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HEU AGE DETERMINATION

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Abstract

A new technique has been developed to determine the age of highly enriched uranium (HEU) in solids. Uranium age is defined as the time since the uranium-containing material was last subjected to a process capable of separating uranium from its radioactive-decay daughters. [Most chemical processing, uranium enrichment, volatilization processes, and phase transformations (especially relevant for uranium hexafluoride) can result in separation of the uranium parent material from the decay-product daughters.] Determination of the uranium age, as defined here, may be relevant in verifying arms-control agreements involving uranium-containing nuclear weapons. The HEU age is determined from the ratios of relevant uranium daughter isotopes and their parents, viz ^{230}Th/^{234}U and ^{231}Pa/^{235}U. Uranium isotopes are quantitatively measured by their characteristic gamma rays and their daughters by alpha spectroscopy. In some of the samples, where HEU is enriched more than 99%, the only mode of HEU age determination is by the measurement of ^{231}Pa since there is negligible quantity of ^{230}Th due to very low atom concentrations of ^{234}U in the samples. In this report the methodology and the data for determining the age of two HEU samples are presented.

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HEU AGE DETERMINATION[1]

by

A. R. Moorthy and W. Y. Kato

INTRODUCTION

The measurement of the age of highly enriched uranium (HEU) where the uranium age is defined as the time since the uranium material was last subjected to a separation process capable of separating uranium from its radioactive-decay daughters has become of interest recently because of the dismantlement of nuclear weapons and the possible availability of the HEU to enter the commercial sector as low enriched uranium after dilution with natural or depleted uranium. It is desirable to ensure that HEU which is undergoing dilution comes from dismantled weapons rather than from new production. While the "older" HEU cannot be conclusively attributed to a dismantled weapon, the age of the HEU provides assurance that the material was not from new production. From the safeguards perspective, for countries which have joined the NPT and given up their nuclear weapons, it is also useful to verify that HEU in storage is from dismantled nuclear weapons rather than from new production.

The HEU age is determined by chemically and quantitatively separating from dissolved HEU samples the daughter products ^{230}Th and ^{231}Pa in the ^{234}U and ^{235}U decay chains as outlined schematically in Figure 1. The procedure is to: 1) completely and homogenously dissolve samples of HEU containing uranium isotopes and their daughter products, 2) quantify ^{234}U and ^{235}U isotopes in the sample by gamma-ray spectroscopy using a Ge detector, 3) inject the HEU solution directly into an anion exchange column for individual separations of protactinium and thorium daughters, 4) prepare, separated alpha-emitting ^{231}Pa and ^{230}Th daughters, for alpha spectroscopy and 5) use the ratios of uranium isotopes and their respective daughters ^{231}Pa and ^{230}Th to determine the HEU age.

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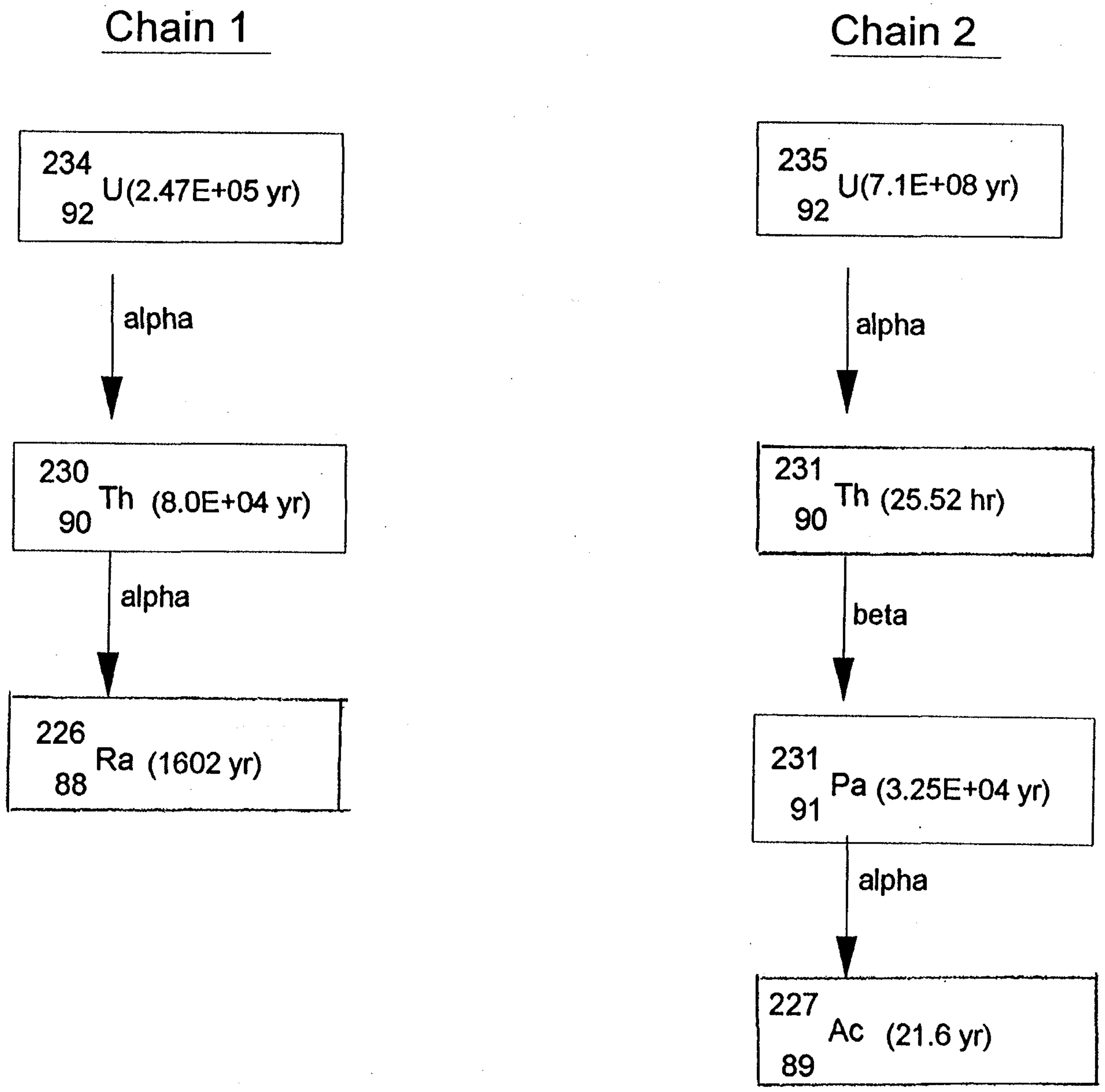


URANIUM ISOTOPES DECAY CHAIN

Figure 1

Two different samples of HEU containing different fractions of ^{234}U and ^{235}U were studied to demonstrate the age determination methodology. The HEU sample from Argonne National Laboratory (ANL) contained 93% ^{235}U while the second sample from Oak Ridge National Laboratory (ORNL) contained more than 99.9% ^{235}U.

