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SUMMARY

The routine determination of natural uranium content in uranyl solutions is done by applying the gamma-ray spectrometric technique. The method here described is based upon the measurement of the U-235 photopeak (185 keV), its height being compared with those obtained using standard uranyl solutions.

The uranium content of uranyl solutions whose concentrations covered the range of 1 to 500 g/l U have been determined by the outlined procedure. Uranium is determined in aqueous uranyl nitrate and sulphate as well as in organic solutions of tributyl phosphate (TBP)-varsol of various uranyl nitrate concentration. Calibration curves of 1-10, 10-100 , 100-300 and 300-500 g/l U were constructed.

The effect of thorium and its daughters present in aqueous uranyl nitrate and sulphate, for thorium concentration up to 10%, was studied, allowing to conclude that Th and its descendants do not interfere.

The determination of uranium by gamma-ray spectrometry was simplified by using a relatively new uranium, obtained by the chemical treatment of monazite sand, and precipitated as sodium diuranate (SDU) only 10 or less years ago. This raw material has its radiochemical equilibrium affected in such a way that the uranyl solutions prepared by dissolving the SDU contained no radioactive descendants that could interfere with the radiometric determination of uranium.

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INTRODUCTION

The method here described for the determination of uranium arised from the necessity of a rapid analysis, without any previous chemical separation. The method is being used routinely at the Chemical Engineering Division of the Instituto de Energia Atômica, São Paulo, since may, 1966.

With a growing analytical work and considerable number of chemical determination, and having the responsibility of the necessary controls for the operation of the pilot plants for uranium purification, the radiometric determination of the uranium content by gamma-ray spectrometry offer an excellent opportunity to accomplish a great number of analysis daily.

The radiometric determination of the uranium content of a considerably great number of uranium salt sample solutions, as here outlined, simplified profitably the analytical task of controlling the chemical steps in the uranium purification and its transformation in nuclear pure compounds. The determination is fast and made using the uranium solutions as they are handed to the analyst, that is, there is no need of any previous chemical separations and purification steps, so saving time and reagents.

The accuracy of the method is reasonably good for the worked range of uranium concentration and for the scope of the outlined procedure: a rapid analysis for the daily control of uranium content in a great number of samples.

For those analysis where best precision and accuracy are needed, the uranium determination is made using more precise methods, but these methods are, on the other hand, much laborious, require several separation steps and are time-consuming. This is the case when uranium is determined by the procedure adopted at our laboratories: extraction of uranyl nitrate with 35% TBP-varsol mixture in 3M HNO_3, scrubbing the organic phase with a 2.2M NH_4NO_3-0.8 HNO_3

solution, stripping the uranium with water, convertion to uranyl sulphate by repeatedly evaporations with H_2SO_4-$HClO_4$ acids, reduction to U-IV with stannous chloride and, finally, oxidation of uranium with ferric chloride and titration of Fe-II with $K_2Cr_2O_7$ using diphenylamine as internal indicator[1,2].

Uranium in aqueous uranyl nitrate and uranyl sulphate prepared by dissolution of crude sodium diuranate (SDU) have being analyzed by the proposed method. Some of these solutions were old enough to have U-238 in transient equilibrium with Th-234, and others solutions were prepared in such a way that the radiochemical equilibrium was broken by a partial or total separation of the U-238 daughters.

Uranium was also determined in organic mixture of TBP--varsol containing extracted uranyl nitrate, being the organic phase directly analyzed.

Generally one try to take advantage of the natural radioactivity of the U and Th series for the determination of these elements in ores. There are in the current literature several published papers advising the use of radiometric determination of uranium, of thorium, or both elements simultaneously.

Lapointe and Williamson[3], and Thommeret[4] used a "beta-gamma" method for determination of uranium; Whitham[5] used a "gamma-gamma" method for the simultaneous determination of U and Th, utilizing the natural radioisotopes Bi-214 for the uranium measurement and Bi-212 for the Th determination, applying the gamma-ray spectrometry technique.

The above mentioned methods require the sample, generally an ore, to be in secular equilibrium.

P.W. De Lange[6] combined the "beta-gamma" and "gamma--gamma" methods and developed a "beta-gamma-gamma" method for the analysis of uranium and thorium in ores, whether the ores are or

not in radiochemical equilibrium. Although the results obtained by Lange are in good agreement with the values reported by the chemical methods, this procedure has the disadvantage of requiring a rather sophisticated equipment and the calculation is time-consuming.

Once again, being mandatory the ore to be in radiochemical equilibrium, the determination will be greatly simplified when a multichannel gamma-ray analyzer is used for the resolution of the individual photopeaks of the U and Th daughters. In this way Adams[7] determined the uranium and thorium content in ores measuring the 1,76 MeV Bi-214 and 2.62 MeV Tl-208 photopeaks, although these radioisotopes have a relatively low activity, but exploring the possibility of a longer counting time.

