelas, MA. 2019. Characterization and adsorption behavior of strontium from aqueous solutions onto chitosan-fuller's earth beads. *Healthcare;* 7: 52-70.

5. Burger A, Lichtscheidl I. 2019. Strontium in the environment: Review about reactions of plants towards stable and radioactive strontium isotopes. *Science of The Total Environment;* 653: 1458-1512.

6. Pathak, P. 2017. An assessment of strontium sorption onto bentonite buffer material in waste repository. *Environmental Science and Pollution Research*; 24: 8825–8836.

7. Sureda, R, Martinez-Llado, X, Rovira, M, Pablo, JD, Casas, I, Gimenez, J. 2010. Sorption of strontium on uranyl peroxide: Implications for a high-level nuclear waste repository. *Journal of Hazardous Materials;* 181: 881–885.

8. Lee, HS, Lee, J, Yoon, B, Yim, Y, Choi, I, Cho, H, Lee, S, Baik, K, Park, JH, Huh, YJ. 2013. Removal characterization of 133Cs and 127I in a water treatment process using a laboratory scale experiment. *Water Supply;* 13(5): 1289–1294.

9. Rahman, ROA, Ibrahium, HA, Hung, YT. 2011. Liquid radioactive wastes treatment: A review. *Water;* 3: 551–565.

10. Tayyebi, A, Outokesh, M, Moradi, S, Doram, A. 2015. Synthesis and characterization of ultrasound assisted ''graphene oxide–magnetite'' hybrid. and investigation of its adsorption properties for Sr(II) and Co(II) ions. *Applied Surface Science;* 353: 350–362.

11. Chen, C, Hu, J, Shao, D, Li, J, Wang, X. 2009. Adsorption behavior of multiwall carbon nanotube/iron oxide magnetic composites for Ni(II) and Sr(II). *Journal of Hazardous Materials;* 164: 923–928.

12. Cicek, E, Cojocaru, C, Zakrzewska-Trznadel, G, Harasimowicz, M, Miskiewicz, A. 2012. Response surface methodology for the modeling of 85 Sr adsorption on zeolite 3A and pumice. *Environmental Technology;* 33(1): 51–59.

13. Cicek, E, Cojocaru, C, Zakrzewska-Trznadel, G, Jaworska, A, Harasimowicz, M. 2008. Response surface methodology for cobalt removal from aqua solutions using Isparta pumice and zeolite 4A adsorbents. *Nukleonika*; 53(S2): 121-128.

14. Cicek, E. 2009. Response surface methodology for cobalt removal from aqua solutions using nevsehir and Kayseri pumice adsorbents. *Asian Journal of Chemistry;* 21(7): 5727-5736.

15. Myers, RH, Montgomery, DC. Response Surface Methodology: Process and Product Optimization Using Designed Experiments; John Wiley &Sons: New York. 2 ed. 2002.

16. Akhnazarova, S, Kafarov, V. Experiment Optimization in Chemistry and Chemical Engineering; Mir Publishers: Moscow. 1982.

17. Khayet, M, Cojocaru, V, Zakrzewska-Trznadel, G. 2008. Response surface modelling and optimization in pervaporation. *Journal of Membrane Science;* 321: 272–283.

18. Cojocaru, C, Macoveanu, M. 2003. Modeling and Optimization of Diesel Oil Spill Removal from Water Surface Using Shredded Strips of Polypropylene as the Sorbent. *Environmental Engineering and Management Journal;* 2(2): 145-154.

19. Le, MH, Behera, SK, Park, HS. 2010. Optimization of operational parameters for ethanol production from Korean food waste leachate. *International Journal of Environmental Science & Technology;* 7: 157–164.

20. Chauhan, B, Gupta, R. 2004. Application of statistical experimental design for optimization of alkaline protease production from Bacillus sp. RGR-14. *Process Biochemistry;* 39(12): 2115–2122.