CHEMISTRY

Protactinium and uranium form anionic complexes with hydrochloric acid. These complexes can be absorbed and desorbed selectively in an anion exchange resin by varying the normalities of the acid eluants. A step-by-step detailed procedure for the chemical separation of thorium and protactinium from uranium is presented in Appendix A.

In an earlier paper (1) the procedure called for the use of acid mixtures of acetic acid and hydrochloric acid of various normalities to effect separation of the isotopes of thorium, protactinium and uranium. However, in this analysis, since the samples which were analyzed were either pure uranium oxide or metal, it was not necessary to use acetic acid in the acid mixture.

An aliquot was prepared which contained a little more than 500 mg of the enriched uranium oxide from the ANL sample. The enriched uranium oxide was weighed and dissolved in about 10 ml of concentrated HNO_3 in a Teflon beaker. In the case of the 99.9% enriched sample, the sample was in the metal form and it took longer to dissolve in nitric acid. The solution was evaporated to dryness at low heat and concentrated HCl was added to convert the material to the chloride form. It was evaporated to dryness again and finally a solution in 10N HCl was made in a volumetric flask. This solution was then ready for the chemical separation of thorium and protactinium.

CHEMICAL RECOVERY EFFICIENCY

Two tracers were needed for the current investigation to measure the chemical recovery efficiency for thorium and protactinium. Gamma-ray emitting ^{233}Pa for quantifying ^{231}Pa and alpha emitting ^{228}Th for quantifying ^{230}Th were used as tracers. Certified ^{228}Th was obtained commercially. About 10 mCi of ^{233}Pa was produced by irradiating pure ^{232}Th (natural thorium nitrate) with 10^{17} thermal n/cm^2 in the BNL High Flux Beam Reactor (HFBR) as shown schematically in Figure 2. Neutron-irradiated natural thorium was dissolved in 10N HCl and

$$ {}^{232}\mathrm{Th} \xrightarrow[\text{(n, Gamma)}]{\text{(HFBR)}\ ⓧ} {}^{233}\mathrm{Th} \xrightarrow[\text{Beta decay}]{\text{(22.2 min)}} {}^{233}\mathrm{Pa} \xrightarrow[\text{(gamma)}]{\text{(27.7 d)}} $$

ⓧ 10 mg of natural thorium nitrate is irradiated in the High Flux Beam Reactor (thermal neutron fluence of 10^{17} n/cm^{2}).

^{233}PA PRODUCTION SCHEME

Figure 2

passed through a 2 ml anion exchange resin polypropylene column. About 20 column volumes of solution (10N HCl) were passed through the column to remove most of the thorium leaving behind ^{233}Pa in the column. ^{233}Pa was eluted with 10 column volumes of solution (8N HCl + 0.1N HF); the effluent was collected in a Teflon beaker. The acid effluent containing ^{233}Pa was then purified with the anion exchange column again. This was done by evaporating the solution in the Teflon beaker to dryness and re-dissolving the residue in 10N HCl and repeating the procedure described above. This yielded the ^{233}Pa tracer solution almost carrier-free. The ^{233}Pa solution was stored in a weighed 25 ml polypropylene container.

GAMMA-RAY COUNTING

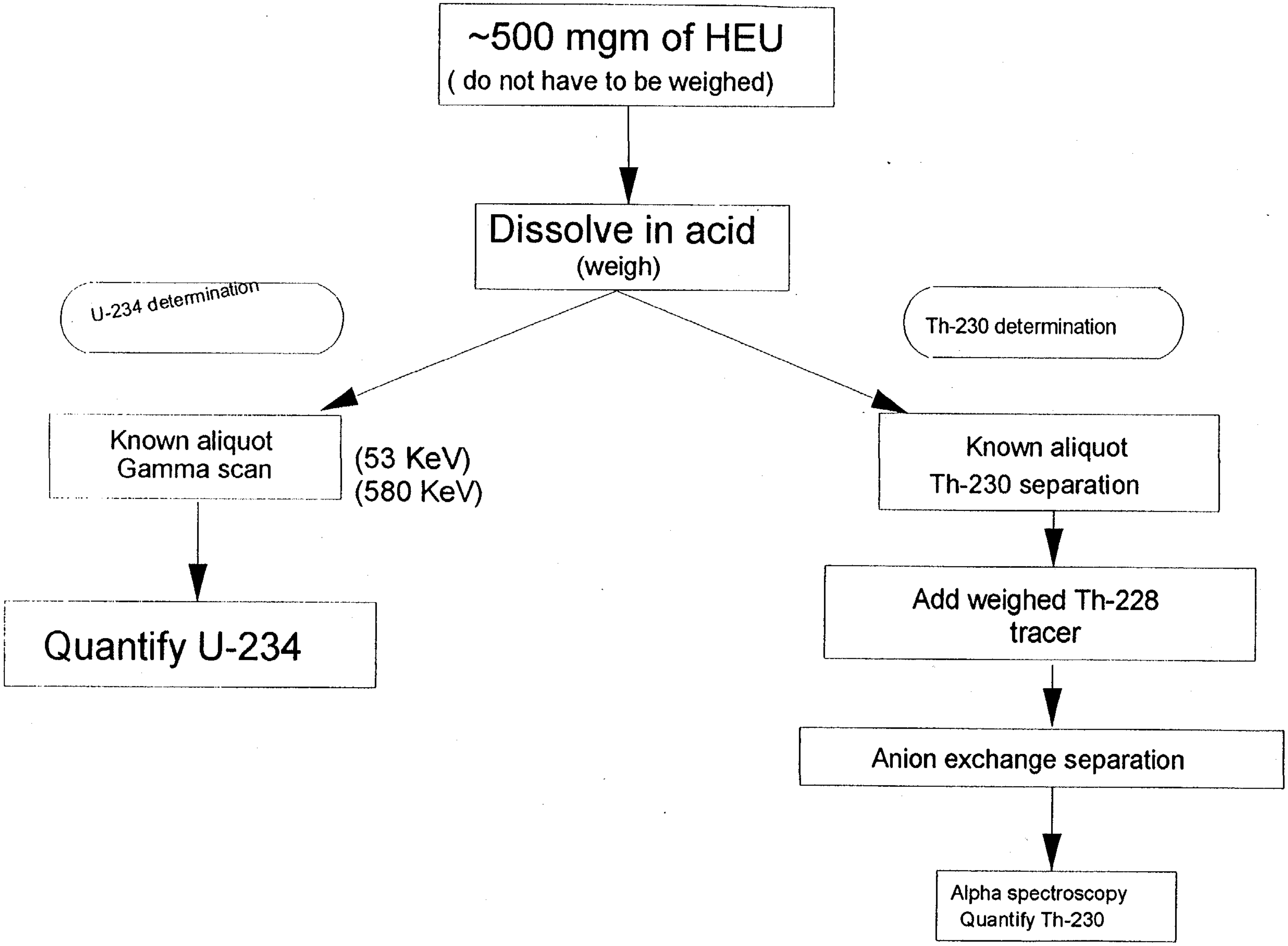
A known aliquot of the solution, representing about 5 mg of uranium oxide, was withdrawn and weighed in a 15 ml polypropylene bottle for gamma-ray counting in an intrinsic Ge detector. The 15 ml bottle geometry was previously calibrated with a National Institute for Standards and Technology (NIST) traceable mixed gamma-ray standard. The contents of the 15-ml bottle was diluted to appropriate volume and gamma-ray activities were counted in the Ge detector for 2000 seconds.

