

The Modeling of Radioactive Cobalt Adsorption on Molecular Sieves

Ekrem Çiçek ^{1,a,*}

¹ Department of Physics, Faculty of Science and Letters, Mehmet Akif Ersoy University, Burdur, Turkey

*Corresponding author

Research Article

History

Received: 07/09/2020

Accepted: 13/01/2022

Copyright



©2022 Faculty of Science,
Sivas Cumhuriyet University

ABSTRACT

Radioactive wastes are products of nuclear activities around the world. Radioactive cobalt is one of the usually found radionuclide in nuclear waste. It is crucial to separate radioactive cobalt from aqueous media. The removal of radioactive cobalt (Cobalt-60) was investigated using molecular sieves in this study. The molecular sieves structure comprises of a microporous and aluminosilicate framework. Due to their chemical composition and structures molecular sieves have excellent sorption capacities. The response surface methodology was utilized to constitute the predictive regression model. The experimental minimum and maximum decontamination factor 2.5 and 11.1 was obtained, respectively. The predicted maximum decontamination factor was 10. Molecular sieves present a high adsorbent capacity for the disposal radioactive cobalt from water solution

Keywords: Adsorption, Radioactive cobalt, Response surface methodology, Molecular sieves.

ekrcicek@gmail.com

<https://orcid.org/0000-0001-6724-9423>

Introduction

The nuclear power stations and nuclear laboratories are increasing [1]. Nuclear activities have produced too much amount of radioactive waste. These wastes cause to significant risks to the environments [2]. They always have been recognized as a serious threat for human health. Owing to their health implications the public concerns are raising on nuclear safety. Disposal of radioactive waste are receiving more attention since nuclear application increased globally [3]. Radioactive contamination is severely dangerous and it is not easy task to handling. For protection against radioactive wastes the new methods were developed by researchers [4,5].

A great quantity of nuclear waste has been produced from nuclear facilities with the fast development of the nuclear field [6]. Cobalt-60 is produced artificially and described as a major environmental contaminant at nuclear region [7]. Radioactive isotope of cobalt has many useful applications [8].

Radioactive isotope of cobalt (Cobalt-60) employed in medicine and industry. The radioactive cobalt is used commonly in radiotherapy units as gamma source for treatment. In nuclear medicine, it (so low activity) is just used as to be flat gamma source to check gamma camera quality control [9]. It is also used to investigate materials and sterilization. The half-life of cobalt-60 is 5.27 years [8].

The disposal of radioisotopes from the environment comprises a number of methods such as filtration, reverse osmosis, precipitation, vacuum evaporation, extraction and adsorption by cation exchange [10]. Adsorption has received rising attention because of its advantages in radioactive wastewater treatment [6].

Different types of sorbents have been utilized to remove nuclear waste. Because of its excellent removal capacity, zeolite is the one of the best choice for radioactive waste adsorption [3].

Zeolites are crystalline materials with three dimensional frameworks [11]. They have micro porous and their frameworks are charged [3]. Alkaline metallic cations and porosity made the zeolite hydrophilic and excellent adsorptive material [2,4,12]. The advantages of zeolites are extensive ion exchange capacity, perfect selectivity and low cost are [6]. The sorption capacity of zeolite is generally depending to pore size [3].

Zeolite is a porous mineral with exchangeable ions. The cavities and channels of zeolites can capture cations, water, and radionuclides. Due to their high cation-exchange capacities, they have been considered for radioactive waste treatment [13]. Removal of radionuclides from aqueous solution is particularly important in cleaning for environmental contamination [3]. Cations exchange and adsorption are the major mechanism for discharging radioisotopes from water solution. Since zeolite comprises uniform small pores they present excellent adsorption and exchange capacity [6].

Adsorption modelling is examined and decontamination factor (response) is predicted by response surface methodology (RSM). The experimental input variables are amount of adsorbent and initial activity of solution.

