Conf-940748--100

JULY 1994
BNL-60690
SSN 94-22

HEU AGE DETERMINATION*

by

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ABSTRACT

A criteria that a sample of highly enriched uranium (HEU) had come from a weapons stockpile and not newly produced in an enrichment plant is to show that the HEU had been produced a significant time in the past. The time since the HEU was produced in an enrichment plant is defined as the age of the HEU in this paper. The HEU age is determined by measuring quantitatively the daughter products ^{230}Th and ^{231}Pa of ^{234}U and ^{235}U, respectively, by first chemical separation of the thorium and protactinium and then conducting alpha spectrometry of the daughter products.

INTRODUCTION

An agreement between the United States and Russia has been signed whereby the United States will purchase 500 MT of highly enriched uranium (HEU) in blended low enriched uranium (LEU) form from Russia over the next twenty years. The HEU is to be derived from the Russian weapons stockpile. A criteria that the HEU comes from the weapons stockpile would be that the HEU had been produced in an enrichment plant some significant time in the past before the delivery of the LEU. The elapsed time since the production of the HEU by enrichment is defined as the age of the HEU in this paper.

Assuming that all of the daughter products are removed from the uranium during the enrichment process, it is possible to determine the age of HEU by quantitatively measuring the amount of one or both of the daughter products of ^{235}U or ^{234}U in a sample of HEU. The assumed nominal isotopic composition of HEU in this paper is as follows:

^{234}U	1%
^{235}U	93%
^{238}U	6%

METHODOLOGY

The natural decay chains for the ^{235}U and ^{234}U isotopes are shown in Figure 1. It can be seen that ^{231}Pa and ^{230}Th, the daughter products of ^{235}U and ^{234}U respectively, are ideal isotopes to determine the age of the HEU because of their

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relatively long half lives, if they can be quantitatively separated from the uranium and measured. As an example, based on their half lives, ten years after the HEU enrichment, there should exist about 15.9 d/s of ^{231}Pa per gram of HEU or 213 d/s of ^{230}Th per gram of HEU. ^{231}Pa disintegrates with a half life of 3.25E+04 yr emitting 4.950 Mev (23%), 5.029 Mev (20%), and 5.013 Mev (25.4%), alpha particles while ^{230}Th disintegrates with a half life of 7.7E+04 yr emitting 4.621 Mev (23.4%), and 4.688 Mev (76.3%) alpha particles.

CHEMICAL SEPARATION

The objective is to dissolve uranium isotopes and their daughter products of the HEU sample completely and homogeneously in an acid solution which can be directly injected into an anion exchange column for individual separations of protactinium and thorium daughters.

The quantitative separation was carried out by introducing a measured quantity of ^{233}Pa, a 27.0 d. half life beta emitter, and ^{228}Th, a 1.913 yr half life alpha emitter into the HEU solution. The ^{233}Pa was produced by irradiation of ^{232}Th with thermal neutrons in the Brookhaven Medical Reactor. The ^{228}Th was obtained as a certified standard solution from a commercial vendor.

Protactinium, thorium and uranium form anionic complexes with nitric and hydrochloric acids and these complexes can be absorbed and desorbed selectively by varying the normalities of the acid eluants[1,2].

Approximately 500 mg of uranium metal or oxide was dissolved in about 5ml of concentrated nitric acid. When the uranium is completely dissolved in the nitric acid, about 5 ml of concentrated hydrochloric acid is added along with known aliquots of ^{228}Th and ^{233}Pa tracers, and the solution is evaporated to dryness at low heat. The residue is dissolved in about 20 ml of 90% acetic acid + 10% (12N HCl + 0.1N HF). At this point the solution should be clear. An anion column with Dowex Ag 1 x 8, with a volume of 30 ml capacity is prepared as shown in figure 2. The column is conditioned with a 90% acetic acid +10% (12N HCl +0.1N HF) mixture. The sample is now introduced as shown in figure 3 at a flow rate of 5 ml/min into the column.

After the sample is completely introduced, the column is washed with the same acid to remove any alkali or alkaline earth products including the daughter products of the tracer ^{228}Th or impurities. Then the thorium daughter products and tracer (^{234}Th, ^{231}Th, ^{230}Th and ^{228}Th) are eluted with 100 ml of mixture of 60% acetic acid + 40% 7N HCl and a known aliquot is gamma counted in an intrinsic germanium detector and another known aliquot is evaporated on a platinum planchet and is beta with a plastic beta counter and alpha counted using a PiP surface barrier alpha spectrometer. ^{231}Pa and the

tracer ^{233}Pa are eluted from the column with 100 ml mixture of 60% acetic acid + 40% (12N HCl +1N HF). An aliquot is gamma counted and another aliquot is evaporated on a platinum planchet and they are beta and alpha counted.