Gamma-ray activities were averaged from two gamma rays (185 and 143 keV) for quantifying ^{235}U in the uranium solution. Similarly ^{234}U was quantified from its respective two gamma rays (53 and 580 keV). A known weight aliquot from the ^{233}Pa solution was counted in the Ge detector using the 310 keV gamma ray for quantification.

After the withdrawal of an aliquot of the sample solution in 10N HCl for gamma-ray analysis, the remainder of the solution was quantitatively transferred to a Teflon beaker. Appropriately weighed amount of tracers ^{228}Th and ^{233}Pa were added to the sample solution. The sample was gently stirred with a magnetic stirrer for homogeneity in preparation for chemical separation of thorium and protactinium.

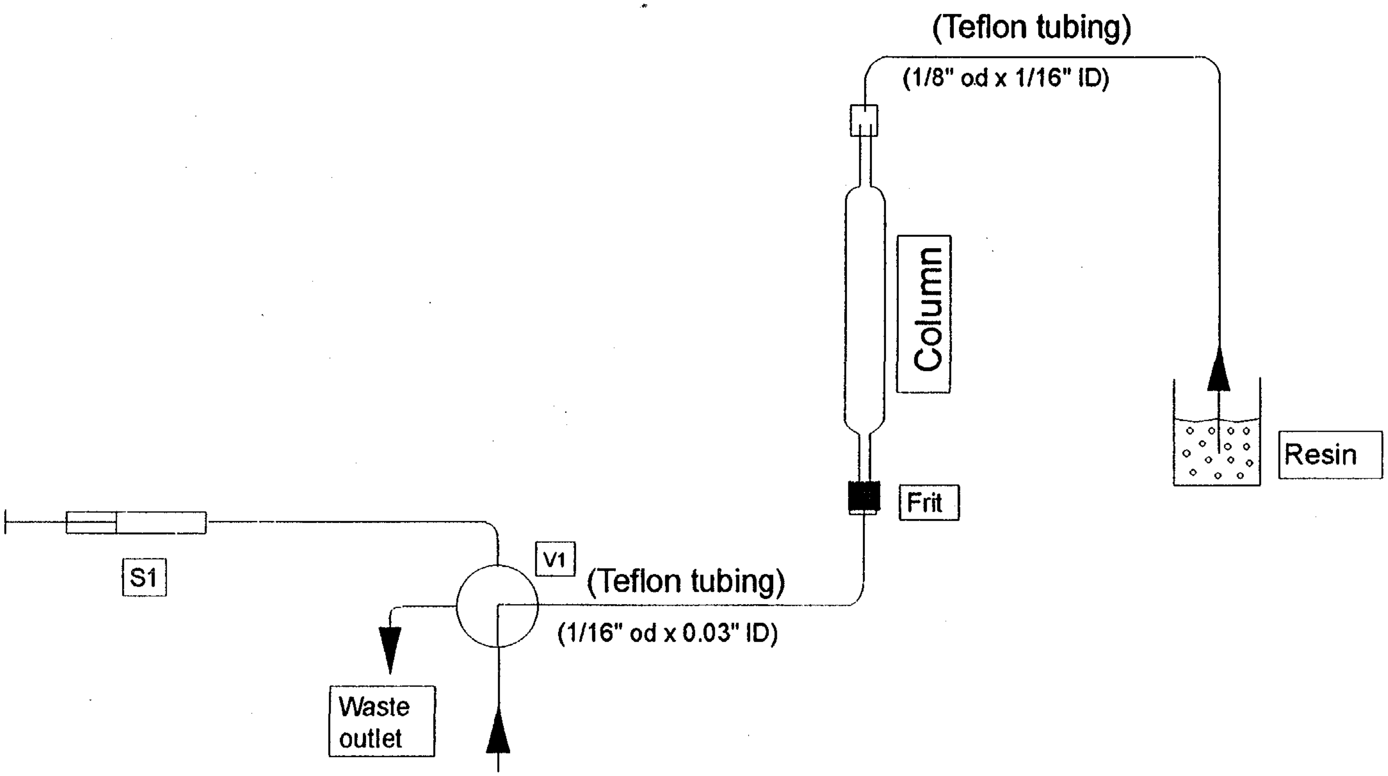
THORIUM SEPARATION

Thorium isotopes were separated as shown schematically in the flow diagram in Figure 3. A 35 ml Teflon column as shown in Figure 4 was filled with anion exchange resin (AG 1x4, from Dowex) using a syringe pump. The column was conditioned with 10N HCl using a piston pump.



