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The search for an elusive 4.4-MeV α emitter in uranium minerals

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THE SEARCH FOR AN ELUSIVE 4.4-MeV α EMITTER IN URANIUM MINERALS

ABSTRACT

We describe our search for an unidentified 4.4-MeV α -emitter in Belgian Congo pitchblende and uranium raffinates, and we give a history of observations of 4.4-MeV activity over the last 55 years in radiogenic haloes, zinc ores, monazite, thorite, huttonite, ultrabasic and other abyssal rocks, osmiridium, uranium ores, and raffinates of uranium. We show no evidence of excess 4.4-MeV activity in any of the chemically separated fractions investigated. We give upper limits for 4.4-MeV α activity in each of four studied samples.

INTRODUCTION

Over the last 55 years, there have been many observations of low-energy α radiation in natural samples. These samples include ultrabasic and volcanic rocks, osmiridium, thorite, monazite, uranium raffinates, ores and chemically separated fractions of zinc, and radiogenic haloes in minerals. Most of these findings have not been reproduced. In this report, we document our failure to find any evidence of the 4.4-MeV α emitter in any of our uranium minerals studied.

Since the identification by J. Joly of pleochroic haloes surrounding zircon as those arising from α decay,¹ a number of haloes have been found whose dimensions suggest α energies that do not agree with the known radioactive decay series stemming from ²³²Th, ²³⁸U, or ²³⁵U. In particular, of the reported haloes corresponding to α energies of 2.2 MeV,²⁻⁶ 3.2 MeV,²⁻⁶ 1-3.4 MeV,⁷ and 4.5 MeV,^{2,8-10} only ¹⁴⁷Sm ($t_{1/2} = 1.08 \times 10^{11}$ y; $E_{\alpha} = 2.23$ MeV) has been proposed as a long-lived natural source.⁹

A series of experimenters led by Josef Schintlmeister at the Vienna Institute of Radium Research reported seven α emitters with energies of 1.3, 1.9, 2.6, 3.3, 4.0, 4.4, and 5.0 MeV between the years 1935 and 1951.¹¹⁻¹³ These emitters were found in foundry products of Upper Silesia zinc ores, mined near Beuthen. The prospect of obtaining nuclear weapons material from fission of the 3.3-MeV α emitter¹² led to research on isolation of this activity source. Based upon an extrapolation of the energies of α emitters known in 1942, Schintlmeister identified the 3.3-MeV α activity as ²⁴⁴94, or eka os-

mium, as it was known in the periodic tables in use during World War II.

Identification by the same method of the 4.4-MeV α emitter as ²⁴⁰94 seemed to agree well with the fact that it could not be chemically separated from the 3.3-MeV α emitter. Only trace amounts of these two emitters were available, but their behavior with carriers in solution included precipitation from strongly acidic solutions with H₂S, insolubility with carbonates in alkali, incomplete precipitation with NH₄OH and NaOH, and volatility in distillation from oxidizing acids. Unfortunately, the raw materials and most of these results were lost during World War II, and later research failed to find these activities in ore samples from the same mine or from residues of different smelting works.^{14,15} Others have also noted the 3.3-MeV α activity associated with zinc.¹⁶⁻¹⁸

Gysae and Korsching have published α -ranges corresponding to 2.3, 3.4, and 3.8-MeV emitters¹⁹ that could be separated from each other but that displayed some common properties: no precipitation from acidic solutions with H₂S, solubility of the hydroxide in ammonium carbonate, and no precipitation with oxalic acid. A later report claimed α emitters of 3.6, 4.1, and 4.5 MeV that would precipitate with HF unless oxidized, with ammonium persulfate being an example of a suitable oxidant.²⁰ The source of these emitters was a monazite extract.

More recently, several observations have been made of α activity in the range 4.2-4.7 MeV that are inconsistent with the natural decay chains. Cherry

et al. published an α spectrum obtained from dissolved thorite separated from Conway granite that showed a peak at 4.4 MeV which could not be associated with the natural $^{235}\text{U}/^{238}\text{U}$ α activity ratio.²¹ This α activity was also detected to a lesser degree in the mineral huttonite from the same granite. These findings could not be reproduced by Petrzhak *et al.*, who separated the heavy fraction of Conway granite by sedimentation in bromoform and prepared α spectrometric sources by a cathode sputtering process, taking care not to lose any volatile oxides.²² They found no excess activity at 4.4 MeV. Gentry *et al.*²³ also failed to find evidence of the 4.4-MeV α emitter in Conway granite. They followed the same preparation techniques used by Cherry.

Cherdyn'tsev²⁴⁻³¹ has claimed the discovery of element 108 in nature, which he called sergenium. This new element displayed a complex α spectra, with the highest energy line at 4.5-4.65 MeV being the most intense. This emitter resembled Os in its chemical properties, including volatility of the oxide. On the basis of excess α activity in the region 4.2-4.7 MeV, sergenium was claimed to be present in samples of ultrabasic and other abyssal rocks, such as iron-bearing minerals, amphiboles, and certain sulfides. Plutonium-239 in amounts much higher than the naturally occurring $^{239}\text{Pu}/^{238}\text{U}$ atom ratio range^{32,33} of 10^{-12} - 10^{-11} was found to be a constant satellite of the element, and it was claimed to be a decay product. Americium-243 and an unspecified isotope capable of spontaneous fission were also claimed to be decay products of sergenium.³⁴ Poor counting statistics and ill-defined α spectra decrease the credibility of Cherdyn'tsev's claim. Others, using rocks supplied by Cherdyn'tsev, have been unable to detect eka-osmium.³⁵ Kush *et al.* demonstrated the existence of technogenic ^{237}Np in the volatile fraction of a lead-zinc concentrate that had been subjected to chemical processing normally used for the separation of osmium.³⁶ They warned against unjustified conclusions from low-energy α spectra obtained from such samples

because of the possible masking effect of technogenic neptunium.