E.A. Uken and Colab.[8] applied the radiometric method for the simultaneous determination of U and Th in ores, using a technique that require once more the samples to be in radiochemical equilibrium. So, with the mineral samples in secular equilibrium, those authors measured the 0.240 MeV Pb-212 peak for the determination of thorium, and the 0.352 MeV Pb-214 peak for the determination of uranium.

Yabubovich and Zaitseu[9] determined, by the radiometric technique, the elements U, Th, Ra and K in ores, measuring the following photopeaks: Th-234 (93 keV), Pb-212 (240 keV), Pb-214 (340 keV) and K-40 (450 keV), but with the requirement of ores to be in secular equilibrium, counting each sample during 30 minutes and solving a mathematical system of 4 equations.

Avril and Grenier[10] applied the Bi-214 gamma photopeak for the pre-concentration of uranium ores by a radiometric choice, for which it is mandatory the ore to be in secular radiochemical equilibrium.

Bourseau, Fabre and Zini[11] determined the U content in uranium ore mills effluents using the U-235 gamma photopeak, being

this peak measured with the aid of a multichannel analyzer. However, the analyzed solutions contained radioactivity due to different concentrations of Ra-226 and Th-230 radioisotopes, these radioisotopes having gamma energy close to the U-235 peak, being so serious interferences. To avoid such interferences the authors were obliged to run chemical separations through CaF_2 precipitation for elimination of Th-230 and Th-234, and $BaSO_4$ for eliminating Ra-226.

The method described in this paper determine the uranium by recording the U-235 gamma-ray photopeak (185 keV) using a single channel analyzer. With this procedure various solutions of uranyl salts were analyzed. These solutions were prepared by the chemical transformation of a relatively new sodium diuranate precipitated 10 or less years before, so the solutions had no Ra-226 and Th-230 activities sufficient high to interfere with the measurement of the U-235 peak.

THE METHOD

The radiometric determination of uranium is based upon the measurement of the height of the peak of U-235 present in the natural uranium. Natural uranium in secular equilibrium have 3 isotopes with the following percentual abundance:

U-238 - 99.28 %
U-235 - 0.71 %
U-234 - 0.0058 %

Of these three isotopes only U-235 presents the possibility of being measured through its gamma-ray emission; U-238 is a gamma emitter of 48 keV, and U-234 has a photopeak of 53 keV. Being both peaks of low energy and little abundance, they are, therefore, difficult to be measured. Consequently, the practical way to determine the uranium content by gamma-spectrometry it is through the 185 keV U-235 photopeak.

The gamma-ray spectrum of natural uranium exhibits two intense peaks of 92 and 185 keV and a third one, very weak, can be seen at 143 keV. The 92 keV peak, intensively high, is associated to natural uranium and due to Th-234 and U-235. The area under the U-235 photopeak (185 keV) or its height is measured and compared with the calibration curve constructed with uranium standards which uranium content was determined by chemical methods.

For each determination are necessary 3 millilitters of solution, pipeted to a vial that is inserted inside the well of the scintillation crystal and the spectrum is recorded. Calibration curves were prepared for determination of uranium in the following ranges: 1-10, 10-100, 100-300, and 300-500 g U/litter. With the counting equipment used, a single channel analyzer and a 2 x 2" NaI(Tl) well type crystal, the determination of uranium in solutions of less than one gram U per litter was not possible.

URANIUM

All the uranium solutions used in this work were prepared by the dissolution of a technical grade sodium diuranate (SDU). This SDU was obtained from the chemical processing of monazite sand, at the industrial plant located in S. Paulo, and operated by Administração da Produção da Monazita, for the Comissão Nacional de Energia Nuclear.

Batchs of SDU precipitated 10 or less years ago were used. This means that the raw uranium salt used had its radiochemical equilibrium disrupted and, consequently, the radioisotopes Ra-226 and Th-230 were separated from uranium. Therefore those radioisotopes are present only at so low activity that they do not interfere with the direct determination of uranium through its U-235 gamma photopeak (185 keV).

The SDU samples used had low thorium content, but it was known that the thorium concentration in the uranium raw material

can varied from 0.3 up to 8%[12]. Therefore, with the scope of to check the interference of Th-232 and its daughters in the radiometric determination of uranium, several determination of U were made using uranyl nitrate solutions to which Th was added to cover the 0.5 to 10% Th/U range.