THORIUM SEPARATION CHEMISTRY

Figure 3



S1: Syringe pumps
V1: Right angle 2 way valve

RESIN FILLING SCHEME

Figure 4

Following the preparation of the column, the sample containing the tracers in 10N HCl and the dissolved HEU were introduced as shown schematically in Figure 5, and passed through the column at a flow rate of 5 ml/minute. The effluent was collected in a 200 ml polypropylene bottle. The column was washed with 10N HCl until 200 ml of the effluent was collected in the bottle. The gamma-ray activity of the contents of the 200 ml bottle was measured using a Ge detector for identification and quantification of the uranium daughters ^{231}Th and ^{234}Th. This was only to trace the intermediate chemical recovery of thorium isotopes.

A 50-ml aliquot of the 200 ml solution was transferred to a Teflon beaker and evaporated to dryness. Dilute acid and 100 microgram of neodymium carrier were added to the dry beaker and gently heated. A quantitative precipitation using a filter paper was performed as described in the references (3, 4, and 5) for alpha spectroscopy. A typical alpha spectrum for the thorium isotopes is shown in Figure 6. ^{230}Th was quantified from the abundance of ^{228}Th.

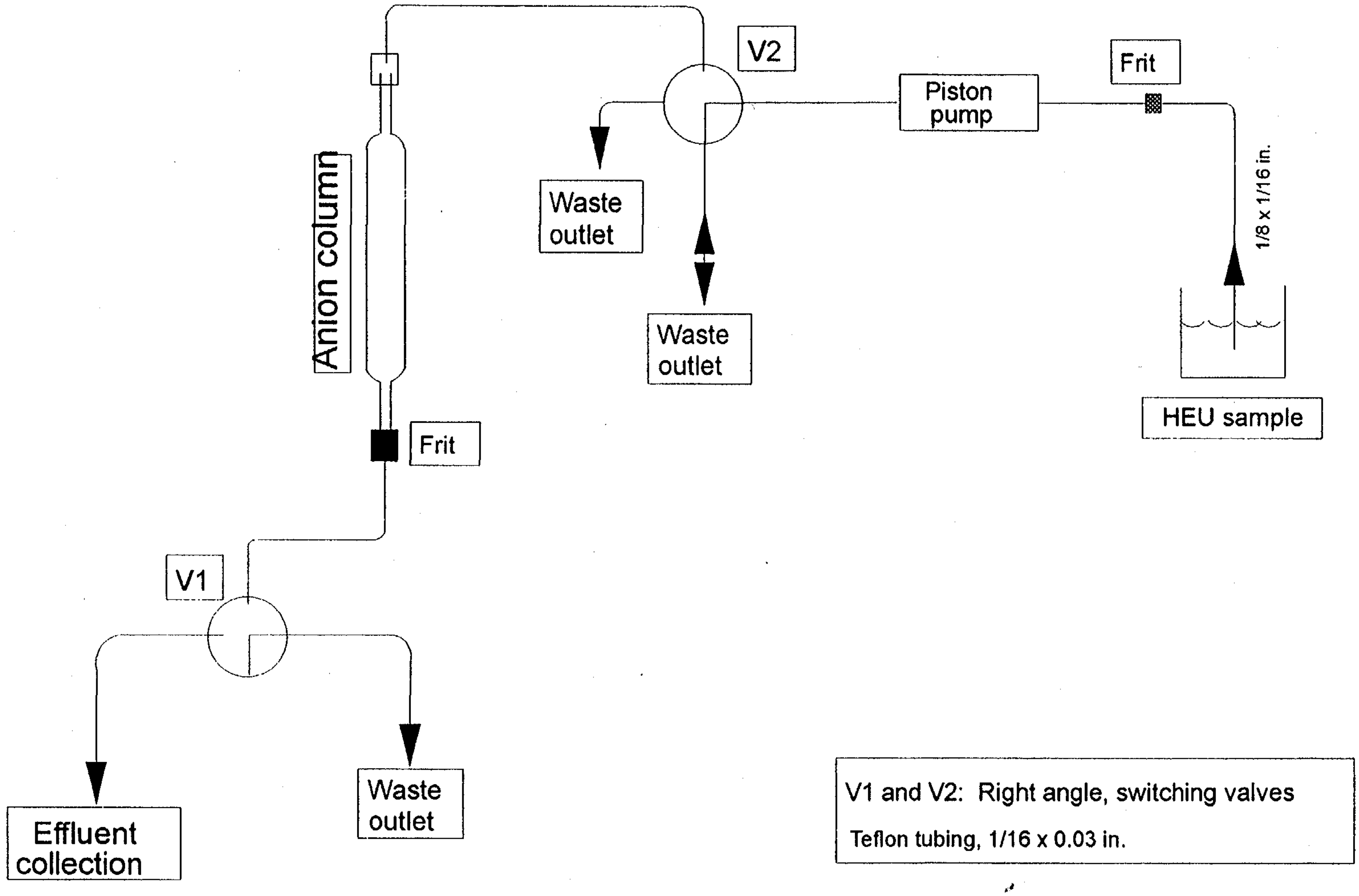
PROTACTINIUM SEPARATION

The process for separation of protactinium from uranium is shown schematically in Figure 7. After separation of thorium from uranium, the ion exchange resin contained mostly uranium and protactinium isotopes. The eluant was changed to an acid mixture of 8N HCl + 0.1 HF to elute protactinium isotopes. Two hundred ml of the acid effluent was collected in a Teflon beaker. The solution was evaporated to dryness and protactinium was purified twice with a 2 ml anion exchange column.

Since ^{234}U is present in the HEU in a much larger quantity than ^{231}Pa, some ^{234}U finds itself in the final ^{231}Pa sample; this poses a difficulty for obtaining well resolved-alpha spectra for ^{231}Pa. It was necessary to purify the separated protactinium twice using additional ion-exchange columns even at the expense of decreasing the chemical recovery to get a reliable alpha spectrum of ^{231}Pa.

