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RADIOCHEMICAL SEPARATION OF THORIUM BY ISOTOPE ION EXCHANGE*

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The aim of the present work is to obtain the separation of ^{233}Th from the radioisotopes formed in the irradiation of Mn, U, Ba, Cs, Co and the lanthanide elements with thermal neutrons, because they may interfere in the neutron activation analysis of Th, when the activity of ^{233}Th is used. The experiments were performed with the resin Bio-Rad AG 50W X-4 and X-8 (100–200 mesh) in the thorium form. The separation of ^{233}Th from the interfering radioisotopes is based on the retention of ^{233}Th by the resin (isotope exchange) and the elution of the interfering radioisotopes with a dilute solution of Th in 0.5M HCl. Batch experiments were made in order to determine the equilibrium time for the isotopic ion exchange of thorium and also the distribution coefficients of the interfering elements between the solution and the resin. Column experiments were carried out with the purpose of establishing the conditions that allow the maximum isotope exchange of ^{233}Th and the minimum retention of the interfering radioisotopes in the resin. With this purpose, a statistical interpretation of a four variable experimental design is presented.

Introduction

The determination of thorium by neutron activation analysis is usually carried out by measuring the gamma-ray activity of ^{233}Pa, without chemical separation. When microamounts of thorium are concerned, the determination requires a long irradiation time and, often, a waiting time to allow the decay of the matrix activity. This procedure is usually employed because the direct determination of thorium, by measuring the ^{233}Th gamma-ray activity, is limited by the low intensity of the total absorption peaks of its gamma-ray spectrum. Depending on the analysis sensitivity required, the use of NaI(Tl) detectors may be indispensable. However,

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as a consequence of the poor resolution of these detectors, the 86 keV main peak of the ^{233}Th gamma-ray spectrum undergoes interference from several radioisotopes that may be formed during sample irradiation. When this happens, a fast radiochemical separation of thorium is required, in view of the short half-life of the ^{233}Th (22.3 min).

TERA and MORRISON[1] and PETERS and DEL FIORE[2] proposed the isotopic ion exchange technique for many radiochemical separations and noted the reliability for short half-life radioisotope separations. As stated by TERA and MORRISON,[1] the technique presents different applications, according to the ion exchange selectivity of the elements concerned. One of them is based on the retention of radioisotope M* by an ion exchanger in the R – M form due to an isotope exchange reaction, whenever the element M has a high ion exchange selectivity. An interfering radioisotope T*, from an element T with a lower ion exchange selectivity than M, will be easily eluted from the exchanger by a solution of M. The steps pointed out by the above authors[1,2] for the separation of M* from T* are

Loading step:

$$R - M + M^* \rightleftarrows R - M^* + M \quad \text{(isotope exchange)} \quad \text{(I)}$$
$$R - M + T^* \rightleftarrows R - T^* + M \quad \text{(ion exchange)} \quad \text{(II)}$$

Elution step:

$$R - M^* + M \rightleftarrows R - M + M^* \quad \text{(isotope exchange)} \quad \text{(III)}$$
$$R - T^* + M \rightleftarrows R - M + T^* \quad \text{(ion exchange)} \quad \text{(IV)}$$

The great ion exchange selectivity of M helps the isotope exchange [Equilibrium(I)]. If the number of gram-equivalents of M in the liquid and resin phases are N_1 and N_2, respectively, the N_1/N_2 ratio must be small in order to force Equilibrium(I) to the right. In order to decrease the sorption of T by the exchanger [Equilibrium(II)], the concentration of M in the liquid phase must be high. As the operation purpose is the separation of M* from T*, the concentration of M in the liquid phase is critical. Consequently, there must be a compromise in the choice of the concentration of M in the liquid phase in order to obtain the maximum separation between the radioisotopes of interest.

In order to facilitate the experiments, as far as the half-life of the tracer is concerned, all the experiments were performed with ^{234}Th (24.1 d) and the best conditions for the maximum separation between ^{234}Th and $^{152-154}$Eu were established. Europium was chosen because the lanthanides have the highest ion

exchange affinity after thorium.[3] It was proved experimentally that ^{233}Th and ^{234}Th show the same ion exchange behaviour in the conditions described.

With the purpose of establishing the conditions that would allow the best separation between ^{234}Th and $^{152-154}$Eu, the effect of the following variables was investigated:

(a) Th concentration in the liquid phase;

(b) HCl concentration in the liquid phase;

(c) Concentration of some interfering ions in the liquid phase;

(d) Flow rate of the liquid phase through the exchanger;

(e) The exchanger cross-linkage.