Meier *et al.* also published spectra showing α activity at 4.3-4.6 MeV, as well as γ activity at 200-220 keV.³⁷ These spectra were obtained from chemically separated fractions of osmiridium that came from various places in Africa and Australia. Not all the details of enrichment are given, but early treatment of the ore included decomposition with NaOH and NaNO₃ in a silver crucible at 700°C, reduction with a stream of hydrogen, driving off Os and Ru by distillation with HNO₃ or H₂SO₄/NaBrO₃, purification of Ag with NH₄OH, refluxing for several days with HNO₃/HCl, precipitation at pH 7 and reduction of the solution with hydrogen. One of their methods of purification involved an ether extraction of an HNO₃ fraction in which the aqueous phase contained the unknown α emitter. A peak at 5.15 MeV in another fraction was given as evidence of ^{239}Pu occurring with the unknown. They have also reported measurements of $^{239}\text{Pu}/^{238}\text{U}$ atom ratios up to 10^{-7} in volcanic samples³⁸ in agreement with Cherdyn'tsev,²⁹ but with no indication of a 4.4-MeV α activity.

In the period 1949-1950, Peppard and Mason observed an unexplained activity at 4.42 MeV during their solvent extraction studies of the major aqueous waste stream of the Mallinckrodt Chemical Works' processing of Belgian Congo pitchblende.³⁹ During 1970, Mason again obtained the activity using a concentrate of the original raffinate. However, the activity disappeared from the separated sample within a few weeks.

G. Van de Steeg of the Kerr-McGee Corporation has described in a private communication his observation of excess α activity in the 4.4-MeV region. The excess was seen in an assay for uranium of a raffinate of uranium ore from Grants, New Mexico.

The mystery of these incomplete and inconsistent results, as well as the prospect of the discovery of a new element, have motivated us to search for the 4.4-MeV α emitter.

EXPERIMENTAL PROCEDURES

SOURCE OF MATERIALS

We obtained our sources for α spectroscopy by chemical separations that were performed on solu-

tions made from four different sources. A sample of the Mallinckrodt concentrate used by Peppard and Mason during their observations of the 4.4-MeV emitter was generously provided to us by

Dr. George Mason of Argonne National Laboratory. Samples of the Belgian Congo ore from which the Mallinckrodt concentrate was derived were provided by the Harvard museum and the Smithsonian Institute. Mound Laboratories, a division of Monsanto Research Corp., supplied a material known as Cotter concentrate—basically Mallinckrodt raffinate which had been chemically reprocessed to remove residual U, Ni, Cu, Co, Th, and Pa. Also, Van de Steeg sent samples of the uranium raffinate from the Kerr-McGee processing.

Mallinckrodt Concentrate

About 70 ml of the Mallinckrodt concentrate was made available for our experiments. The solution was viscous, yellow-green in color, and effervescent. All of the components of the original ore that were soluble in HNO_3 were present.⁴⁰ Uranium and lead were also present but were reduced to small fractions of their original amounts by the Mallinckrodt processing. The solution was 1–2M in free HNO_3 and nearly saturated in $\text{Al}(\text{NO}_3)_3$.⁴¹ The principal α activity was ^{230}Th at $\sim 10^7$ α -decays/m per 50 μl . Daughter activities from the Th and U decay chains were also present.

Belgian Congo Ore

The Belgian Congo ore used in our investigation was the source of the uranium used during World War II to produce the first man-made atomic reactor, at Stagg Field, at the University of Chicago. The 70 g of material provided by the Harvard museum was a shiny, black exfoliate that emitted about 60 mr/h β and γ radiation at contact, while the Smithsonian ore was a hard grey-green rock weighing 150 g, and emitting about the same amount of β and γ radiation. The ores contained about 5 g/ton ^{230}Th and had a $^{231}\text{Pa}/^{230}\text{Th}$ mass ratio of about 17×10^{-3} , and are extremely rich in uranium, as they contained about 38% uranium oxide.⁴⁰ Iron, calcium and the lanthanides were also present in varying amounts as the principal mass contaminants.

Cotter Concentrate

The Cotter concentrate was obtained because of its genetic relationship to the Mallinckrodt concentrate and the original Congo ore. The bulk of the raffinate from the Mallinckrodt Chemical Works' processing of the Congo pitchblende for uranium

was stored until the late 1960's at the end of an airplane runway in St. Louis, Missouri. The material came to be known as the St. Louis residues. The Cotter Corporation moved it to Colorado for processing of the residual U, Ni, Cu, Co, Th, and Pa. Di-(2-ethylhexyl) phosphoric acid (HDEHP) in kerosene was contacted with a sulfuric acid leach of the St. Louis residues to remove, with varying degrees of efficiency, the U, Th, and Pa. The solid concentrate from evaporation of the carbonate containing liquid raffinate was eventually shipped to Mound Laboratories for storage.