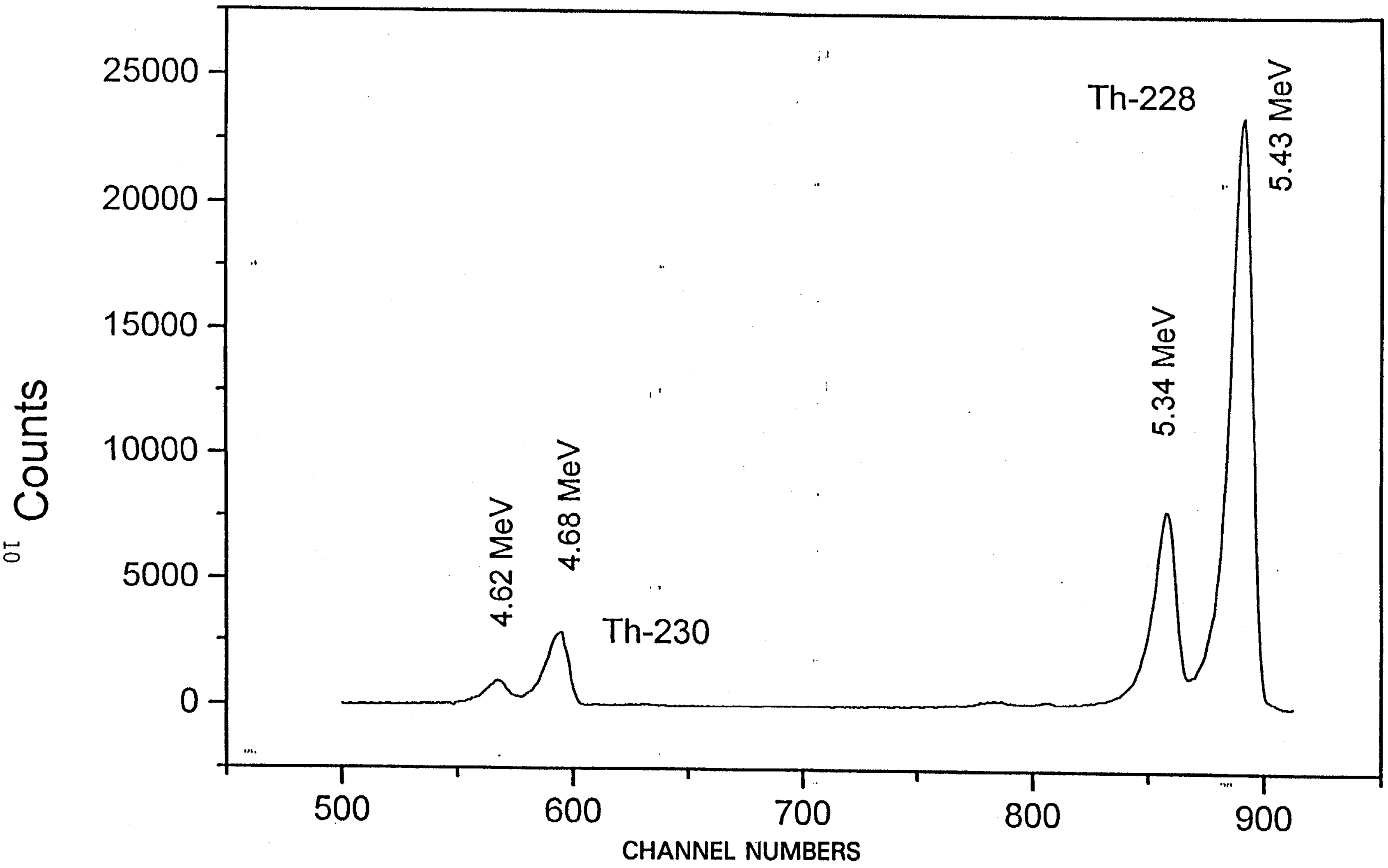
The twice purified protactinium fraction was subjected to alpha spectroscopy as described in the procedure for thorium. The ^{231}Pa alpha spectrum which was obtained is shown in Figure 8.

Uranium was eluted from the 35 ml column with 200 ml of 0.1N HCl and stored as waste.