A hydrochloric acid medium was employed, because it is suitable for the Th^{4+} ion exchange separation from almost all the other cations. According to STRELOW,[3] the distribution coefficient for Th^{4+}, when using Bio-Rad AG 50W X–8 cation exchanger, is the highest, except for ZrO^{2+} ions.

In order to decide which elements must be regarded as interfering, the following factors were taken into account:

(a) the radioisotopes with gamma-ray energies between 50 and 130 keV;

(b) the elements that, due to their nuclear characteristics, induce high activity in the matrix;

(c) the affinity of the elements for the resin, according to values of the distribution coefficients, determined by STRELOW.[3]

Consequently, the behaviour of: Eu^{3+}, La^{3+}, Yb^{3+}, UO_2^{2+}, Co^{2+}, Mn^{2+}, ZrO^{2+}, Ba^{2+}, Cs^{+} and Fe^{3+} was studied.

Experimental

Reagents and tracers

All reagents used were of analytical grade.

Bio Rad AG 50W (X4 and X8) cation exchange resin was employed after purification with concentrated HCl and deionized water.

^{234}Th was obtained by percolating a uranyl nitrate 0.3M HF solution through aluminium oxide, as described by ABRÃO.[4] All the other radioactive tracers were produced by irradiating the oxides of the respective stable elements.

A solution containing all the interfering elements was prepared by dissolving their carbonates or chlorides in 2M HCl. The final solution (T solution) contained 200 μg/ml of each element in 1M HCl.

Apparatus

A Nuclear Chicago single channel analyzer coupled to a 3.7 cm X 6.2 cm well-type NaI(Tl) crystal was used for the measurements of the activity, when only one radioactive tracer was involved. When interfering activities were present, a TMC 400 channel analyzer coupled to a 7.5 cm X 7.5 cm well type NaI(Tl) crystal was used.

An automatic fraction collector "Fractomat-Buchler" was employed to collect the column effluent.

The apparatus used for the radioisotope separation consisted of a glass column (5.0 cm long and 0.5 cm diameter) fitted with a small glasswool plug at the bottom. The column was joined to a glass vessel (60 ml) where the testing and washing solutions, one after the other, were placed.

Resin saturation with thorium

The two resin types (X–4 and X–8) were transformed to thorium form by percolating a 50 mg/ml $ThCl_4$ solution through the column. When the saturation was reached, the resins were washed with deionized water until a negative test for thorium was obtained in the effluent.

Batch experiments

A portion of the resins (X–4 and X–8) in the thorium form was dried at about 80 °C for a period of about 24 hours and then 0.5 g was taken for each batch experiment. The resin and 10 ml of a 0.5N HCl solution containing a known thorium concentration and a known activity of radioactive tracer (M* or T*) were added to a 50 ml Erlenmeyer flask. The time required for reaching the equilibrium between the phases was established by shaking the flasks for periods of 5 minutes to 2 hours. It was verified that ion exchange is a slower process than isotope exchange whose equilibrium was achieved in 25 minutes. The activity of appropriate aliquots from all solutions was then measured after a shaking time of 30 minutes and the distribution coefficients were calculated by means of the following equations:

$$D_T = \frac{|T^*|_R}{|T^*|_S} \quad \text{and} \quad D_M = \frac{|M^*|_R}{|M^*|_S} = \frac{|M|_R}{|M|_S}$$

where the subscripts R and S refer to the resin and solution, respectively.

In order to verify the variation of the distribution coefficients with the change of the concentration of thorium in the liquid phase, experiments with thorium solutions from 0.10 to 0.75 mg/ml were performed. The distribution of ^{234}Th in the system was calculated, for the equilibrium conditions, from the thorium content in the phases.

Although the distribution coefficient is commonly defined for the equilibrium conditions, the same nomenclature to state the distribution of a generic radioisotope

Table 1
Values of the distribution coefficient for thorium and interfering elements as a function of the thorium concentration in the liquid phase. Liquid phase: 5 ml of 0.5M HCl; exchanger: 500 mg of R – Th (4 or 8% of DVB)

Resin % of DVB	Th, mg/ml	Distribution coefficient								
		Th	U	La	Eu	Yb	Ba	Co	Mn	Cs
4	0.10	2240	4	57	44	30	12	4	5	7
	0.25	900	2	38	30	22	11	2	4	5
	0.50	448	2	33	26	17	9	2	3	4
	0.75	298	2	32	24	16	8	2	3	3
8	0.10	2180	3	14	14	12	10	3	2	2
	0.25	872	2	11	12	10	9	2	2	2
	0.50	436	2	10	10	9	8	2	2	2
	0.75	290	2	9	9	8	7	2	2	2

