REMOVAL OF ^{226}Ra FROM WASTE WATERS RESULTED FROM MINING AND PROCESSING OF URANIUM ORES,WITH NATURAL ZEOLITES

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ABSTRACT

The zeolites were of clinoptilolyte type and the used waste waters were: mine drainage waters, waste waters from radiometric sorting and tailings pond waters from a uranium processing plant.

On the basis of experimental results, the parameters of the procedure have been established, wich enables one to obtain a radium removal efficiency higher than 95% at the passage of over 2,000 BEV waste water.

INTRODUCTION

The rasearch was done on laboratory scale and was started to find out an efficient procedure for removal of ^{226}Ra from industrial effluents resulted from exploitation and processing of uranium ores.

For type of zeolitic tuffs from three autochthonous deposits were tested. The zeolite occuring in these deposits is of clinoptilolyte type.

METHODOLOGY

The experiments were done in two steps: in the first step were used synthetic ^{226}Ra solutions to obtain information regarding the removal of the radionuclide by natural zeolites and in the second step using real waste waters resulted from uranium industry.

The experiments were conducted at ambient temperature and atmospheric presure, in dynamics, the ^{226}Ra solutions being passed through sorption columns. The diameter of columns was 10mm, the sorption bed being made up by zeolitic tuff grains of 1-2mm in diameter, previously washed out to remove the fine particles of tuff.

The effluents of columns were collected in fractions having different volumes, depending on the specificity of the experiment. The fractions were analysed for their ^{226}Ra content, being measured the remanent activity of the radionuclide. The radionuclide was measured using its daughter ^{222}Rn.

RESULTS AND DISCUSSION

The experiments were conducted simultanously using the four type of zeolitic tuffs with clinoptilolyte contents ranging between 64 and 77%. The behaviour of the four tuffs was very similar, so that only the obtained average values will be reported.

Removal of radium 226 from synthetic solutions.

Synthetic ^{226}Ra solutions used in the first step of the experiments were prepared by contamination of tap water by Ra Br_2 solution with known activity concentration.

The experiments proved that in the same geometry of the sorbent bed(height-diameter ratio:h/d) the removal efficiency depends on the specific lauding(burden) of the column and implicity on the contact time between the liquid phase and the sorbent. So, passing a solution having 3.7 Bq^{226}Ra/L through a sorption column with h/d=8.4, at different specific burdens were obtained the following results:

Specific burden [BEV/h]	Remanet activity [10^{-3}Bq/L]	Removal efficiency [%]
10	18	99.5
15	18	99.5
20	55	98.5
25	100	97.3
30	174	95.3

It can be noted the very elevated removal efficiency of radium by zeolites, in the conditions when the volume of radium solution passed through the columns was 120 BEV(Bed Equivalent Volume).

The dependance of removal efficiency on the contact time between the liquid phase and the sorbent is shown in Figure1. The optimum time in experimental conditions is of 4 minutes corresponding to a specific burden of 15 BEV/h.

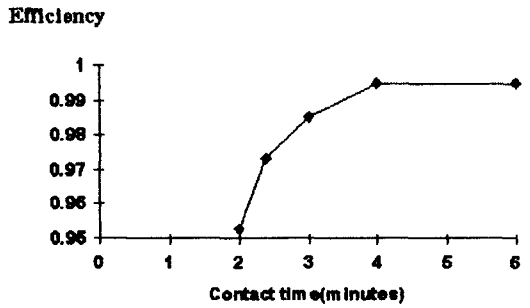

Figure 1. The relationship between removal efficiency and contact time

At this specific burden of 15 BEV/h, synthetic solutions having 0.92, 3.7 and 37Bq.^{226}Ra/L, respectively, volumes of 160 BEV of each were passed through sorption columns having the ratio h/d = 16.8.

The average remanent activities were $7\cdot 10^{-3}$, $18\cdot 10^{-3}$ and $451\cdot 10^{-3}$ Bq^{226}Ra/L, respectively, the corresponding removal efficiencies being 99.2,99.5 and 98.8%, respectively.Thus, one can note that for a large range of the activity concentrations, the removal efficiency has very high value.

The geometry of sorption columns proved to be unimportant.Thus, at the passing of 120 BEV radium solution of 3.7Bq/L through columns having h/d = 4.2, 8.2, 16.8, and 33.6, at the specific burden of 15 BEV/h were obtained similar efficiencies(99-99,5%).