Finally HEU is eluted with about 200 ml of 0.1 ml of HCl and collected into waste container. The column is monitored for any contamination and can be reused for the next run.

QUANTITATIVE ALPHA AND BETA COUNTING

In the case of the protactinium, the ^{231}Pa content is determined by alpha counting and the ^{233}Pa content is determined by beta and gamma counting. The purity of the ^{233}Pa isotope is checked by measuring the 27 day half life by beta counting. The ratio of the separated ^{233}Pa beta counting rate to the original ^{233}Pa counting rate provides the chemical separation factor. In addition ^{233}Pa is quantified by gamma counting.

The chemical separation factor for thorium from the HEU solution is determined by alpha counting the ^{228}Th sample before and after separation. Since ^{228}Th has a half life of 1.913 yr and emits 5.4233 Mev (71%) and 5.3406 Mev (28%) alpha particles, these alphas can easily be separated from the lower energy alphas of the longer half life ^{230}Th as shown in Figure 4 which is the alpha spectrum of the mixture.

ACCOMPLISHMENTS TO DATE

1. Chemical procedures for the separation of thorium and protactinium from uranium using an anion exchange column chemical process have been tested and shown to be adequate for the HEU age determination.

2. ^{230}Th has been separated from an HEU sample.

3. The age of an HEU sample has been estimated to be 22 years based on the ^{230}Th methodology using the ratio of the ^{230}Th to the ^{234}U alpha activities.

4. The age of the HEU sample based on the quantitative separation of ^{231}Pa from the HEU is currently being determined.

REFERENCES

1. Kim, J.I. and Born, H.J., Anion Exchange Behavior of Thorium, Protactinium, Uranium, and Other Elements in Hydrochloric and Acetic Acid Mixture, Radiochimica Acta, Band 14, Heft 1, 1970, p.35.

2. Kim, J.I. and Born, H.J., Anion Exchange Behavior and Separation of Thorium, Protactinium, and Uranium in Nitric Acid and Acetic Acid Mixtures, Radiochimica Acta, Band 14, Heft 2, 1970, p.65.

ACKNOWLEDGEMENT

The authors would like to acknowledge the contributions of L. Milian and D. Kaurin in

carrying out the chemical separation and the setting-up and calibration of the alpha, beta, and gamma counting equipment.

* This work was sponsored by the Office of Research and Development of the Office of Nonproliferation and National Security in the U.S. Department of Energy.