Table 2
Values of the separation factors (α) for the interfering elements as a function of the thorium concentration in the liquid phase. Liquid phase: 5 ml of 0.5M HCl; exchanger: 500 mg of R – Th (4 or 8% DVB)

Resin % of DVB	Th, mg/ml	Separation factor (α)							
		U	La	Eu	Yb	Ba	Co	Mn	Cs
4	0.10	560	39	51	75	187	560	448	320
	0.25	450	24	30	41	82	450	225	180
	0.50	224	14	17	26	50	225	150	113
	0.75	150	9	13	19	38	150	100	100
8	0.10	727	156	156	182	218	727	1090	1090
	0.25	435	79	73	87	97	435	435	435
	0.50	218	44	44	48	54	218	218	218
	0.75	145	32	32	36	41	145	145	145

T*, after 30 minutes of stirring, was used, even though the equilibrium had not been reached.

Table 1 presents the values of D found experimentally and Table 2 the separation factors α, calculated by means of the distribution coefficients obtained under the same experimental conditions

$$\alpha_{Th/T} = \frac{D_{Th}}{D_T}.$$

Column experiment

One milliliter of the resin saturated with thorium was introduced into the column and a little glasswool plug was placed at the top. In order to maintain the equilibrium between the exchanger and the liquid phase, a $ThCl_4$ solution, with the same HCl and Th concentrations as the sample solution, was percolated through the column. By means of this procedure, the ^{232}Th daughters that may have grown during the storage were almost completely eluted, as may be seen in Fig. 1. When the eluate and the influent solution presented the same thorium concentration, it was assumed that equilibrium was reached. The concentration of thorium was measured by activation analysis through the ^{233}Th activity. The test solutions were prepared

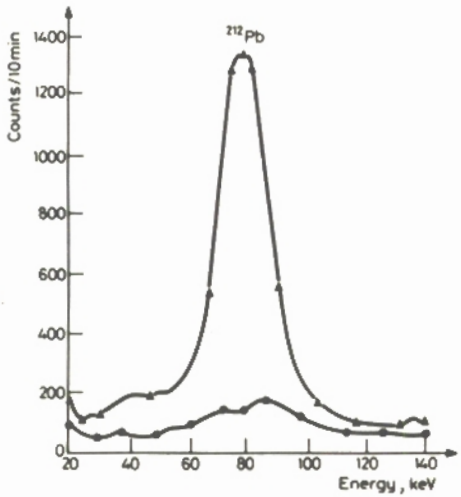

Fig. 1. Gamma ray spectrum of the ^{232}Th daughters in the thorium form resin; • counting immediately after washing, ▲ 23 days after washing

by mixing a convenient volume of the T solution with the radioactive tracer (M* or T*). After drying, the residues were taken with 5 ml of the $ThCl_4$ solution to be used in subsequent experiments. The test solutions so obtained were percolated through the column until the top of the exchanger was reached. Five ml of the washing solution were immediately percolated to remove any residue that might have been left behind. The remaining 45 ml of the washing solution were percolated directly afterwards. The washing solution had the same HCl and Th concentrations as the test solution. Whenever it was necessary to increase the flow rate in the column, air pressure was applied. Two milliliter fractions of the eluate solution were collected and the activity was measured by gamma counting. The resin was quantitatively transferred into a polyethylene tube and the activity was also measured by gamma counting.

Selection of the HCl, Th and interfering ion concentration in the liquid phase

Europium was chosen as representative of the interfering ions.

In order to obtain the best separation between ^{234}Th and $^{152-154}Eu$, the suitable HCl, Th and interfering ion concentrations were selected from previous experiments carried out with the same resin (X–4 or X–8) and by keeping the liquid flow rate constant through the column (2 ml/min). Each experiment was executed twice in order to study ^{234}Th sorption by the exchanger and $^{152-154}Eu$ elution individually.

The best concentration intervals established are shown in Table 3.