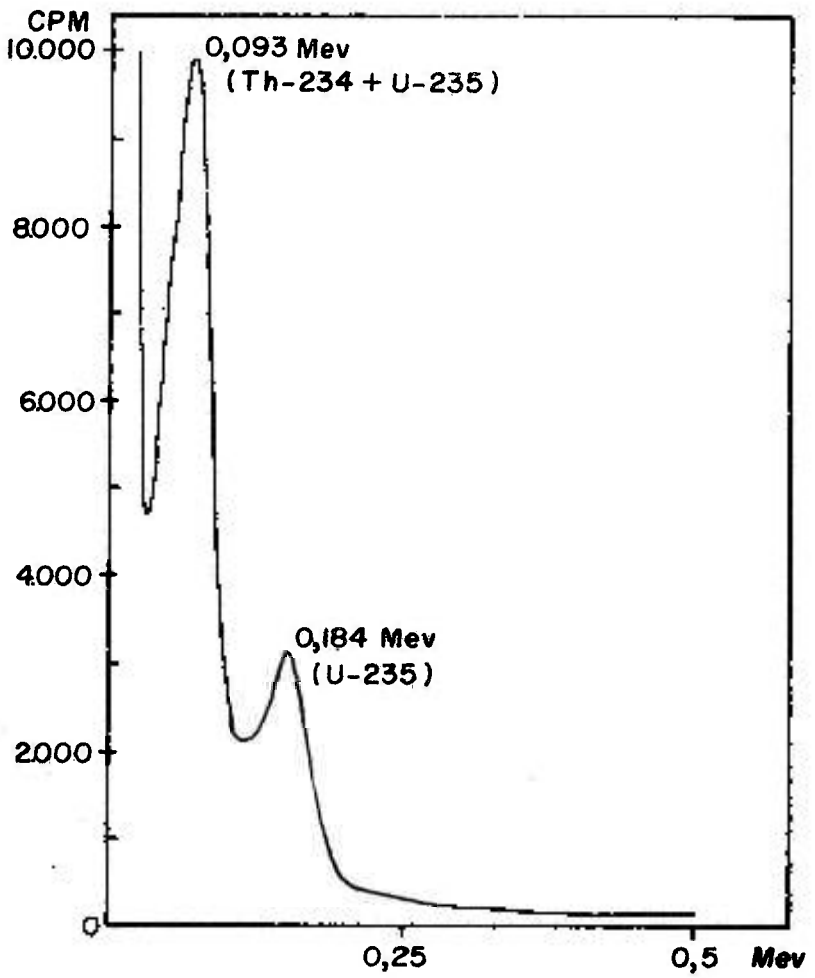


Fig. 1 Gamma ray Spectrum of an uranyl nitrate solution

Figure 1 shows the gamma-ray spectrum obtained for an uranyl nitrate solution prepared by the direct dissolution of SDU with nitric acid, without filtration or any other chemical steps that could contribute to disrupt the radiochemical equilibrium of

the sample, except any possible radioactivity loss of gaseous radioisotopes (Rn-219 and Rn-222). As can be seen the spectrum is very clean, appearing only the peaks of U-235 (185 keV) and Th-234 (93 keV). On the other hand, the spectrum recorded using an uranium sample at secular equilibrium exhibits, in the 0 - 1.0 MeV range , the following well defined peaks: Th-234 (63 and 93 keV), U-235 (185 keV) + Ra-226 (187 keV), Pb-214 (242, 352 and 610 keV). These peaks can be seen in the figure 2 obtained by scanning an euxinite sample.