The Cotter concentrate powder was a red, iron-oxide color, with specks of bright yellow. It was emitting about 10 mr/h β and γ at contact. Gross activity was 4×10^9 decays/m per 1.5 kg. A considerable amount of "filter aid" constituted a major component of the material. The Cotter concentrate was of less interest than the Mallinckrodt concentrate or the Congo ore because it was only distantly related to the two previously mentioned sources.

Kerr-McGee Solution

The Kerr-McGee solution was pale yellow and clear, with pH 1–2, a high sulfate and chloride content, and containing about 5×10^5 decays/m per liter of uranium and its daughters. The solution also contained iron and the lanthanides. It was originally an oxidizing medium, although the chlorate initially present was largely decomposed when we received the solution.

PREPARATION AND ANALYSIS OF THE SAMPLE

Previously published descriptions of the 4.4-MeV α emitter are either unclear or inconsistent with respect to its half-life; however, the past observations were made following the execution of chemical procedures requiring several hours or more. Our chemical separations were likewise designed to produce samples suitable for α pulse-height analysis (PHA) within hours.

Samples were deposited on 0.051-mm thick platinum plates by either flash volatilization from a hot tungsten filament in a vacuum, evaporation of a solution under a heat lamp, or by electroplating. To produce our electroplated samples, we followed a variation of Talvitie's method which required a plating cell of glass and teflon⁴² Often, the final solution, obtained after several steps of separation

and purification, was divided, and counting plates were prepared by both electroplating and volatilization. Such double samples were a vital check on the volatility of the unknown.

The α spectroscopy of all samples prepared in this work was performed with 450-mm² surface-barrier detectors. An ND600 pulse-height analyzer coupled to an LSI-11 computer was used for data acquisition, storage and output of 1024-channel spectra. Alpha energies were determined by calibration with ²³⁷Np, ²⁴¹Am, and ²³⁹Pu sources, and internally with isotopes of the natural decay chains that were present in many samples. The region 2.8-11 MeV was analyzed for each sample. The background in the 4.4-MeV region was 0.15 counts/channel per day at the start of measurements, and this amount gradually increased with time. Depending on their activity level, samples were counted for periods ranging from less than a day up to thirteen days. Because of previous reports concerning excess ²³⁵U activity, we counted samples containing uranium until the ²³⁵U peaks contained at least 150 counts.

Figures 1 through 4 show flow charts of the separations initially used in the preparation of samples from the Mallinckrodt concentrate, Congo ore, the Cotter concentrate, and the Kerr-McGee solution, respectively. The chemical separations that we used to prepare samples from the Mallinckrodt concentrate and the Kerr-McGee solution were precisely that which Mason and Van de Steeg used in their respective observations of the unexplained 4.4-MeV α activity. The samples from the Mallinckrodt stock and Kerr-McGee stock were purified and prepared for α counting, as outlined in Figs. 1 and 4, respectively. The Congo ore was dissolved and samples from the stock solution prepared by the Mason and Van de Steeg procedures. Figure 2 shows the dissolution procedure for the Congo ore. The dissolved Cotter concentrate was subjected both to the Kerr-McGee and Peppard-Mason separation methods. A variation of these separations was also investigated in which actinide and lanthanide-like elements could be separated from any solution, as shown in Fig. 5.

The solutions made from each of the four source materials presented unique chemical problems. Uranium was the most abundant element in solutions made from the Congo ore. The Mallinckrodt concentrate primarily contained Al and the lanthanides, with thorium providing most of the α activity from this source. The Kerr-McGee

material was chiefly composed of iron and the lanthanides. A mixture of Na₂CO₃, organic material, iron, and the lanthanide elements was nearly equally abundant in the Cotter concentrate. The main components of each of these solutions were removed by an appropriate chemical separation because determination of the α energies, and therefore identification of the α emitters, was not possible in the presence of these bulk constituents.

A 15-ml aliquot of the stock solution from the Congo ore, prepared according to the procedure outlined in Fig. 2, contained 490 mg of uranium. The uranium was eluted from a Dowex-1 anion column with 0.1M HNO₃, and an aliquot was electroplated. Pulse-height analysis of the sample showed peaks attributable to ²³⁴U, ²³⁵U, and ²³⁸U. The ²³⁸U/²³⁵U α ratio of 21.3 \pm 1 was consistent with natural uranium samples. The Th and rare-earth fraction eluted from an anion-exchange column using HCl was treated according to the procedure illustrated in Fig. 5. For some samples, Ra was removed by a Ba(NO₃)₂ precipitation with red, fuming HNO₃. A major portion of the Ra was removed from other samples by incomplete extraction into di-n-octylphosphoric acid. Radium separation by either method was sufficient to produce samples in which tailing from the 4.78-MeV α from ²²⁶Ra did not obscure the 4.4-MeV energy region.