DECAY CHAIN OF ^{234}U AND ^{235}U ISOTOPES

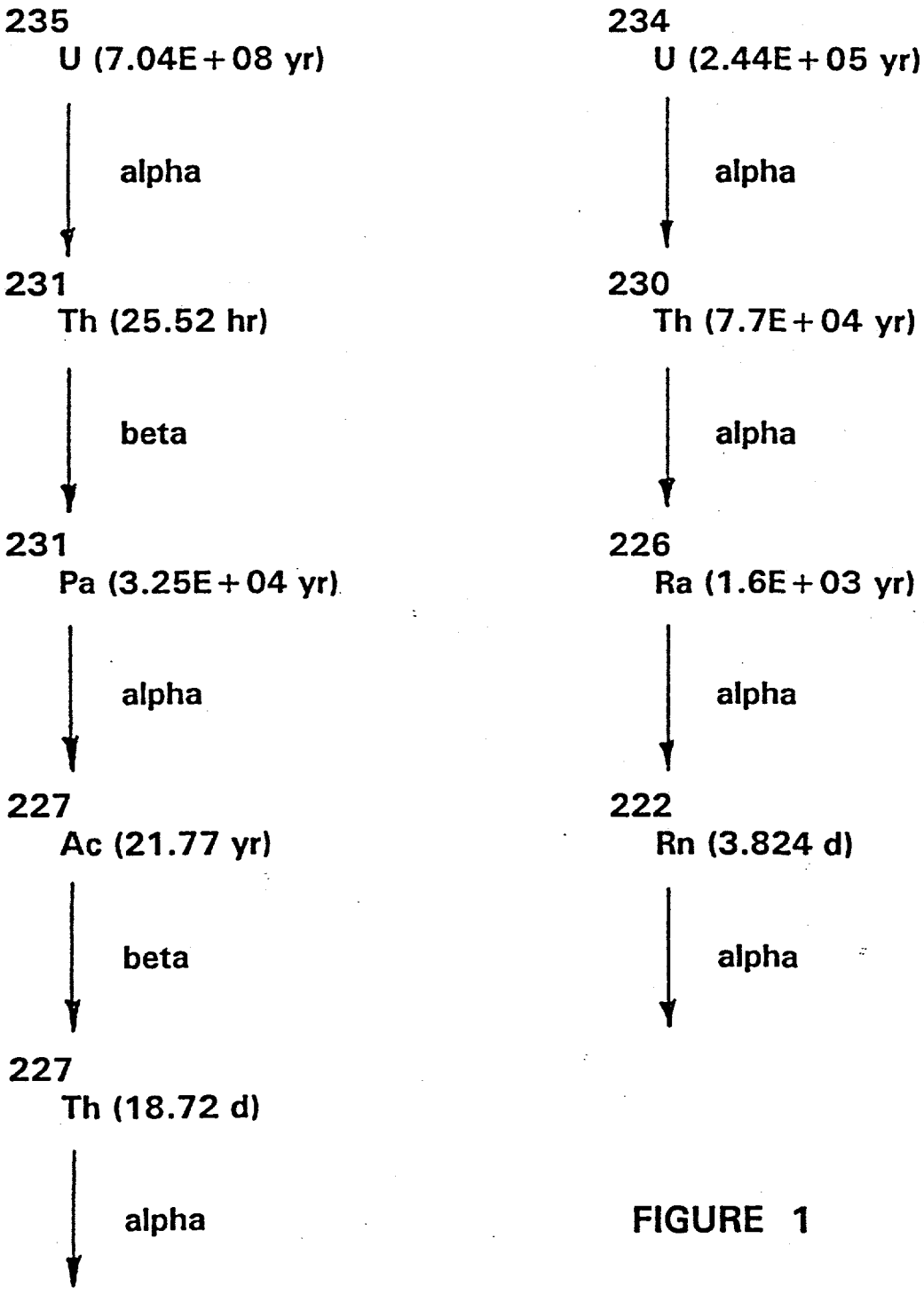
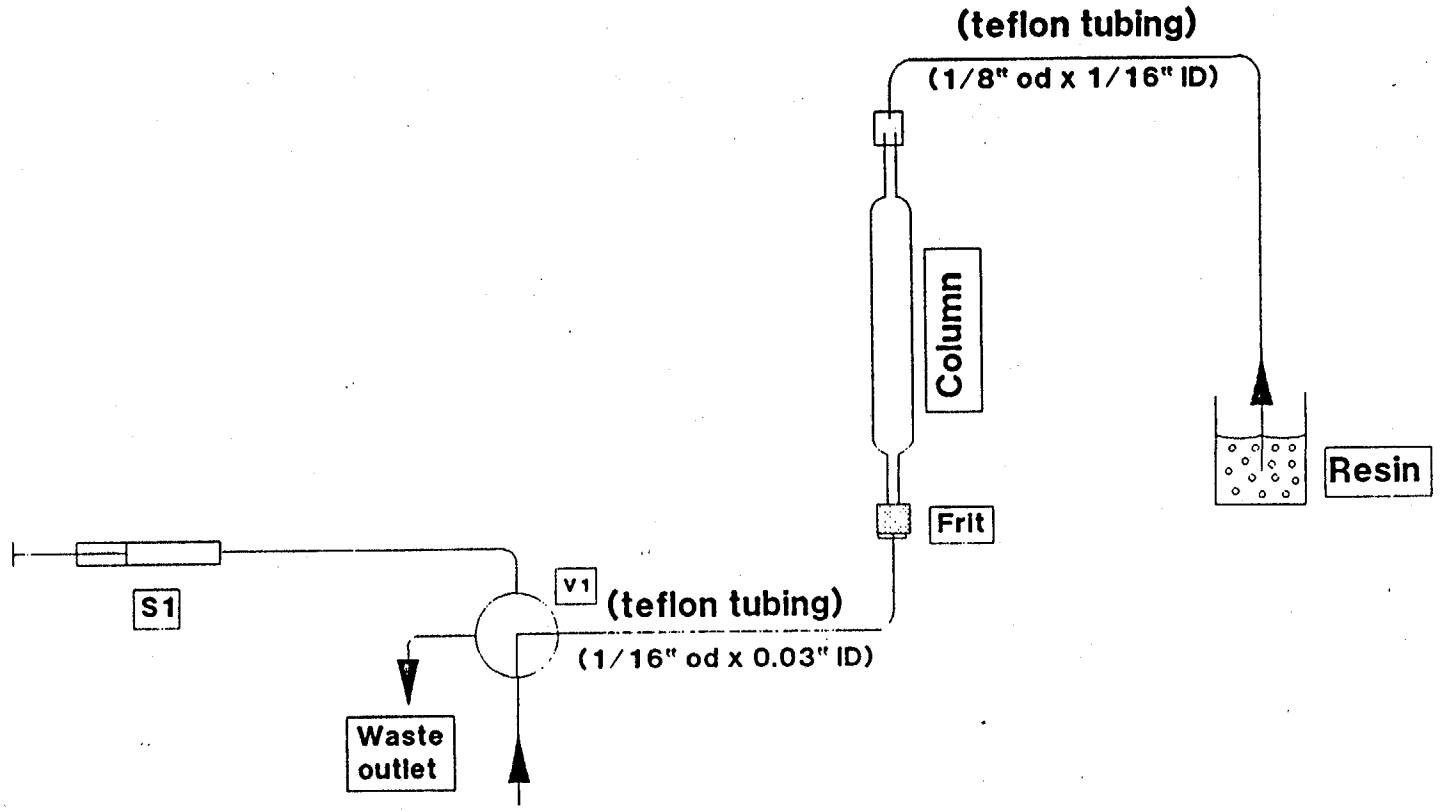