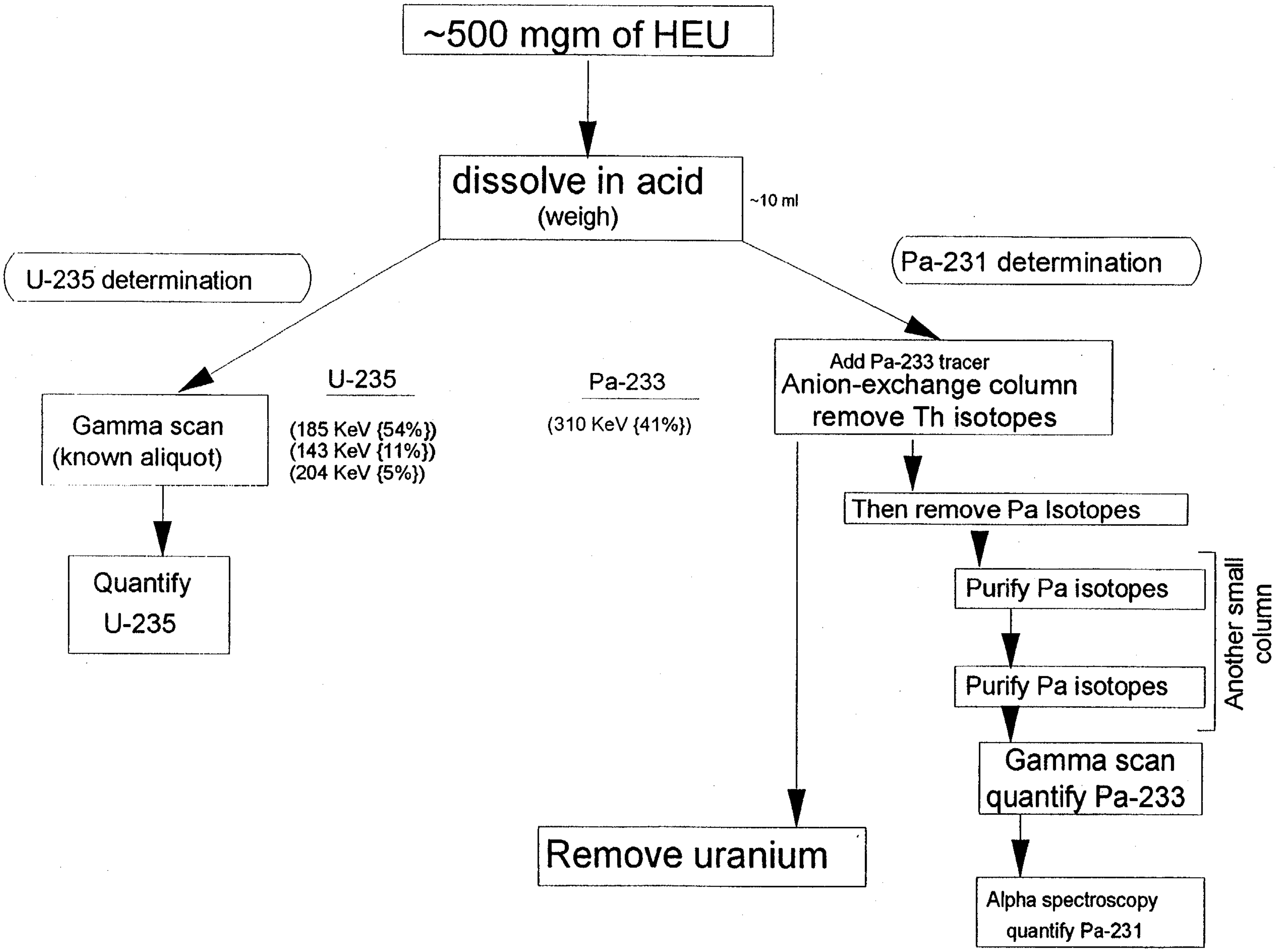
HEU AGE DETERMINATION CHEMISTRY

Figure 5



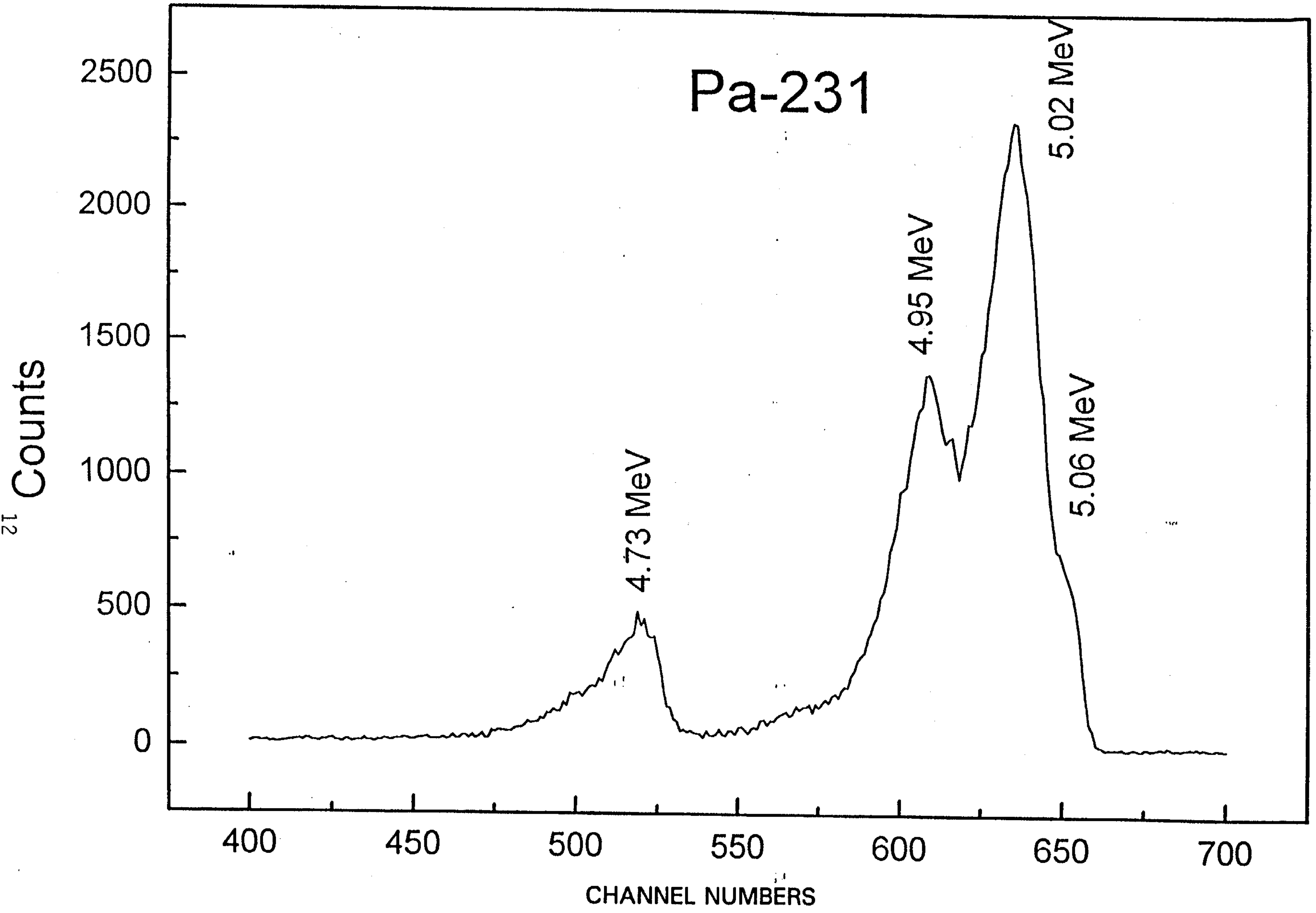
TYPICAL ALPHA SPECTRUM FOR THORIUM ISOTOPES

FIGURE 6



PROTACTINIUM SEPARATION CHEMISTRY

Figure 7



^{231}PA ALPHA SPECTRUM

FIGURE 8

RESULTS

For the 93% ^{235}U sample, the $^{230}Th/^{234}U$ ratio yielded an age of about 30 years, whereas the $^{231}Pa/^{235}U$ ratio yielded an age of about 24 years as shown in Table 1. The age of the HEU sample with 99.9% ^{235}U isotopic abundance, was determined to be 30 years as shown in Table 2. In HEU samples containing negligible quantities of ^{234}U, and therefore ^{230}Th, the only way to determine their age is by using the $^{231}Pa/^{235}U$ ratio methodology.