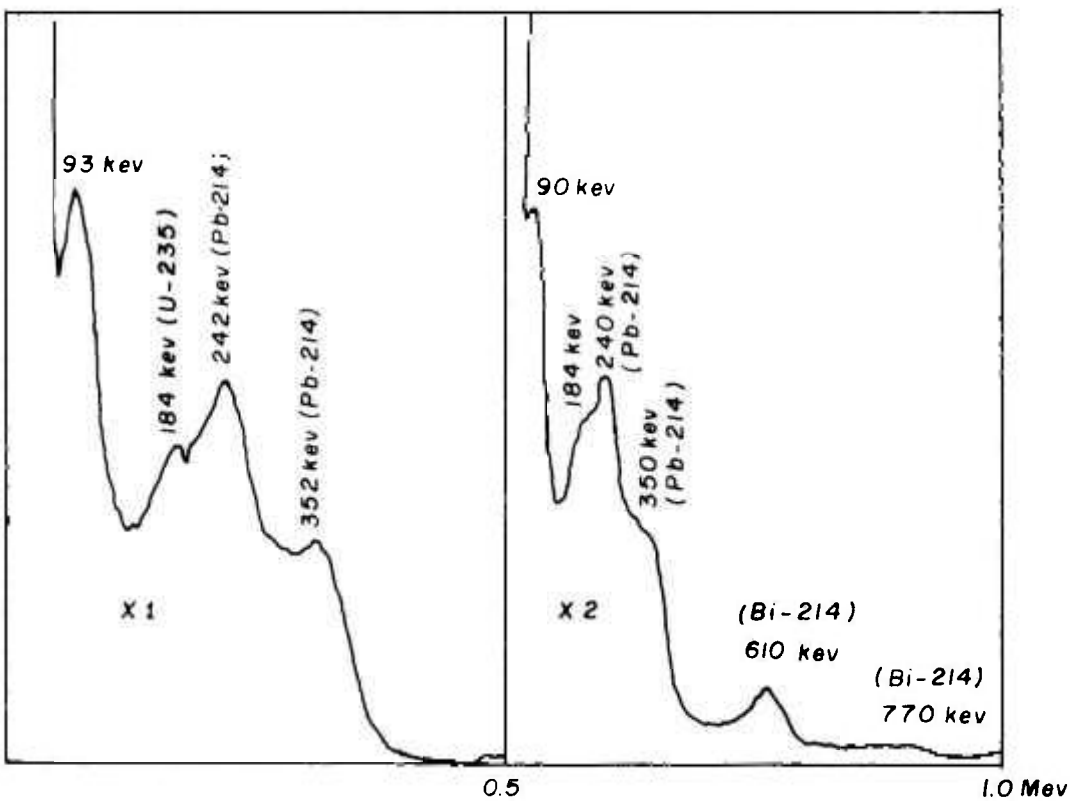


Fig. 2 Gamma spectrum of an euxenite sample (Santa Clara Farm, Pomba, Minas Gerais, Brasil).

The detection of Ra-226 was search in the uranium used in this work, looking for the three photopeaks of Pb-214 ,

descendants of Ra-226, the gamma-energy of these peaks being 242, 295 and 352 keV, respectively.

In a first tentative, 10 ml of an uranyl nitrate solution (120 g/l U), prepared by dissolution of SDU with HNO_3, were extracted with a mixture of 35% TBP-varsol, using for the extraction equalvolumes of aqueous and organic phases. The extracted aqueous phase (raffinate) was evaporated and ascertained to 3 ml and the gamma-ray spectrum was recorded (Figure 3), where only the photopeak of Th-234 (93 keV) is shown.

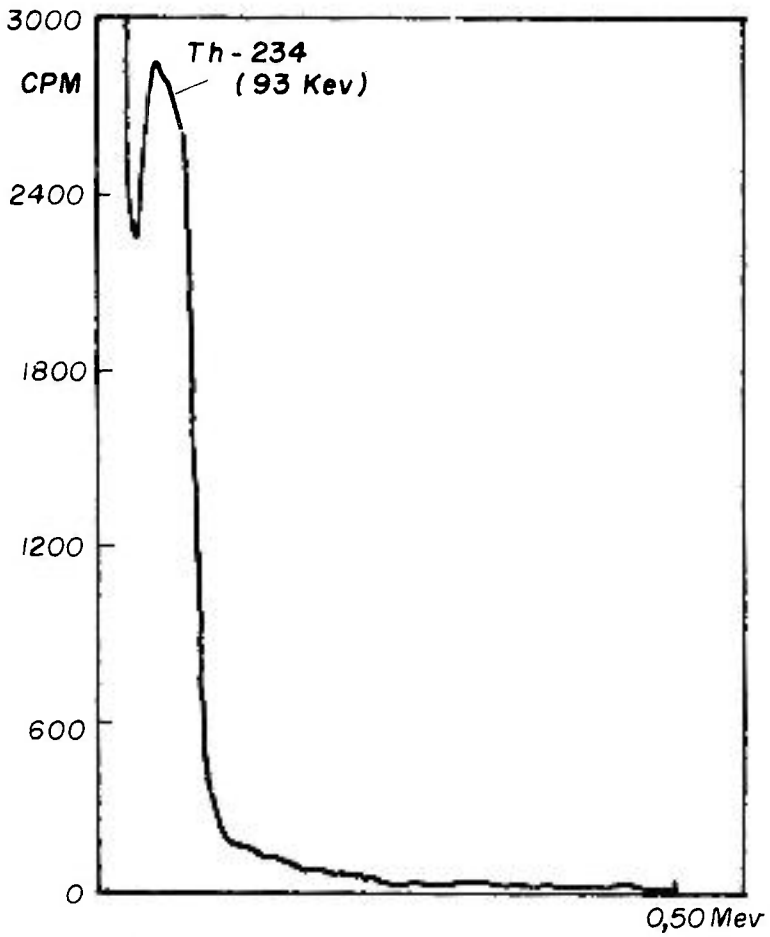


Fig. 3 Spectrum of an aqueous raffinate from TBP extraction of uranyl nitrate.

In a second trial the coprecipitation of Ra-226 with barium sulphate was experimented, using 10 ml of the same stock uranyl nitrate solution (120 g/l U). The gamma spectrum of the $BaSO_4$ was recorded covering the 0-0.5 MeV range, the scanning showing again only the 93 keV Th-234 peak, and no radioactivity due to Ra-226 (187 keV) sufficient to be detected was observed. As it is well known, barium sulphate coprecipitate efficiently radium and thorium[13].

These experiments allowed us to conclude to be negligible the amount of Ra-226 ingrown since the precipitation of SDU that occurred only about 10 or less years ago, and no activity due to Th-230 was noticed as well. Consequently, there is no interference due Ra-226 (187 keV) peak chosen for the determination of natural uranium.