Some experiments were carried out in order to eliminate one of the five variables. It was supposed that the effect of the flow rate of the liquid phase

Table 3
Nomenclature adopted for the variables and their values

Variables	Level	
	(1)	(2)
Th concentration	A_1 = 0.25 mg/ml	A_2 = 0.50 mg/ml
HCl concentration	B_1 = 0.2M	B_2 = 0.5M
Concentration of each interfering ion	C_1 = 1.0 μg/ml	C_2 = 10.0 μg/ml
Resin cross-linking degree	D_1 = 4% DVB	D_2 = 8% DVB

through the column might be disregarded, considering that a fast separation is needed. Some experiments were executed at different flow rates (1.5; 2.0 and 2.5 ml/min), leaving all the other conditions constant. When the flow rate applied was 2.5 ml/min, a loss of ^{234}Th in the eluate solution was observed. So, a flow rate of 2.0 ml/min was chosen for all the experiments. By doing so, it was possible to make a 16-experiment design by combining the four variables.

The experiments were identified by one letter or by combinations of letters; for instance, the experiment carried out in the conditions $A_1B_2C_1D_2$ was indicated by BD. Only the variables with the highest subscripts appear in the identification. The only exception made was the experiment $A_1B_1C_1D_1$ which was indicated by (1). All the experiments were executed twice in order to obtain a good estimation of the experimental error.

Table 4 shows the values obtained for the percentages of isotope exchange of thorium and the ion exchange of europium, as well as the separation factor calculated from the average for each pair of results.

Table 4

Percentage of ^{234}Th and $^{152-154}$Eu retained by the resin and separation factors obtained in each experiment. Variables: A – thorium concentration in the solution; B – HCl concentration; C – concentration of interfering elements; D – resin cross-linking degree. Indexes 1 and 2 refer to the smallest and the greatest value, respectively

Variable				Experimental conditions	^{234}Th, %		$^{152-154}$Eu, %		Separation factor
A_1	B_1	C_1	D_1	(1)	99.7	98.1	5.4	4.9	19
			D_2	D	99.9	99.9	10.1	11.7	9
		C_2	D_1	C	100.3	99.4	5.7	7.4	15
			D_2	CD	99.8	99.5	10.7	14.7	8
	B_2	C_1	D_1	B	99.8	100.1	2.1	1.9	50
			D_2	BD	99.4	100.9	9.1	12.5	9.
		C_2	D_1	BC	99.6	99.1	4.6	3.3	25
			D_2	BCD	99.6	100.3	13.0	9.6	9
A_2	B_1	C_1	D_1	A	99.0	99.5	1.0	0.9	105
			D_2	AD	89.9	87.2	12.8	11.6	7
		C_2	D_1	AC	99.4	98.9	0.7	0.9	124
			D_2	ACD	89.6	83.9	11.6	13.2	7
	B_2	C_1	D_1	AB	96.6	97.9	0.6	0.8	139
			D_2	ABD	99.1	97.3	7.3	8.4	12.
		C_2	D_1	ABC	99.4	98.8	0.6	0.9	132
			D_2	ABCD	96.9	98.1	9.3	8.1	11

A statistical interpretation of the results was made in order to point out the best conditions for thorium separation and also the effect of the different variables. The procedure indicated for the statistical interpretation of the results from a four-variable experimental design,[5] in which the variable have two values and each experiment is made twice, was applied.

In order to check the effect of each variable and of the combination of variables in the isotope and ion exchange, the F test at a 0.05 significance level was applied.

The results for which the experimental F value was lower than the tabulated one were used to improve the estimation of the experimental error. The experimental F values are listed on Table 5. It may be observed that all the experimental conditions listed, exception made for condition AB, contribute significantly to the separation factor.

Therefore, it may be concluded that the following conditions are the most favourable for the separation of ^{234}Th from interfering radioisotopes:

- Thorium concentration in the solution: 0.50 mg/ml;
- HCl concentration in the solution: 0.5M;
- Interfering element concentration: range: 1.0 – 10.0 μg/ml;
- Percentage of DVB in the resin: 4%.

After the most favourable conditions were established, the behaviour of ^{234}Th and interfering radioisotopes, using only one radioisotope in each experiment, was studied. The retention percentages of the elements in the resin are listed in Table 6, where three results obtained with ^{233}Th as tracer are also presented in order to demonstrate the uniformity of ^{233}Th and ^{234}Th behaviour. Two milliliter fractions

Table 5
Experimental F values and sign of the average effect caused by the variables and their combinations in the separation factors

Experimental conditions	F	Average effect
A	180	(+)
B	10.1	(+)
AB	5.1	(+)
D	337	(–)
AD	177	(–)
BD	333	(–)
ABD	6.2	(–)