Fifty- μ l aliquots of the Mallinckrodt stock were used for chemical separations and analyses. This aliquot size was the same as that used by Mason in his 1970 investigation. Dilutions of the 50- μ l aliquots were then made to achieve 0.1M HNO₃ in order to duplicate Mason's previous work. Extractions were also made with stock solutions diluted to 0.05M and 0.2M HNO₃. Since the selectivity of thenoyltrifluoroacetone (TTA) as an extractant decreases with increasing pH, a series of experiments were performed in which the pH of the initial solution was adjusted to 2 and then back-extracted. By varying the acid concentration in extractions with TTA, we could control the distribution of radioactivity between the aqueous and organic (TTA) phase. At pH 2, TTA extracts the elements that have a valency of 3⁺ or greater. This back-extraction technique reduces the likelihood that our failures to reproduce Mason's results were due to extractions done at a different pH. Figure 6 shows the variation in extracted α activity that was found by this procedure, and this agrees with the expected behavior.⁴³ Samples taken after each back-extraction were analyzed by α spectroscopy. Each

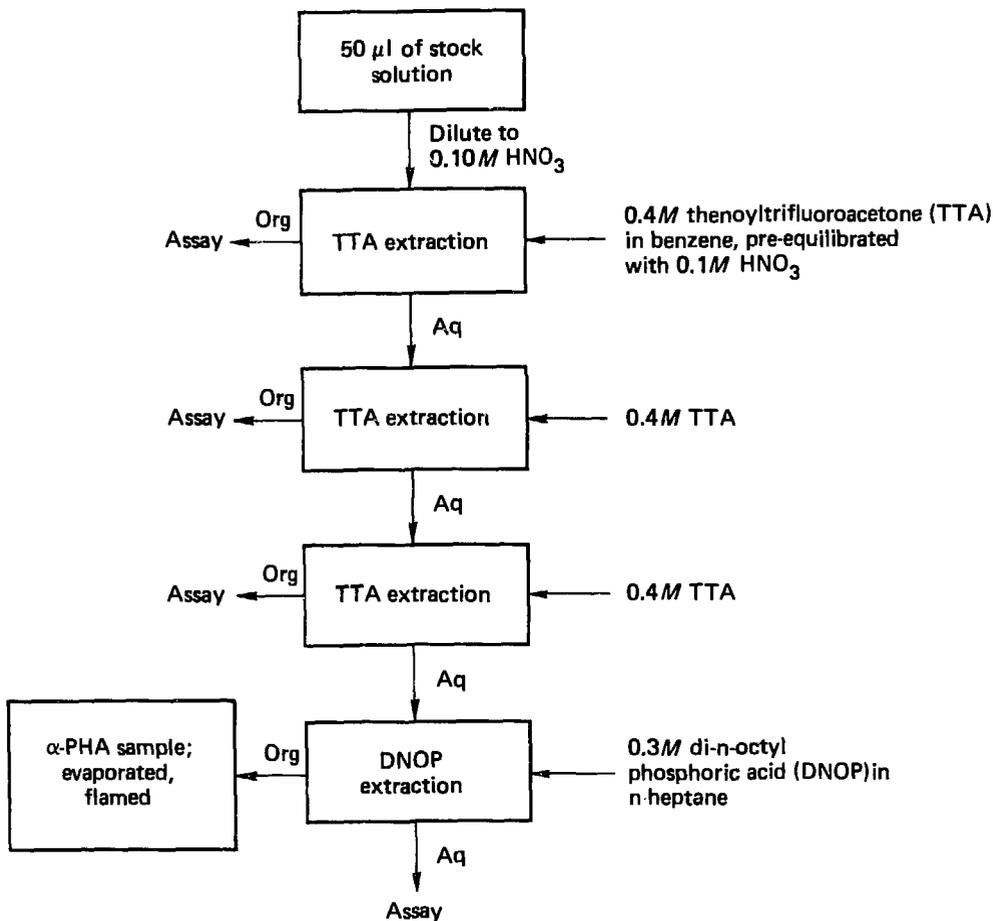


FIG. 1. Peppard-Mason separation procedure for investigating 4.4-MeV activity.

of these samples contained α peaks attributable only to Th, U, or their daughters. The TTA extractions, which Mason used in his 1970 observations, were very effective in removing Th from solution. To prevent a change in pH, the TTA was pre-equilibrated by washing the 0.4M TTA in benzene with the same acid concentration as the solution to be extracted. Table 1 shows the relative amounts found in each phase after successive extractions of the Mallinckrodt stock at pH 1.

Uranium-232 was used to determine the uranium concentration in each of the four solutions. Plutonium-236 was added to a 50- μ l sample of the Mallinckrodt concentrate to determine the $^{239}\text{Pu}/^{230}\text{Th}$ α activity ratio. The gross α count of aliquots, coupled with measured α spectra, were used to determine the yields in all other cases. Uranium yields in the Van de Steeg separations were typically 78% overall, an amount consistent with his yield.