In this study, molecular sieves (a kind of zeolite) were exploited for the discharging of radioactive cobalt from aqueous media.

Materials and Methods

Experimental procedure

We used molecular sieve obtained from Merck (105705) in this study. In order to remove water from molecular sieve, it was activated with temperature for two hours at 873 K. After activation process, molecular sieve was added to the radioactive cobalt and suspensions were mixed for 4 h. Thereafter the mixtures were filtered with a syringe filter (Whatman Syringe Filter 25 mm diameter, 0,2 µm pore size). The activities of radioactive cobalt were measured with Polon Warszawa Analyzer (A-22p HT Power supply ZW N-21M HT Control 0/2000V). For all liquid radioactive cobalt measurements, a volume of 10 mL solution was utilized. The measurements were repeated for 10 times. The experimental decontamination factor was presented as below:

$$DF = A_0 / A_f \tag{1}$$

where A_0 was the initial activity of radioactive cobalt feed solution (Bq L⁻¹) and A_f was the final radioactivity (Bq L⁻¹).

More details concerning experiment can be found in previous studies [13-15].

We utilized RSM for analysis and modeling. It is a useful statistical and mathematical technique as described previous studies in details [13-17]. The decontamination factor (response) can be described which is influenced by controllable various input values in RSM model.

The general form of RSM can be demonstrated as below:

$$\hat{DF} = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{12} x_1 x_2 \tag{2}$$

$$\beta = [\beta_0 \beta_1 \beta_2 \beta_{11} \beta_{22} \beta_{12}]^T \tag{3}$$

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

where: β – present regression coefficients;

X –input variables;

DF – decontamination factor (response).

Adsorption of radioactive cobalt upon molecular sieves;

- regression model with actual variables:

$$\hat{DF} = 5.63 - 56.3SD - 0.0005A_0 + 764SD^2 - 0.00256SDA_0 \tag{5}$$

valid for the range: $0.05 \leq SD \leq 0.15$ (%w v⁻¹); $7600 \leq A_0 \leq 15200$ (Bq L⁻¹)

The modeling details can be seen in previous studies [13-18]. All calculations were done by means of Minitab 19 software.

Results and Discussion

The ANOVA was used to examine the accuracy of the calculated model. According to the results the model was compatible where the probability value was 0.028. F value was 8.75, it was pointed out that the experimental decontamination factor obtained by changing the factor levels were statistically meaningful at the 92% confidence limit. R² value should be close to 1 for a good statistical model. The mathematical model is adequate for the prediction radioactive cobalt removal by molecular sieves sorption since R² = 0.92 > (0.75). Lack of fit F and p values are 38.71 and 0.117 (P>0.05), respectively. Lack of fit was not significant and this means that the model is good [18,19].

Table 1. Radioactive cobalt experimental design for molecular sieves

Run number (N)	Factors (input values)				Final activity Bq L ⁻¹	Response	
	Amount of sorbent (g 100ml ⁻¹)		Initial activity of radioactive cobalt Bq L ⁻¹			Decontamination Factor (DF)	
	Sorbent Dosage (%w v ⁻¹)	level a x1	CO (mg L ⁻¹)	level a x2		Experimental DF	Predicted DF
1	0.15	1	15200	1	1492.61	10.2	10.0
2	0.05	-1	15200	1	4317.62	3.5	4.2
3	0.15	1	7600	-1	683.32	11.1	9.9
4	0.05	-1	7600	-1	3026.31	2.5	2.2
5	0.15	1	11400	0	1433.73	8.0	9.4
6	0.05	-1	11400	0	3732.90	3.1	2.7
7	0.1	0	15200	1	2678.49	5.7	5.2
8	0.1	0	7600	-1	2875.68	2.6	4.2
9	0.1	0	11400	0	2568.94	4.4	4.1
10	0.1	0	11400	0	2378.59	4.8	4.1

^a -1 = low. 0 = center. +1 = high.