These encouraging results obtained for the removal of ^{226}Ra from synthetic solutions using columns fieled up with natural zeolites, induced the attempt of using of the same sorbent bed in more cycle after the elution of retained radium. In a test, the same sorbent bed was used in 4 cycle of sorption-desorption , the elution of radium being made using a 5%NH_4NO_3 solution. The specific burdens were 15 BEV/h at sorption and 5 BEV/h at elution, the volumes being 100 BEV for the 3.7 Bq^{226}Ra/L solution and 20 BEV for the NH_4NO_3 one. In these conditions, even in the fourth cycle, the removal efficiency was over 99%.

<u>Removal of radium226 from true waste waters.</u>

Three uranium industry waste waters were used :mine drainage water, water resulted from radiometric sorting of uranium ore, and effluent resulted from processing of uranium ores.

Due to fact that the activty concentrations the three industrial waters were very low(0.19, 0.56 and 0.17 Bq ^{226}Ra/L, respectively), the sorption test were made using industrial effluent contaminated with 3.7Bq^{226}Ra/L. So, waste waters having an actual chemical composition and an appropriate activity concentration of radium were obtained. The waste water volumes pased through columns were at least 160 BEV. The experimental results are shown in Table 1.

From the data shown in Table 1 it can be seen that the chemical composition of waste waters has had a negative effect on the removal efficiency of radium, this being the most obvious for the waste water from uranium ore processing. Also, it can be noted that the removal efficiency is not more independent of the columns geometry as for synthetic radium solutions, the decrease of h/d ratio being folowed, generally, by the decrease of removal efficiency.

The detrimental effect of chemical composition of waste waters can be counteract using serial columns, the enhancement of removal efficiency being exactly in the case of processing waste water for which the chemical composition has had the most detrimental effect upon the removal efficiency. Using the same sorbent volume in two columns is more favourable, as for removal efficiency, in comparison to one column.

It can be noted that using the serial column procedure, even at a ratio h/d = 3, the remanent activity concentrations are low, the removal efficiency being over 95% even at specific burdens of 20 BEV/h.

An experiment , using two serial columns with h/d = 3, in which were passed through 2,320 BEV of uranium ore processing waste water, fieled of remanent activity concentration of 0.19 Bq^{226}Ra/L. Taking into account that in actual Romanian uranium industry waste waters can not be find such high ^{226}Ra activity concentrations, it can be said that in the case of uranium processing effluents it can be achieved an appropriate decontamination using two serial columns (h/d = 3), being able to pass through at least a volume of 2,500 BEV of waste water. These parameters are more assuring for radiometric sorting and mine drainage waters.

Removal of radium from waste waters

Table 1

Column characteristics				Water source					
Number of columns	Sorbent volume (cm^3)	$^h/_d$	Specific burden (BEV/h)	M.D.[1] (3.89Bq/L)		R.S.[2] (4.62Bq/L)		P.P[3] (3.87Bq/L)	
				RAC*	RE**	RAC	RE	RAC	RE
1	10	12	15	0.07	98.2	0.11	97.4	0.39	89.9
			20	0.07	98.2	-	-	0.59	84.7
1	5	6	15	0.14	96.4	0.17	95.7	0.31	92.0
			20	0.17	95.6	-	-	0.39	89.9
1	2.5	3	15	0.36	90.7	0.34	92.0	0.51	86.8
			20	0.34	91.2	-	-	0.76	80.3
2	2x5	6	15	0.06	98.5	0.10	97.6	0.08	97.9
			20	0.05	98.7	-	-	0.12	96.9
2	2x2.5	3	15	0.12	96.9	0.07	98.4	0.12	96.9
			20	0.08	97.9	0.08	98.1	0.16	95.8

1) Mine drainage
2) Radiometric sorting
3) Processing plant

* Remanent activity concentration (Bq/L)
** Removal efficiency (%)

CONCLUSIONS

- Clinoptilolyte zeolitic tuffs have a very high affinity for ^{226}Ra, showing high removal efficiencies;
- The chemical composition of waste water can have a detrimental effect upon the removal efficiency of ^{226}Ra by natural zeolites;
- The inhibitory effect of waste water chemical composition can be counteracted using serial columns;
- Natural zeolite column can be used, if it is appropriate, in more sorption-desorption cycles.

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