Table 1

93% ANL HEU SAMPLE

Age of HEU sample using $^{231}Pa/^{235}U$ ratio

Sample weight (gm)	^{235}U gamma (dpm)	^{231}Pa alpha (dpm)	Age (years)
0.5041	1900320	969	24
0.5590	2023000		

(Age of HEU sample using $^{230}Th/^{234}U$ ratio)

Sample weight (gm)	^{234}U gamma (dpm)	^{230}Th alpha (dpm)	Age (years)
0.501	51948000	23975	30

Table 2

99.9% HEU SAMPLE

Age of HEU sample using $^{231}Pa/^{235}U$ ratio

Sample weight (gm)	^{235}U gamma (dpm)	^{231}Pa alpha (dpm)	Age (Years)
0.5347	2245178	1280	28
0.5710	2481516	1638	32

CONCLUSION

A radiochemical thorium and protactinium separation technique combined with alpha spectroscopy has been developed to measure the ratios of $^{234}Th/^{234}U$ and $^{231}Pa/^{235}U$ to determine the age of HEU samples. This technique can be used routinely to determine the HEU age.

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APPENDIX A

HEU CHEMISTRY

The detailed step-by-step procedure for the dissolution of the HEU samples, the chemical separation of (a) the thorium and (b) the protactinium from the uranium samples is described in this appendix.

Dissolution of the HEU Sample

1) Dissolve approximately 500 mg of the HEU into a clean 500 ml Teflon beaker and cover the beaker with a Teflon cover.

2) Gently add about 5 ml of concentrated nitric acid while the beaker is still covered. There will be a vigorous reaction.

3) Digest the sample at low heat on a hot plate for about 30 minutes. The sample should be in complete solution. If not, add another 5 ml of concentrated nitric acid and continue to heat until the solution is a clear yellow solution.

> The above procedure is valid for HEU oxide sample. If the sample is of uranium metal, nitric acid is replaced with aqua regia (mixture of 2:1 ratio of concentrated HCl and HNO_3). If unsure, one can always start with aqua regia dissolution as aqua regia dissolves both oxide and metal samples.

4) Wash the inside of the cover and sides of the beaker with 8N HNO_3.

5) Evaporate the solution to dryness at a low heat. This may take about 8 hours.

6) When the beaker appears dry, add about 5 ml of concentrated HCl and evaporate to dryness at low heat as before.

7) Repeat addition of HCl and evaporation to dryness two more times. This will convert all the nitrates into chlorides.

8) Dissolve the dried salt in 50 ml of 10N HCl. Gently warm but do not evaporate lest the normality of the acid changes.

9) Transfer the solution to a previously weighed 100 ml polypropylene (PP) bottle. Wash the Teflon beaker with 10N HCl and transfer the washings to the PP bottle and secure the screw cover.

10) Reweigh the PP bottle with the solution. Compute the net weight of the solution.

Gamma-ray counting of the HEU sample:

1) Pipet about 1 ml of the solution from the 100-ml PP bottle into a previously weighed 15-ml PP gamma-ray counting vial and weigh both PP bottles again.

The difference in weights will give the quantity of the HEU solution that will be used for gamma counting.

2) Increase the volume in the 15-ml PP counting vial to the 15-ml mark with 1N HCl.

3) Place the 15-ml PP bottle inside the specially prepared lucite holder that centers the bottle on top of the intrinsic Ge detector.

4) Preset the counting time to 4,000 seconds and start the counting.

5) At the end of the counting period of 4,000 seconds, gamma-ray data of ^{235}U, ^{234}U, ^{231}Th and ^{234}Th are noted.

The calibration procedure for the Ge detector for 200-ml, 100-ml and 15-ml volumes is provided below:

Gamma-Ray Calibration Procedure:

A mixed gamma-ray certified standard solution that is traceable to NIST, is commercially available in microcurie quantities from the Amersham Company. The solution contains nine different gamma rays spanning the energy range from 88 keV to 2.3 MeV. The standard comes in a sealed glass ampoule, which can be diluted with acid.

1) Inside a hood, place a disposable plastic tray. Wear gloves and open the glass ampoule carefully with a file and the ampule located over the plastic tray. This is to confine any spill to the tray.

2) Quantitatively transfer the solution from the ampoule to a previously weighed 50-ml PP bottle. Wash the ampoule several times with dilute HCl and transfer the washings into the PP bottle.

3) Weigh the bottle again and quantify the gamma standard per unit weight. The 50-ml bottle is called "gamma-ray standard".

<u>15 ml standard:</u>

1) Weigh a 15-ml PP bottle and transfer to it an aliquot representing an equivalent of about 0.1 microcurie of the gamma standard.

2) Again weigh the 15-ml and 50-ml gamma-ray standard bottles after the transfer and quantify the gamma activity in the 15-ml bottle.

3) Increase the volume in the 15-ml bottle to the 15-ml mark, using 1N HCl. The standard is ready to be used.

4) Place the 15-ml bottle inside the specially designed lucite holder that centers the bottle on top of the Germanium detector and count the standard for 4,000 seconds. Repeat several times to get at least five separate counts.

5) Average the nine different gamma-ray values spanning from 88 keV to 2.3 MeV (counts/min) and enter the results in the software package that generates the calibration curve. This curve (15-ml geometry) will be used to quantify the gamma activity in 15-ml bottles.

<u>100- and 200-ml geometries:</u>

1) Prepare two more gamma-ray standards using 100- and 200-ml PP bottles, and the original gamma standard solution, as before. This time the diluted volume will be 100 and 200 mls.

1) Inside a hood, place a disposable plastic tray. Wear gloves and open the glass ampoule carefully with a file and the ampoule located over the plastic tray. This is to confine any spill to the tray.

2) Quantitatively transfer the solution from the ampoule to a previously weighed 50-ml PP bottle. Wash the ampoule several times with dilute HCl and transfer the washings into the PP bottle.

3) Weigh the bottle again and quantify the gamma standard per unit weight. The 50-ml bottle is called "gamma-ray standard".

15 ml standard:

1) Weigh a 15-ml PP bottle and transfer to it an aliquot representing an equivalent of about 0.1 microcurie of the gamma standard.