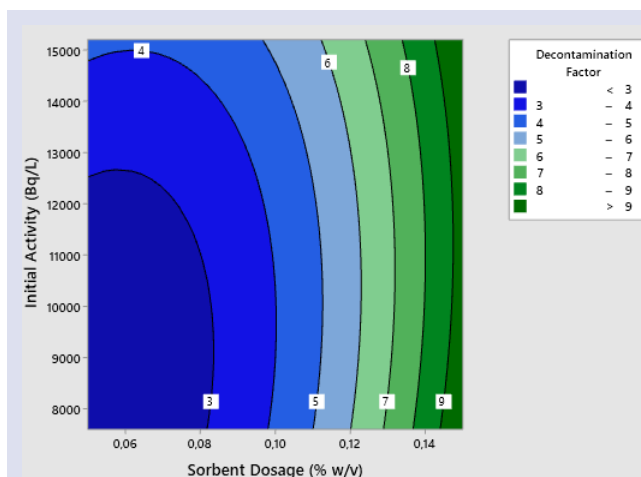


Figure 1. Contour plot of decontamination factor for molecular sieves - radioactive cobalt

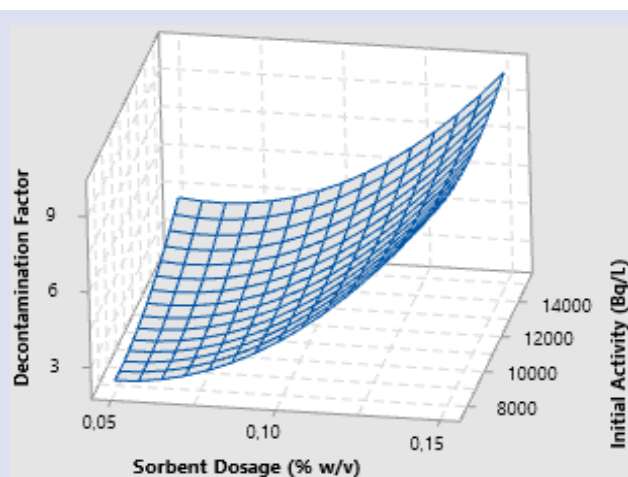


Figure 2. 3D-Response surface plot of decontamination factor for molecular sieves- radioactive cobalt

Conclusion

We examined the ability of removal of the radioactive cobalt by molecular sieves in aqua solution in this study. Initial cobalt activity and the influence of molecular sieves amount on decontamination factor were investigated by means of RSM. RSM can predict decontamination factor for different amount of sorbent dosage and initial activity. We suggested that molecular sieves might be used as an adsorbent for the removal of radioactive cobalt in nuclear waste. Also. RSM can prevent many experimental runs.

Conflicts of interest

The authors declare no conflict of interest.