URANIUM DETERMINATION

All the determinations here mentioned were runned using uranyl nitrate solutions prepared by acid solubilization of SDU. As yet mentioned, the determination was achieved through the measurement of the height of the U-235 (185 keV) gamma photopeak, using 3 ml of solution for each analysis, the solution pipeted into a plastic vial when aqueous solutions were used, and into a glass vial when organic solutions were analyzed, the last ones being TBP-varsol mixtures containing dissolved uranyl nitrate.

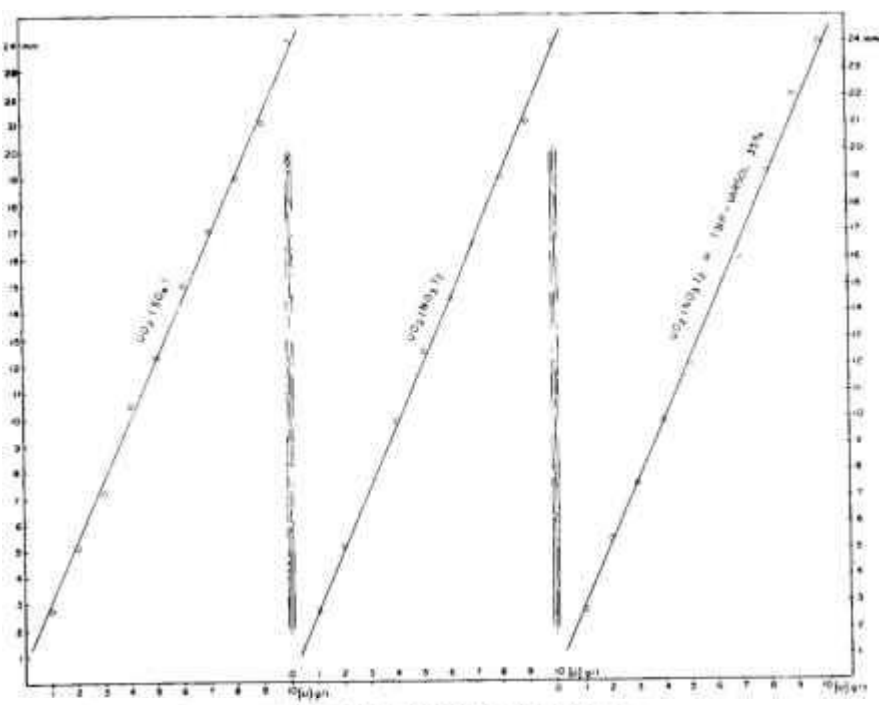
The vial containing the uranyl nitrate was inserted into the well of the 2 x 2" NaI(Tl) scintillation crystal and the gamma-ray spectrum was recorded using a single channel analyzer.

The uranium content of each solution was calculated with a calibration curve obtained using the same technique and using uranyl nitrate solutions prepared from the solubilization of uranium oxides obtained by the transformation of SDU. The uranium concentration of the solutions used for construction of the calibra

tion curve was determined by chemical methods through the following steps: solvent extraction with 35% TBP-varsol in 3M HNO_3 medium , scrubbing of the organic phase with a 2.2 NH_4NO_3-0.8M HNO_3 solution, stripping with water, transformation to uranyl sulphate with sucessive H_2SO_4-$HClO_4$ evaporations, reduction to U-IV with $SnCl_2$, reoxidation with $FeCl_3$ and titration of Fe-II with potassium dichromate and diphenylamine as internal indicator.

Calibration curves were constructed for uranyl nitrate and uranyl sulphate, covering the following ranges: 1-10, 10-100 , 100-300 and 300-500 g/l U (figures 4,5,6,7). For the determination of uranium in TBP-varsol solutions, the calibration curves were determined as for the uranyl nitrate, using several dissolutions of uranyl nitrate in the TBP-varsol mixture, being the U determined in the organic phase by chemical method after the complete stripping of the uranium.

For all the calibration curves the height of U-235 peak was used.



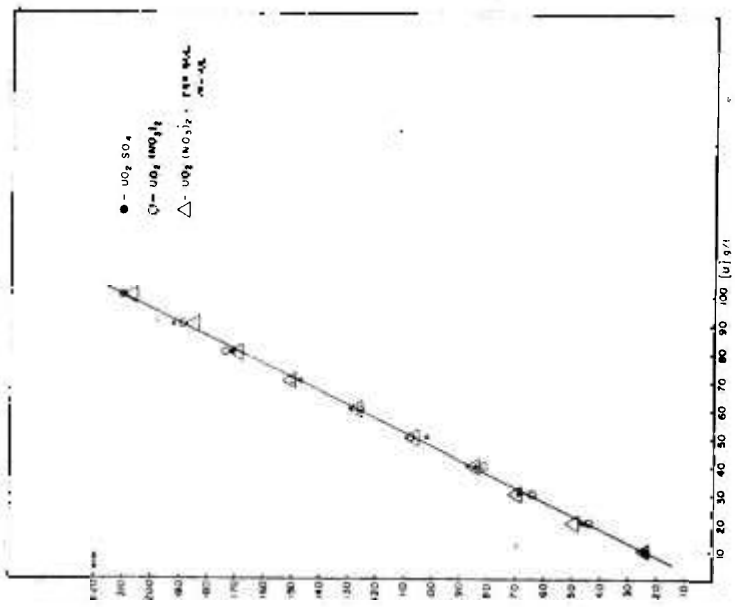


Fig. 5 Calibration curves for aqueous uranyl nitrate and uranyl sulfate, and uranyl nitrate in TBP – varsol. U-235 peak height in mm.