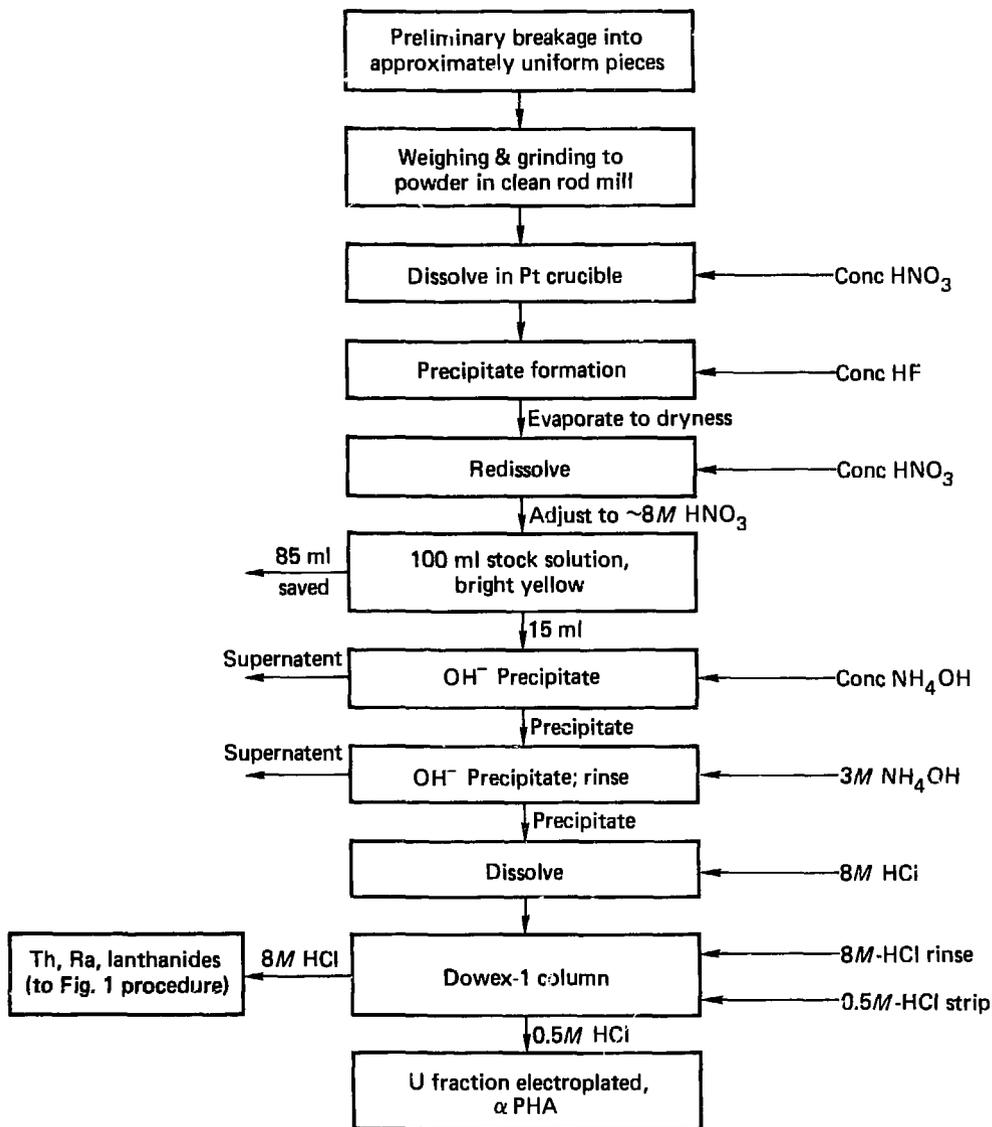


FIG. 2. Procedure for dissolution and separation of Belgian Congo pitchblende.