References

- Zhong Q.Q., Zhao Y.Q., Shen L., Hao B., Xu X., Gao B.Y., Shang Y.N., Chu K.Z., Zhang X.H., Yue Q.Y., Single and binary competitive adsorption of cobalt and nickel onto novel magnetic composites derived from green macroalgae, *Environ. Eng. Sci.*, 37(3) (2020) 188-200.
- Lee H.Y., Kim H.S., Jeong H.K., Park M., Chung D.Y., Lee K.Y., Lee E.H., Lim W.T., Selective removal of radioactive cesium from nuclear waste by zeolites: on the origin of cesium selectivity revealed by systematic crystallographic studies, *J. Phys. Chem. C*, 121(19) (2017) 10594–10608.
- Ovhal S., Butler I.S., Xu S., The potential of zeolites to block the uptake of radioactive strontium-90 in organisms, *Contemporary Chemistry*, 1(1) (2018) 1-13.
- Sadeghi M., Yekta S., Ghaedi H., Babanezhad E., Effective removal of radioactive ^{90}Sr by CuO NPs/Agclinoptilolite zeolite composite adsorbent from water sample: isotherm. kinetic and thermodynamic reactions study, *Int. J. Ind. Chem.*, 7 (2016) 315–331.
- Olatunji M.A., Khandakar M.U., Mahmud H.N.M.E., Amin Y.M., Influence of adsorption parameters on cesium uptake from aqueous solutions- a brief review, *RSC Adv.*, 5 (2015) 71658-71683.
- Fang X.H., Fang F., Lu C.H., Zheng L., Removal of Cs, Sr²⁺ and Co²⁺ ions from the mixture of organics and suspended solids aqueous solutions by zeolites, *Nucl Eng Technol.*, 49(3) (2017) 556-561.
- Handley-Sidhu S., Mullan T.K., Grail Q., Albadameh M., Ohnuki T., Macaskie L.E., Influence of pH, competing ions and salinity on the sorption of strontium and cobalt onto biogenic hydroxyapatite, *Sci. Rep.*, 6 (2016) 1-8.
- Herrfinez-Barrales E., Granados-Correa F., Sorption of radioactive cobalt in natural Mexican clinoptilolite, *J. Radioanal. Nucl. Chem.*, 242(1) (1999) 111-114.
- Myers M.J., Lavender J. P., de Oliveira, J. B., Maseri, A., A simplified method of quantitating organ uptake using a gamma camera, *Br. J. Radiol.*, 54(648) (1981) 1062–1067.
- Munthali M.W., Johan E., Aono H., Matsue N., Cs⁺ and Sr²⁺ adsorption selectivity of zeolites in relation to radioactive decontamination, *J. Asian Ceram. Soc.*, 3(3) (2015) 245-250.
- Yeritsyan H., Sahakyan A., Harutyunyan V., Nikoghosyan S., Hakhverdyan E., Grigoryan N., Hovhannisyan A., Atoyan V., Keheyanyan Y., Rhodes C., Radiation-modified natural zeolites for cleaning liquid nuclear waste (irradiation against radioactivity), *Sci. Rep.*, 3 (2013) 2900.
- Frising T., Leflaive P., Extraframework cation distributions in X and Y faujasite zeolites: a review, *Microporous Mesoporous Mater.*, 114(1–3) (2008) 27–63.
- Çicek E., Cojocar C., Zakrzewska-Trznadel G., Harasimowicz M., Miskiewicz A., Response surface methodology for the modeling of ^{85}Sr adsorption on zeolite 3A and pumice, *Environ. Technol.*, 33(1) (2012) 51–59.
- Çicek E., Cojocar C., Zakrzewska-Trznadel G., Jaworska A., Harasimowicz M., Response surface methodology for cobalt removal from aqua solutions using Isparta pumice and zeolite 4A adsorbents, *Nukleonika* 53(S2) (2008) 121-128.
- Çicek E., Response surface methodology for cobalt removal from aqua solutions using nevşehir and kayseri pumice adsorbents, *Asian J. Chem.*, 21(7) (2009) 5727-5736.
- Khayet M., Cojocar V., Zakrzewska-Trznadel G., Response surface modelling and optimization in pervaporation, *J. Membr. Sci.*, 321 (2008) 272–283.
- Cojocar C., Macoveanu M., Modeling and Optimization of Diesel Oil Spill Removal from Water Surface Using Shredded Strips of Polypropylene as the Sorbent, *Environ. Eng. Manage. J.*, 2(2) (2003) 145-154.
- Le M.H., Behera S.K., Park H.S., Optimization of operational parameters for ethanol production from Korean food waste leachate, *Int J Environ Sci Te*, 7 (2010) 157–164.
- Chauhan B., Gupta R., Application of statistical experimental design for optimization of alkaline protease production from *Bacillus* sp. RGR-14, *Process Biochem.*, 39(12) (2004) 2115–2122.