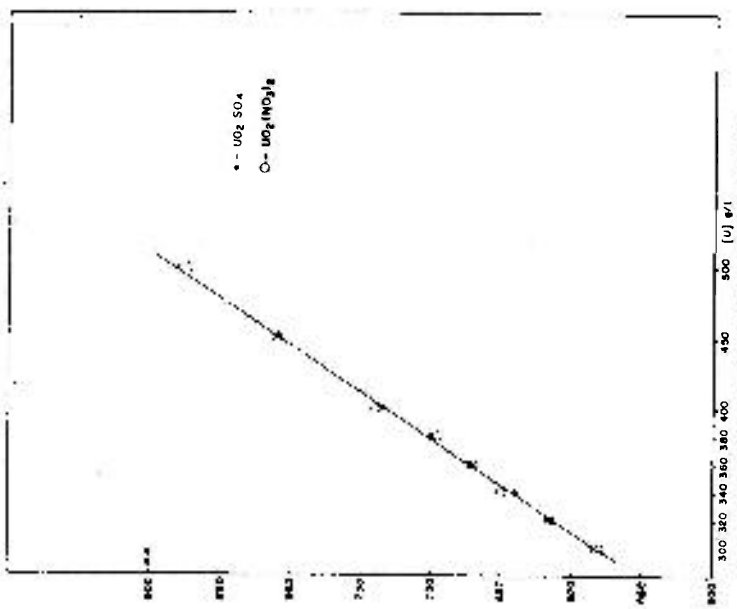


Fig. 6 Calibration curves for aqueous uranyl sulfate and uranyl nitrate. U-235 (185 kev) height in mm.

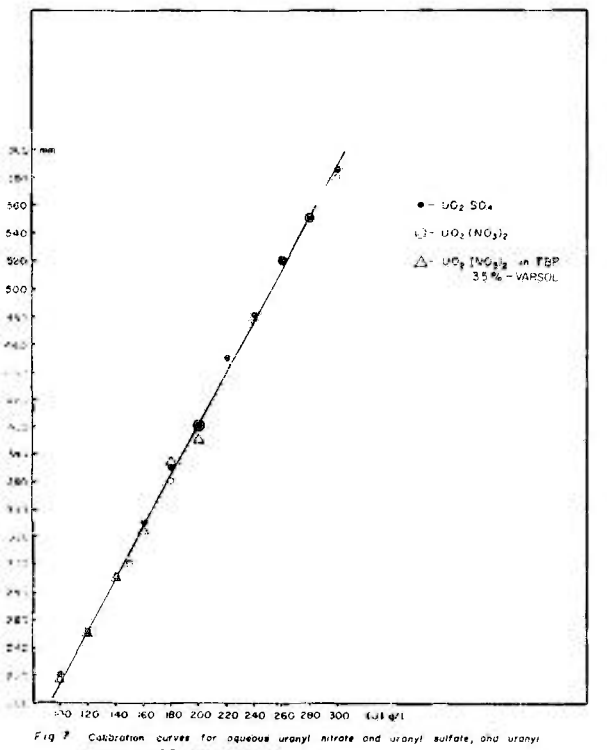


Fig 3 Calibration curves for aqueous uranyl nitrate and uranyl sulfate, and uranyl nitrate — TBP - [illegible]
U-235 peak height [illegible].

RESULTS

In Table I are some results of uranium determination by the radiometric method here described, compared with the ones obtained by chemical method.

For the radiometric method were used calibration curves previously constructed and the results were calculated by direct comparison with the height of the U-235 peak recorded after the sample spectrum, using an uranium standard of close concentration.

For solutions of concentration up to 10 g/l U the errors are as high as 7%, decreasing as the uranium concentration increase.

T A B L E I

Results of U determination by the radiometric method

Det. nr.	Chemical Method U g/l	Radiometric Method			
		by calibration curve		by close standard	
		U g/l	error	U g/l	error
1	5.50	5.4	1.8	5.9	7.2
2	8.80	9.0	2.2	-	-
3	11.60	11.5	0.9	11.3	2.5
4	13.90	14.0	0.7	14.1	1.4
5	23.20	24.0	3.4	23.5	1.3
6	27.60	29.0	5.1	28.2	2.1
7	32.60	33.5	2.7	33.2	1.8
8	46.50	47.0	1.1	46.4	0.2
9	58.44	60.0	2.6	57.9	0.9
10	65.50	63.5	3.0	62.8	4.1
11	69.90	69.0	1.2	68.6	1.8
12	71.90	73.0	1.5	-	-
13	88.80	87.0	2.0	88.7	0.1
14	112.20	110.0	1.9	112.9	0.6
15	117.60	119.0	1.2	120.0	2.0
16	174.90	178.0	1.7	178.0	1.7
17	197.80	200.0	1.1	200.0	1.1
18	335.60	332.0	1.1	-	-
19	401.10	394.4	1.6	-	-