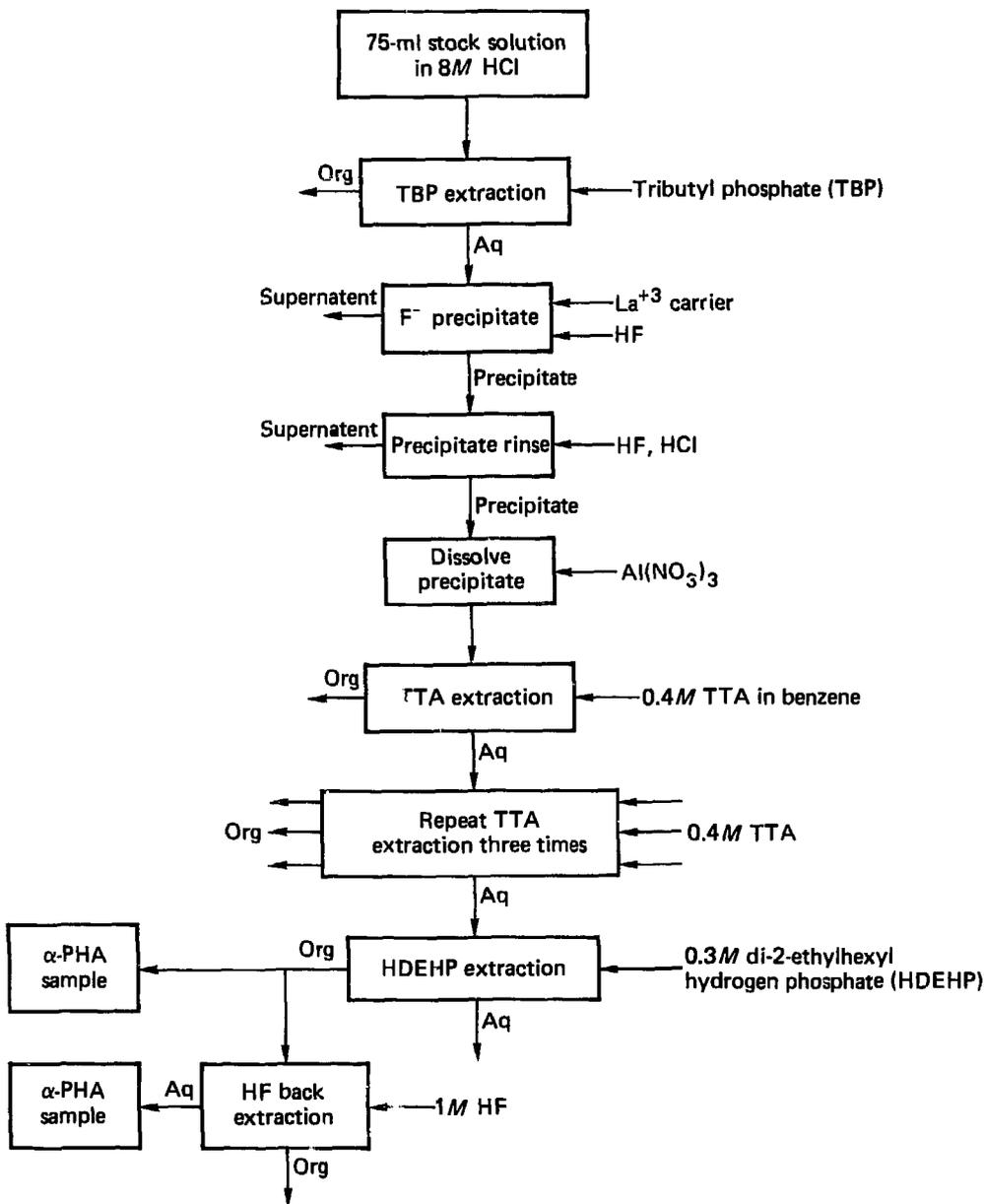


FIG. 3. Cotter concentrate separation procedure for investigating 4.4-MeV activity.