FIGURE 1

SCHEME FOR FILLING RESIN



S 1: Syringe pumps
V 1: Right angle 2 way valve

Figure 2

HEURESIN.DRW

HEU AGE DETERMINATION CHEMISTRY

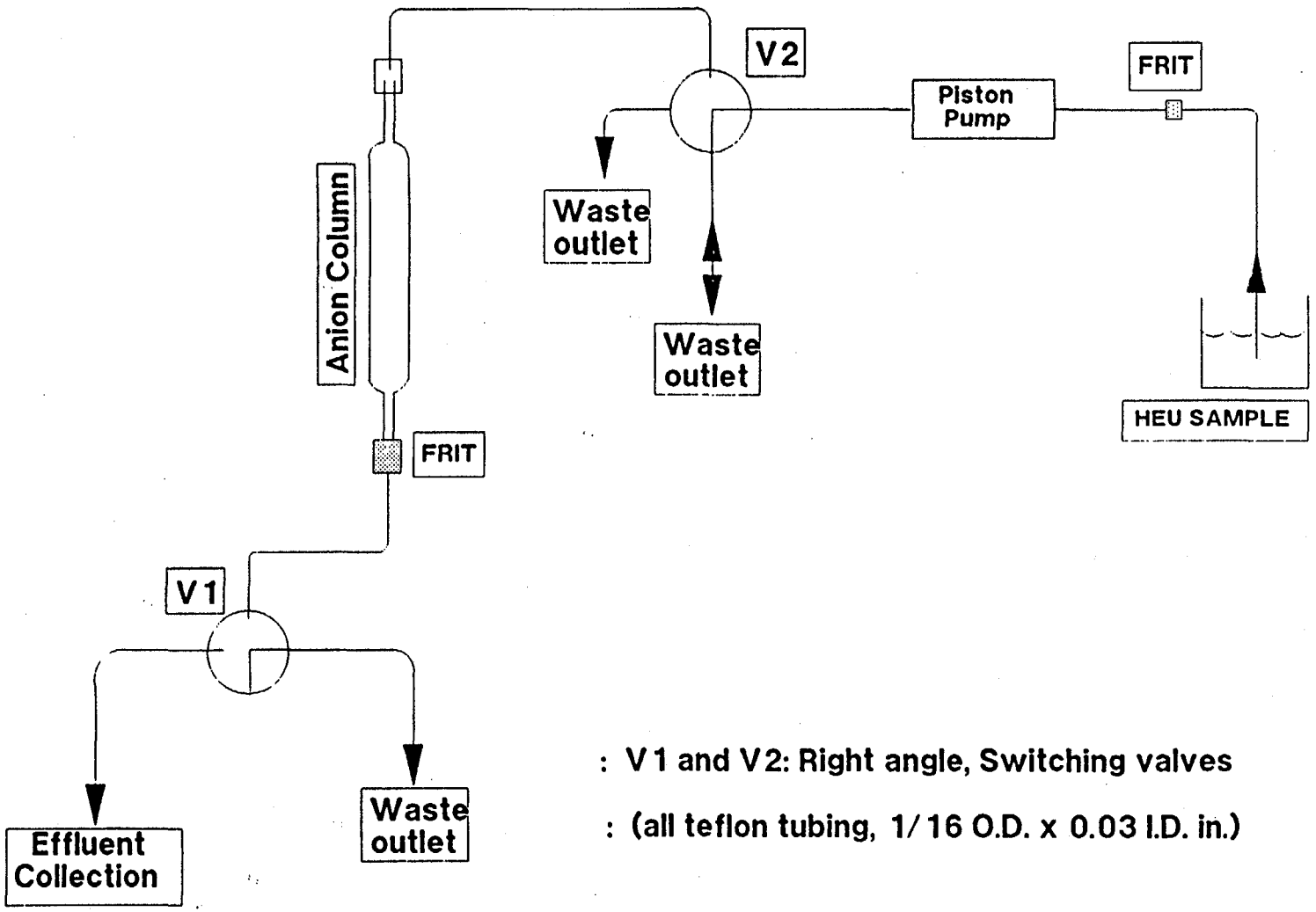


Figure 3

HEUPUMP 1.DRW

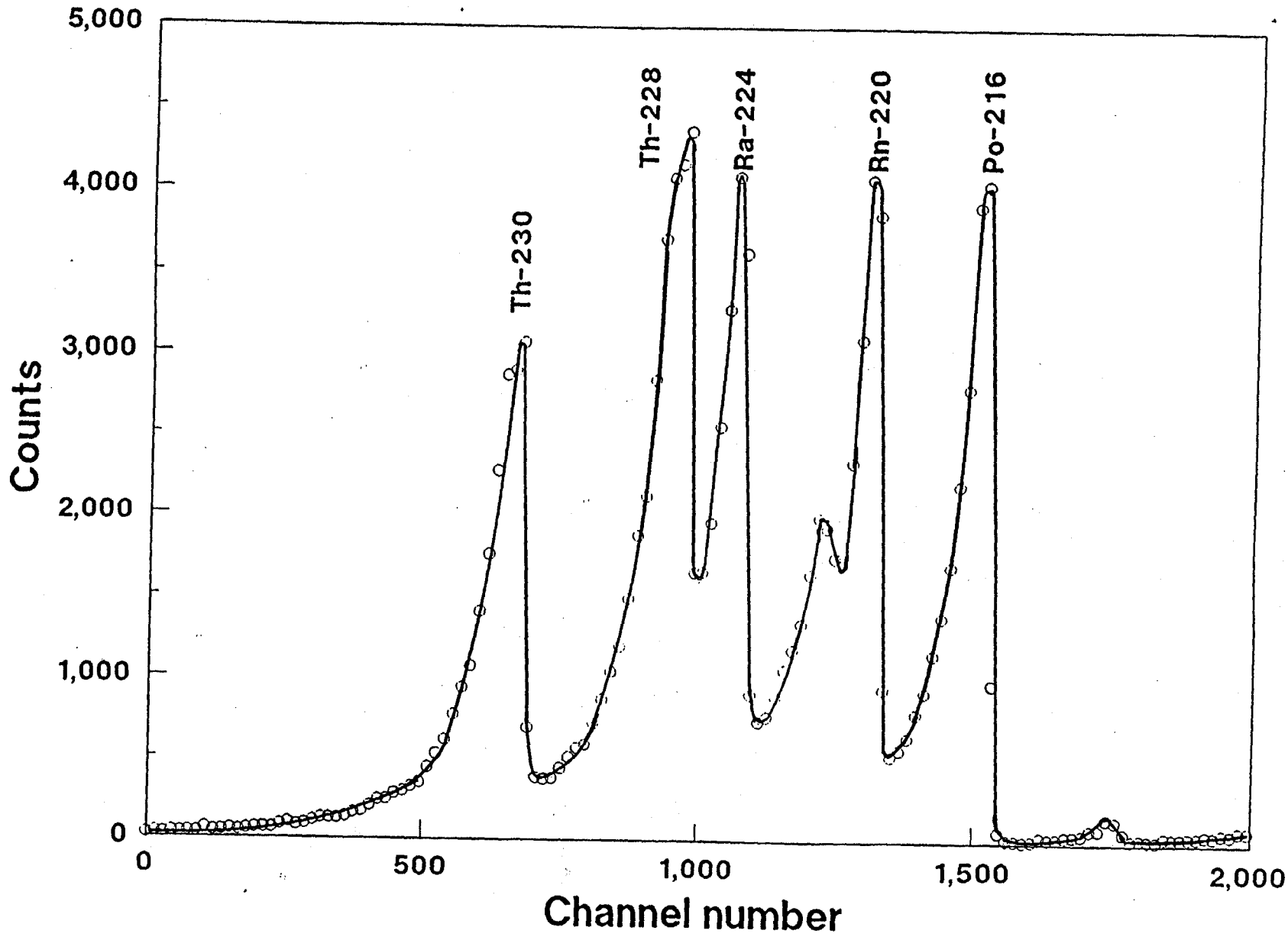


Figure 4

JTh-230.drw