2) Again weigh the 15-ml and 50-ml gamma-ray standard bottles after the transfer and quantify the gamma activity in the 15-ml bottle.

3) Increase the volume in the 15-ml bottle to the 15-ml mark, using 1N HCl. The standard is ready to be used.

4) Place the 15-ml bottle inside the specially designed lucite holder that centers the bottle on top of the Germanium detector and count the standard for 4,000 seconds. Repeat several times to get at least five separate counts.

5) Average the nine different gamma-ray values spanning from 88 keV to 2.3 MeV (counts/min) and enter the results in the software package that generates the calibration curve. This curve (15-ml geometry) will be used to quantify the gamma activity in 15-ml bottles.

100- and 200-ml geometries:

1) Prepare two more gamma-ray standards using 100- and 200-ml PP bottles, and the original gamma standard solution, as before. This time the diluted volume will be 100 and 200 mls.

2) These standards will be used to generate calibration curves for 100-ml and 200-ml geometries.

2) These standards will be used to generate calibration curves for 100-ml and 200-ml geometries.

Preparation of ^{233}Pa standard:

1) Weigh a 10 mg alequot of spectrascopically pure thorium nitrate in a quartz ampoule and flame-seal it.

2) Irradiate the sample in a thermal fluence of about 10^{17} n/cm^2.

3) Cool for about two days. Wear gloves and open the ampoule with a file inside a hood and over a plastic tray.

4) Empty the contents into a clean Teflon beaker. If necessary, wash the ampoule with 10N HNO_3 transferring the contents to a beaker. Add about 10-ml of 10N HCl to the beaker and gently warm the beaker to dissolve the salts. At this point there should be a clear solution.

5) Using a 6-ml PP anion exchange column ion exchange column, which was previously conditioned with 10N Hcl, pass the 10N HCl solution containing the irradiated thorium salt at a flow rate of about 1 ml/min.

6) Clean the column with 50-ml of 10N HCl to remove all the thorium from the column, leaving behind the ^{233}Pa tracer.

7) Elute the ^{233}Pa with 50-ml of 8N HCl + 1N HF mixture and collect the effluent in a Teflon beaker.

8) Evaporate the HCl + HF mixture to dryness at low heat.

9) Add about 5-ml of 8N HCl and reevaporate to dryness.

10) Add about 10-ml of 10N HCl to dissolve the traces of thorium salt and ^{233}Pa.

11) Repeat steps 5 to 10 two more times to separate ^{233}Pa tracer from the thorium impurities.

12) Dissolve the protactinium fraction in the beaker using about 10-ml of 8N HCl + 1N HF solution.

13) Transfer the solution to a previously weighed 15 ml bottle and weigh the bottle again to get the weight of the solution. This solution is called "^{233}Pa tracer standard."

14) Pipet a weighed quantity (200 ul) of the standard into another previously weighed 15-ml PP bottle.

15) *Increase* the volume to the 15-ml mark using 8N HCl. Gamma scan the sample for about 600 seconds. ^{233}Pa emits a characteristic gamma ray of 310 keV with a half-life of 27.7 days.

16) Using the 15-ml geometry that was developed earlier with the Amersham mixed gamma-ray standard, quantify ^{233}Pa in the "^{233}Pa tracer standard." This is the standard that will be used for protactinium chemistry.

^{228}Th preparation:

A certified ^{228}Th standard that is traceable to NIST is available from Amersham in 0.1 uCi quantities.

1) Open the glass ampoules as before and quantitatively transfer the contents to a previously weighed 15-ml PP bottle.

2) Wash the ampoule with 1N HCl and transfer the washings to the 15-ml bottle.

3) Add about 10-ml of 1N HCl in to the bottle and weigh. The bottle will be called "^{228}Th tracer standard."

4) Quantify "the ^{228}Th standard" using information from the certificate. This solution will be used in thorium chemistry.

Separation of thorium isotopes:

The balance of the HEU solution in the 100-ml PP bottle from step 1 under "Gamma counting of HEU sample" is used for the chemical separation of the thorium and protactinium isotopes.

1) Pipet 200 microliter representing about 0.1 uCi of ^{233}Pa tracer directly to the HEU solution and mix uniformly by swirling. Weigh the PP bottles

which contain the HEU solution and the ^{233}Pa standards both before and after the transfer.

2) Pipet a known amount of the ^{228}Th tracer and mix the solution as before. Prepare the anion exchange column setup.

3) Condition the column with 250-ml of 10N HCl by placing the acid in the inlet and maintaining a flow rate of 5 ml/min.

4) Discard the effluent and place the resin in a clean and empty 200-ml PP bottle for the thorium fraction.

5) Place the HEU sample under the inlet now and maintain the sample flow rate at 5 ml/min.

6) When most of the sample has passed through the column, stop the pump and weigh the near-empty HEU sample bottle.

7) Change the eluant to 10N HCl at the inlet and collect 200 of the solution at the exit in the 200-ml PP bottle.

8) Secure the 200-ml PP bottle with its screw cap and count the bottle for gammas from ^{231}Th and ^{234}Th on a Ge detector for 4,000 seconds.

The anion-exchange resin absorbs only uranium and protactinium isotopes under these conditions leaving thorium daughters to pass unabsorbed.

9) The solution is ready for alpha spectroscopy.

Protactinium Separation:

Uranium and protactinium isotopes are left in the column.

1) Introduce the eluant mixture of N HCL + n HF (HCL + HF) at the inlet and collect 200-ml of the effluent at the exit in a new 200-ml PP bottle.

2) Gamma-ray scan for ^{233}Pa isotope in the effluent mixture.

3) Evaporate the solution containing protactinium in a Teflon beaker, to dryness.

4) Add 10-ml of 10N HCL to the beaker and digest at low heat.

5) Pass the solution through 5-ml ion exchange column.

6) Clean the column with 25-ml of 10 N HCL discarding the effluent.

7) Elute protactinium with 25-ml of n HCL + n HF mixture and evaporate to dryness as before.

8) Repeat steps 5 through 7.

9) Quantify the ^{233}Pa in the solution, by gamma-ray counting the 25-ml effluent.

10) The solution is ready for alpha spectroscopy using the procedures of References 3, 4, and 5.