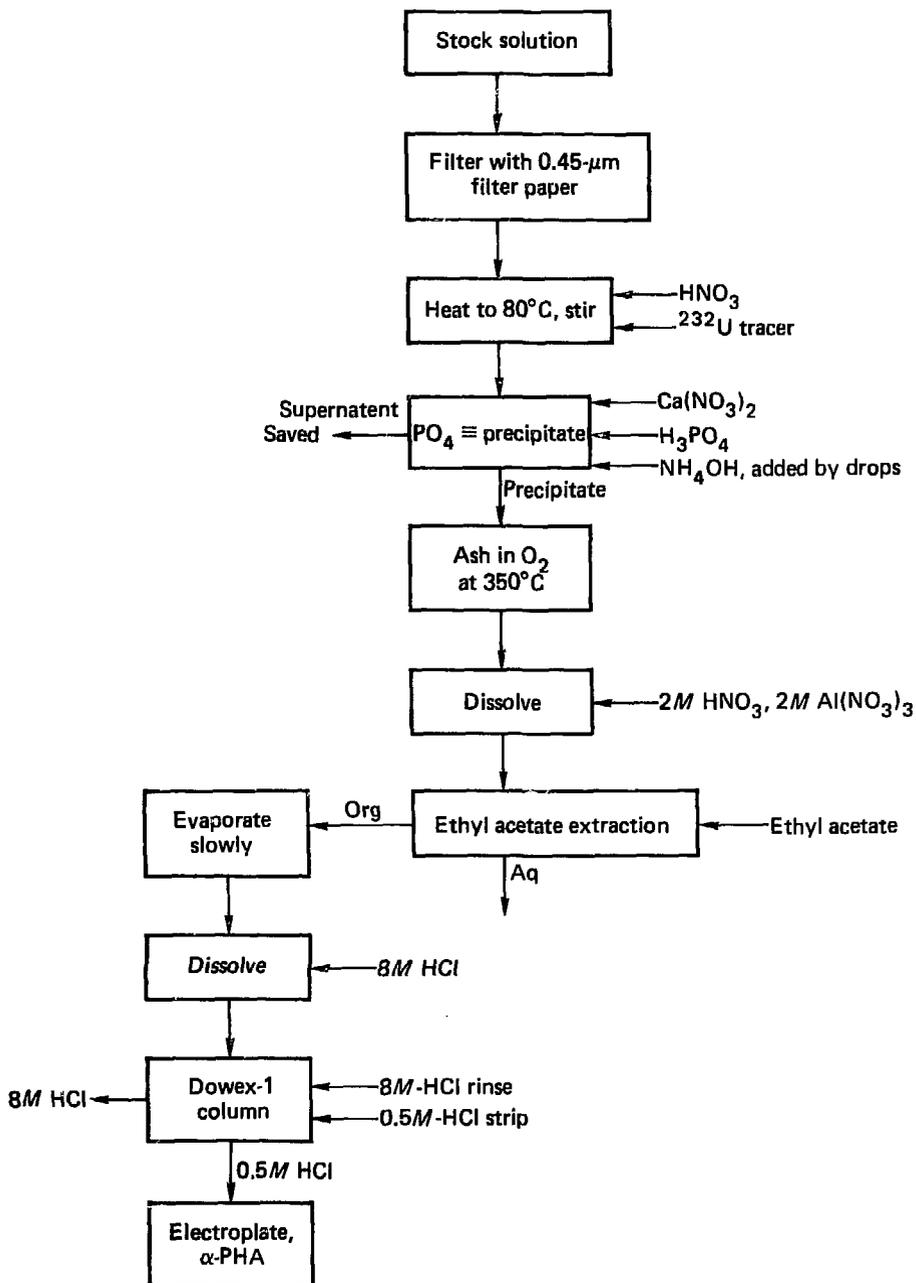


FIG. 4. Kerr-McGee raffinate separation procedure for investigating 4.4-MeV activity.

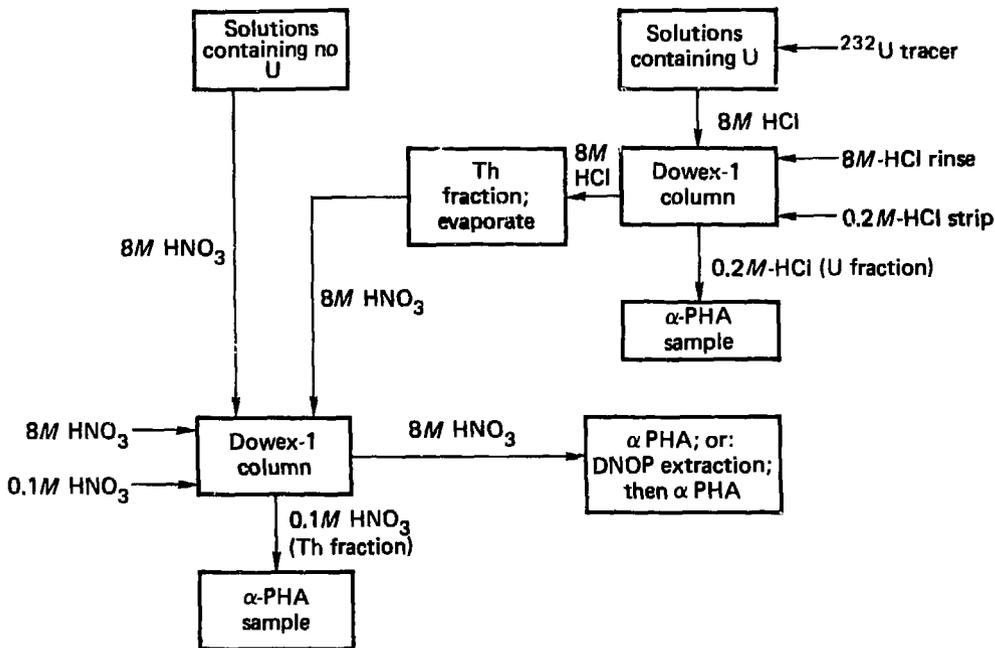


FIG. 5. Lanthanide separation procedure.

TABLE 1. Gross α activity after successive TTA extractions from 50- μ l Mallinckrodt distillate diluted to 0.1M HNO₃.

	Organic	Aqueous	Contact time
1st TTA	644×10^4 counts/m	42×10^4 counts/m	30 s
2nd TTA	36×10^4 counts/m	6.3×10^4 counts/m	30 s
3rd TTA	1.4×10^4 counts/m	4.7×10^4 counts/m	30 s
4th TTA	1.3×10^4 counts/m	4.8×10^4 counts/m	30 s