$F_{0.05(1.8)} = 5.32$

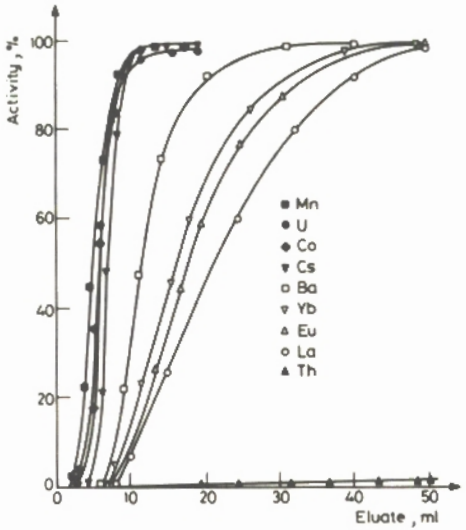

Fig. 2. Elution curves of ^{234}Th, ^{239}U, ^{140}La, $^{152-154}$Eu, ^{169}Yb, ^{139}Ba, ^{60}Co, ^{56}Mn and ^{134}Cs through the thorium saturated resin. Exchanger: 4% DVB; solution: 0.5M HCl containing 0.50 mg Th/ml and 10 μg/ml of interfering ions; flow rate: 2.0 ml/min

Table 6
Retention of ^{233}Th, ^{234}Th and interfering radioisotopes by the thorium saturated resin and value of the separation factor

Tracer	Retention on resin, %	Separation factor*
^{234}Th	98.3	
^{233}Th	98.5–97.8–99.0	
^{239}U	0.2	500
^{140}La	1.3	75
$^{152-154}$Eu	0.8	100
^{169}Yb	0.5	200
^{139}Ba	0.3	300
^{60}Co	<0.1	>1000
^{56}Mn	<0.1	>1000
^{134}Cs	<0.1	>1000

*These values represent only an of order magnitude because the contribution of the experimental error is significant. Conditions: solution: 0.5M HCl containing 0.50 mg Th/ml and 10 μg/ml of interfering ions; flow rate: 2.0 ml/min; exchanger: 4% DVB R – Th

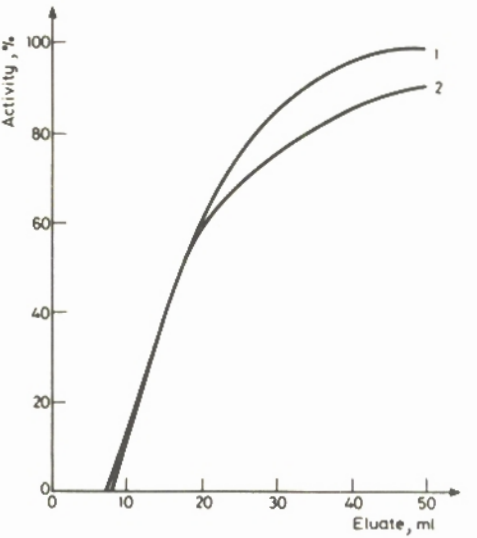

Fig. 3. Elution curves of $^{152-154}$Eu obtained with 4 and 8% of DVB. Exchanger: 4 and 8% DVB; solution: 0.5M HCl containing 0.50 mg Th/ml and 10 μg/ml of interfering ions; flow rate: 2.0 ml/min. Curve 1 – 4% DVB, curve 2 – 8% DVB

of the effluent from the exchanger were collected directly in counting tubes by means of an automatic sample collector. The elution curves are depicted in Fig. 2.

The elution of $^{152-154}$Eu through a column with a 8% DVB resin in the thorium form was studied in order to point out the difference caused by the cross-linkage of the resin on the elution curves. In Fig. 3 the elution curves for $^{152-154}$Eu through exchangers with 4 and 8% of DVB are depicted.

Verification of the influence of zirconium and iron on thorium isotope exchange

With this purpose, increasing masses of zirconium or iron and ^{234}Th were added to the loading solutions. After the usual procedure, the ^{234}Th activity retained by the exchanger was measured and compared with the known activity initially added.

Tables 7 and 8 show the values for the isotope exchange of ^{234}Th when ZrO^{2+} and Fe^{3+}, respectively, are present.