DISCUSSION

The radiometric determination of uranium through the photopeak of U-235 (185 keV) present in the natural uranium, applied to several solutions of uranyl salts prepared by the solubilization and transformation of a relatively new SDU, precipitated approximately 10 or less years ago, can be used with advantage. As was demonstrated, the unique U-238 daughter still present with significant radioactivity it is Th-234. The radioisotopes Ra-226 and Th-230, that could interfere, do not exhibit sufficient radioactivity to contribute to the gamma-ray spectrum, since there was not enough time to regenerate them.

The method is rapid, allowing to run a great number of analyzes daily. The presence of stable elements is tolerable and do not constitute interference.

Knowing about the possibility of the presence of some thorium in the SDU, since this salt is obtained from the chemical processing of monazite sand, experiments were performed with the scope of to know whether Th-232 and its daughters behave or not as interference. For this the spectra of several uranyl nitrate solutions containing thorium at concentrations covering the range of 0.5 to 10% Th/U were recorded. This range of thorium contamination in uranium was chosen since it is known that the thorium content of SDU rangesfrom 0.3 to 8%[12]. As we can see by figures 8 and 9, it is possible to determine the U content of uranyl solutions having the indicated amounts of thorium.

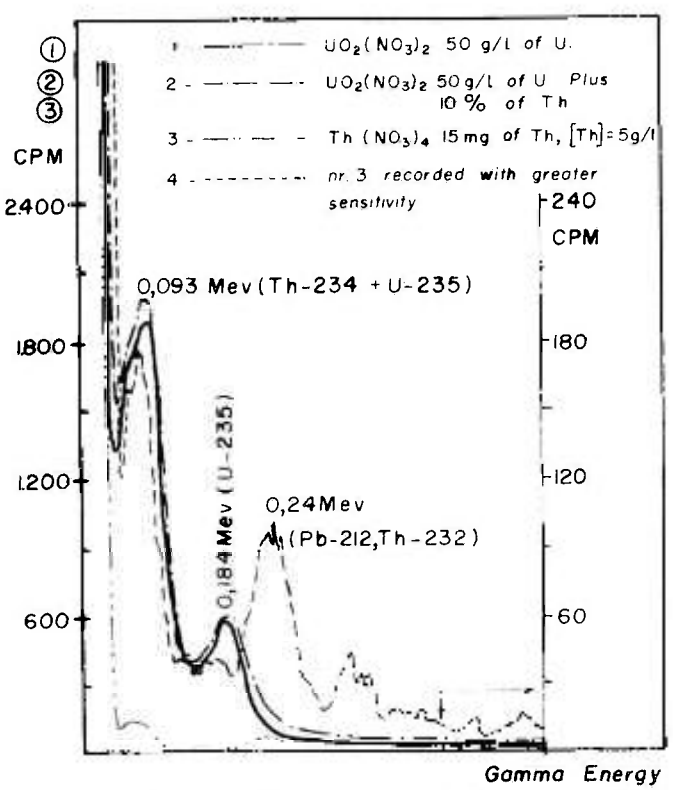


Fig. 8 U-235 peak recorded for U solutions impurified with Th-232.