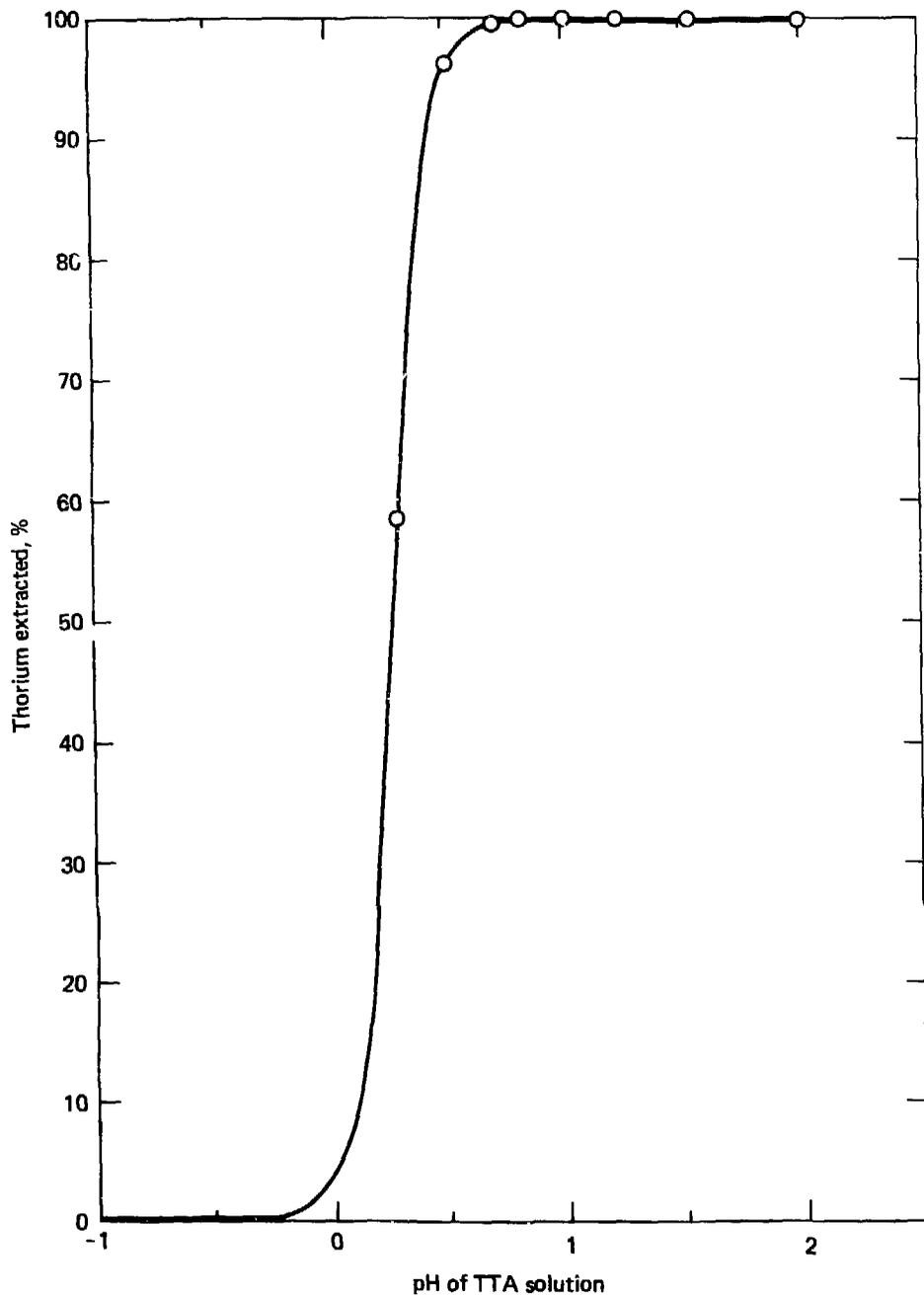


FIG. 6. Efficiency of extracting thorium with 0.4M TTA.

RESULTS AND DISCUSSION

Although we were unable to find any unusual α activity at 4.4 MeV, our investigations allowed us to place limits on the amount of activity that could be present in relation to the known elements. Table 2 shows the α activity from natural uranium, ^{230}Th , and ^{226}Ra in each of the four solutions. If the 4.4-MeV α emitter had followed the chemistry of thorium, it would have been present in amounts less than 0.1% of the ^{230}Th activity. Similarly, an upper limit of 0.1% for α activity was set for the emitter in the ^{226}Ra fractions. A 4.4-MeV activity would be required in greater than 1% of the uranium activity to have been detected in the uranium fractions. Upper limits of the 4.4-MeV activity in the lanthanide-actinide fractions, relative to the initial uranium activity of the solutions, are 10^{-8} , 10^{-7} , 10^{-4} , and 10^{-3} for the Congo ore solution, Cotter concentrate, Kerr-McGee solution, and Mallinckrodt concentrate, respectively.

Plutonium-239 was present in the Mallinckrodt concentrate at a level of 865 α decays/m \cdot ml. This amount is consistent with the findings of Peppard *et al.*, who determined the $^{239}\text{Pu}/^{238}\text{U}$ atom ratio in

the Belgian Congo ore³³ to be 1.5×10^{-11} . Americium-243 and ^{243}Cm were present at ≤ 1 decays/m \cdot ml in the Mallinckrodt concentrate. This eliminated the possibility that the ^{239}Pu , present in higher amounts than that found in other pitchblende and monazite sources,³² arose from a long-lived parent in an α decay chain.

Our failure to observe the 4.4-MeV α emitter cannot be ascribed to a unique shortcoming in either our chemical separation or our data analysis. The solutions we investigated contained α activity attributable only to thorium, uranium, and their daughters. Irreproducibility of results is a distressing theme which permeates the history of the observation of low-abundance α emitters in nature. Because of this, we believe that most, if not all, of the previous observations have been in error. In many cases, the errors may have resulted from poor α energy calibrations, unknown backgrounds in the detectors, or primitive systems for measuring the α energies. We have found no evidence for a 4.4-MeV α emitter in any of our four sources.

TABLE 2. Major α activities in the four solutions.

	U	Th	Ra
Mallinckrodt solution	2.41×10^3 α decays/m \cdot ml	2.82×10^8 α decays/m \cdot ml	2.0×10^6 α decays/m \cdot ml
Belgian Congo solution	4.92×10^4 α decays/m \cdot ml	2.36×10^4 α decays/m \cdot ml	2.36×10^4 α decays/m \cdot ml
Cotter concentrate	3.68×10^4 α decays/m \cdot ml	2.62×10^6 α decays/m \cdot ml	1.44×10^4 α decays/m \cdot ml
Kerr-McGee solution	8.55 α decays/m \cdot ml	4.79×10^2 α decays/m \cdot ml	4.30×10^1 α decays/m \cdot ml

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