Table 7
Isotope exchange of ^{234}Th as a function of zirconium concentration in the solution

Zr, μg/ml	Retention of ^{234}Th in the resin, %
10	99.1
20	98.7
30	98.8
40	98.5
50	98.5
60	96.8
70	96.3

Table 8
Isotope exchange of ^{234}Th as a function of iron concentration in the solution

Fe, mg/ml	Retention of ^{234}Th in the resin, %
0.8	99.0
2.0	98.9
4.0	98.2
6.0	97.6
8.0	96.2
10.0	95.7

Discussion

The batch experiments were made to point out the thorium concentration in the solution for which the highest separation factors and smallest loss of ^{233}Th in the effluent were obtained. Two types of resin (4 and 8% DVB) were used to observe the cross-linking effect on the separation factors.

The results presented in Table 1 show that, when the resin with 4% DVB was used, the decontamination from the interfering ions increased rapidly until a thorium concentration of 0.25 mg/ml in the solution was achieved. This effect is less remarkable in the 8% DVB resin, where the retention of interfering ions is small. Consequently, it was concluded that the values of the separation factors (Table 2) are favoured by increasing the percentage of DVB in the resin.

It may be seen from Table 2 that the lanthanides are the most difficult elements to be separated from thorium by the proposed method, in accordance with the

distribution coefficient values determined by STRELOW.[3] Table 2 shows also that the most favourable range of thorium concentration in the solution in order to obtain the best decontamination is from 0.25 to 0.50 mg/ml

Hydrochloric acid concentrations of 0.2 and 0.5M were chosen, having in mind the values of the distribution coefficients calculated by STRELOW[3] and also because these concentrations were adopted in previous works[1,2] in which the same technique was used.

It is known that the retention of the ions by the exchanger increases as the cross-linkage percentage increases. Therefore, although the batch experiments have shown that the exchanger with the highest cross-linking improves the separation factors, column experiments with both resins, 4% and 8% of DVB, were made.

From the Equilibrium Reaction (II), it was presumed that the concentration of the interfering ions in the solution must be low. This condition is generally obtained when chemical separations are performed before sample irradiation. Therefore, it was assumed that, after the chemical separations, some micrograms of interfering elements still remain in the solution. Based on this assumption, the concentration of each interfering element in the loading solution was 5 or 50 μg/ml.

Table 4 shows the values obtained in a column experiment design, where all the possible combinations with the two values for each variable were used.

The statistical interpretation of the results, presented in Table 4, shows that the variation of thorium concentration in the solution (Experimental Condition A), affects both the isotope exchange and the ion exchange. The negative value of the average effect of this variable indicates that, with increasing thorium concentration in the solution, a decrease in ^{234}Th and $^{152-154}$Eu retention is observed. As both effects have the same sign, no indication of the effect of this variable on the separation factor may be inferred. To settle this matter, a variance analysis of the separation factor values was applied. Then, it was possible to conclude that the effect of variable A is highly significant (Table 5) and its positive sign indicates that an improvement in the separation factor is caused when the thorium concentration in the solution is increased from 0.25 to 0.50 mg/ml. Similar considerations concerning the other variables allow to conclude that the conditions which make it possible to obtain the highest separation factors are:

- thorium concentration in the solution: 0.50 mg/ml;
- HCl concentration in the solution: 0.5M;
- concentration of interfering ions: no effect between 1.0 and 10.0 μg/ml;
- 4% DVB resin.

Once the conditions that allow the best separation between ^{234}Th and $^{152-154}$Eu were established, they were applied in determining the separation factors of the elements studied in column experiments. Table 6 shows that the lowest separation

factor value was obtained for lanthanum, followed by europium and ytterbium. Among the radioisotopes produced by (n, γ) reaction from the lanthanides, ^{153}Sm is the one that interferes most strongly in the activation analysis of thorium. It may be foreseen that the separation factor for this element must be a little less than 100.

It can also be observed that the differences between results obtained with ^{233}Th and ^{234}Th, as tracers, are within the experimental error.

Figure 3 shows that europium elution is faster when a less cross-linked resin is used. This proves the greatest readiness of interfering ion elution when they are retained in a low DVB percentage resin.

After determining the ideal conditions for obtaining a high separation factor, the interferences of ZrO^{2+} and Fe^{3+} ions on ^{234}Th isotope exchange were studied. The values presented in Table 7 show that for up to 50 μg of zirconium per milliliter (250 μg in 5 ml total), the isotope exchange of ^{234}Th is virtually unaffected. As for iron, Table 8 indicates that it is tolerable up to 4.0 mg Fe/ml (20 mg total).

From the study presented in this paper it may be concluded that the method proposed for radiochemical separation of ^{233}Th is feasible for use in thorium activation analysis through ^{233}Th activity.

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