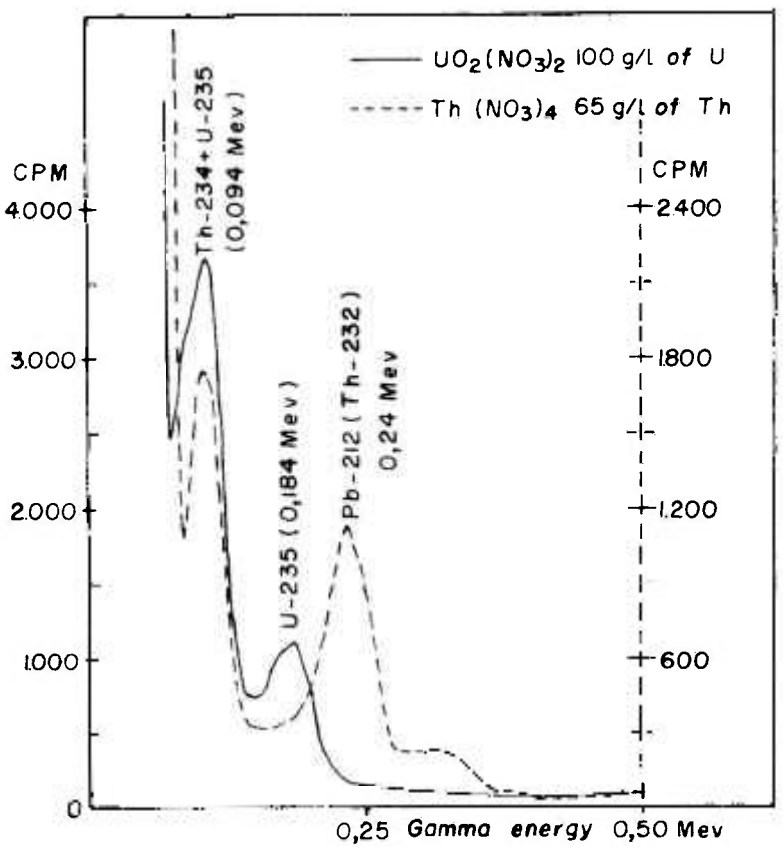


Fig. 9 Spectra of uranyl nitrate and thorium nitrate solutions

For the determination of uranium in uranyl nitrate dissolved in TBP-varsol mixtures, as was in the case of solvent extraction, the analysis is still more facilitated, since only uranium is present.

Having at ours analytical and technological laboratories, as well as at the uranium purification plants, considerable number of experiments where uranyl nitrate, uranyl sulphate, and uranyl nitrate dissolved in TBP-diluents are used, having these solutions a variable range of uranium concentration, therefore demanding a great number of U determinations, the radiometric method here put in practice permited to overcome the analyzes demand.

Although the sensitivity of the method, using a 2 x 2"

NaI(Tl) crystal, well type, and a single channel analyzer, do not make possible the determination of U concentration lower than 1 g/l, and with errors up to 7% when compared with the chemical determination of uranium, the simplicity of the radiometric method permit a rapid determination, with remarkable advantage.

An additional advantage offered by the radiometric method is the uranium solutions purification control concerning the thorium and lead. As was demonstrated, when the gamma-ray spectrum for the U analysis is recorded, it is possible also to follow the thorium behavior through the Th-234 tracer as to follow the lead separation, as well, through the Pb-212 thorium daughter. Being the Chemical Engineering Division of the Instituto de Energia Atômica involved in uranium purification and its transformation in nuclear grade products, this control of Th behavior during the purification steps is of enourmous value.

RÉSUMÉ

La determination routinière d'uranium naturel est faite en solution de sels d'uranyle, employant la technique de spectrométrie de rayons gamma. Cette méthode est basée sur la determination du photopic de l'U-235 (185 keV), dont l'hauteur est mesurée et comparée avec celles obtenues à partir de solutions échalon.

On a fait des determinations d'uranium en solutions de sels d'uranyle de concentrations de 1 à 500 grammes U/litre. On a determiné uranium en solution aqueuses de nitrate et de sulfate d'uranyle et en solutions organiques de phosphate de tributyle-varsol de diverses concentrations de nitrate d'uranyle. On a construit des courbes de calibration de 1-10, ... 10-100, 100-300 et 300-500 grammes/litre d'uranium.

On a étudié l'effet de la présence du thorium et ses descendants dans les solutions aqueuses de nitrate et de sulfate d'uranyle, en concentrations jusqu'à 10% de Th, concluant inexister d'interférence.

Dans ces études la determination d'uranim par spectrométrie gamma a été simplifiée par l'emploi d'uranium relativement neuf, obtenu par le traitement de la monazite et précipité sous forme de diuranate de sodium il y a moins de dix ans. Ainsi l'equilibre radiochi-

mique avait été rompu et les solutions de sels d'uranyle préparées par la dissolution de ce diuranate de sodium ne contenaint par des descendants radioactifs interférents.

RESUMO

A determinação rotineira de urânio natural é feita em soluções de sais de uranila usando-se a técnica de espectrometria de raios gama. O método baseia-se na determinação do fotopico do U-235 (185 keV) cuja altura é medida e comparada com as medidas obtidas usando-se soluções padrões.

Foram feitas determinações de urânio em soluções cujas concentrações em U variaram de 1 a 500 gramas por litro. Urânio foi determinado em soluções aquosas de nitrato e sulfato de uranila e em soluções orgânicas constituidas por misturas de fosfato de tributila .. (TBP)-varsol com variadas concentrações de nitrato de uranila. Foram preparadas curvas de calibração de 1-10, 10-100, 100-300 e 300-500 g/l U.

Foi estudado o efeito da presença de tório e seus descendentes nas soluções aquosas de nitrato e sulfato de uranila, para concentrações de até 10% de Th presente no urânio, concluindo-se não haver interferência.

Neste trabalho a determinação de urânio por espectrometria gama foi muito facilitada pelo fato de ter sido usado urânio relativamente nôvo, obtido pelo tratamento químico da monazita e precipitado na forma de diuranato de sódio, há aproximadamente dez anos. Assim, o equilíbrio radioquímico fôra rompido, não apresentando as soluções de sais de uranila, preparadas a partir da dissolução dêste diuranato de sódio, descendentes radioativos interferentes.

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