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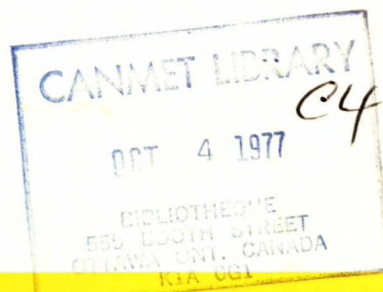
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THE DETERMINATION OF RADIUM - 226 IN URANIUM ORES AND MILL PRODUCTS BY ALPHA ENERGY SPECTROMETRY

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Laboratory for Chemical Analysis

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by

J.B. ZIMMERMAN* and V.C. ARMSTRONG**

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ABSTRACT

A reliable routine procedure for determining ^{226}Ra by alpha energy spectrometry is described. Radium is isolated as sulphate from the sample matrix by co-precipitation with a small mass of barium and analysed using a ruggedized silicon surface barrier detector. The method is capable of providing high accuracy over a large ^{226}Ra concentration range and is applicable to materials such as uranium ores, uranium mill products and effluent streams.

Samples resulting from nitric acid leach experiments with Elliot Lake ores were examined using the procedure. The distribution of ^{223}Ra, ^{224}Ra and ^{226}Ra between the leach products, (residue and leach liquor), is discussed.

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LA DETERMINATION DU RADIUM-226 DANS DES MINERAIS D'URANIUM ET DE PRODUITS D'USINE A L'AIDE DE LA SPECTROMETRE A L'ENERGIE ALPHA

par

J.B. Zimmerman* et V.C. Armstrong**

RESUME

Ce rapport décrit un procédé de routine sûr utilisé pour déterminer ^{226}Ra par la spectrométrie à l'énergie alpha. Le radium est isolé, comme sulfate, de la matrice de l'échantillon par la co-précipitation avec une petite masse de barium et est analysé à l'aide d'un détecteur de barrière avec une surface rugueuse de silicone. La méthode peut être très exacte sur une grande échelle de concentration du ^{226}Ra et peut être appliquée à des matériaux comme des minerais d'uranium, des produits d'uranium d'usine et des courants d'eau usée.

Des échantillons provenant d'expériences de lixiviation à l'acide nitrique avec des minerais d'Elliott Lake ont été examinés à l'aide de ce procédé. La distribution de ^{223}Ra, ^{224}Ra et de ^{226}Ra entre les produits de lixiviation, (résidu et solution de lixiviation) est discutée.

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INTRODUCTION

A reliable routine procedure was required for deter-
mining quantitatively the distribution of ^{226}Ra in process
products (i.e. solids, residues, leached liquors and washes)
resulting from a CANMET investigation into the nitric acid
leaching of uranium ores from the Elliot Lake area. It was also
anticipated that such a procedure would be valuable for deter-
mining ^{226}Ra over a wide concentration range in various other
sample types. For example, uranium mill wastes, in which ^{226}Ra
together with ^{228}Ra, ^{227}Ac and ^{230}Th concentrate, are of parti-
cular concern to environmentalists owing to the "very highly
hazardous" nature[1] of these contents. Since the release of ^{226}Ra
to the environment is coming under increasingly stringent Govern-
ment controls[2], accurate monitoring of its distribution is becoming
more important. It was necessary therefore to develop an analysis
procedure which would be both sensitive and selective and yet
sufficiently straightforward for its routine execution in a
suitably equipped laboratory.

Until quite recently ^{226}Ra determinations were performed
almost exclusively using either non-specific gross alpha counting
techniques[2], or the classical radon emanation method[4,5]. The
former may require long waiting periods before counting and/or
multiple counts at well separated times. The latter, while capable
of providing high accuracy and sensitivity, is also time consuming
and employs a specially constructed apparatus. The introduction of
the energy sensitive solid-state detector has facilitated the
accurate direct analysis of ^{226}Ra in the presence of the other

alpha-emitting nuclides found in the three naturally occurring radioactive series.

This report describes the isolation of radium from a variety of sample types in a form suitable for the direct determination of ^{226}Ra using a ruggedized solid-state detector system.

EXPERIMENTAL

The quantitative isolation of radium by precipitation can be achieved with barium as a carrier. To obtain well-resolved alpha spectra, the mass of the barium precipitate must be extremely small and the radiopurity of the radium nuclides high. Using the system described good recovery and adequate resolution may be obtained with 0.5 to 1 mg of barium.

Intrumentation

Preliminary activity checks on the barium precipitates and small insoluble residues could conveniently be made using a Baird Atomic University II series 530 single-channel analyser and Atomic model FC-110 windowless 2π proportional flow counter. A mixture of 90% argon and 10% methane was used as counting gas. Alpha energy spectra were obtained with an Ortec 450 mm^2 ruggedized silicon surface barrier detector, Ortec model 428 detector bias supply, model 121 charge-sensitive preamplifier, model 485 linear amplifier, model 408A biased amplifier, and a Northern Scientific NS 600 512-channel analyser. Samples were mounted

for counting and placed in an Ortec model 805 vacuum chamber, data printed by a series 3300 Teletype and spectra recorded with a Hewlett Packard 7004B X-Y recorder.

The resolution of the detector plus associated electronic equipment was found to be 46 keV FWHM using an Amersham/ Searle Corporation alpha reference source. This comprised an active area of 20 pCi ^{241}Am vacuum deposited onto a 25 mm diameter lightly oxidized stainless steel disc. Energy calibration was performed with an Amersham/Searle Corporation mixed alpha source (0.16 µCi ^{241}Am + ^{244}Cm + ^{214}Pu). The amplifier and biased amplifier were adjusted so that the spectrum of ^{226}Ra and its descendent nuclides (^{222}Rn, ^{218}Po and ^{214}Po) could be collected in 256 channels of the multi-channel analyser.

Owing to the "thickness" (and hence alpha energy degradation) of the samples obtained by the precipitation procedure, no effect on resolution would be expected by varying the sample to detector distance. Normally measurements were made with a source-to-detector distance of 1 cm. Counting efficiencies (E) were estimated by comparing the number of counts observed from a 1.6 cm diameter source, ($\sim$10nCi ^{226}Ra in equilibrium with its daughters), using the solid-state system with those using the 2π proportional flow counter. Then,

$$E = \left(\frac{\text{number of counts registered by Si detector}}{\text{number of counts contained in } 2\pi \text{ geometry}}\right)100\%$$

Apparatus

Electrically-ignited Paar Bomb

Variable heat hot plate

Millipore filtration apparatus (to hold 25 mm and
   45 mm diameter filters)

Magnetic stirrer + stirring bar

Platinum dish (50 mℓ)

Suction flask (500 mℓ), beakers (100, 600 mℓ)

Pipettes (1, 5, 25 mℓ), graduated cylinder (250 mℓ).

Reagents

Sodium peroxide: Reagent Grade powder

Sugar charcoal

Hydrochloric acid: concentrated

Citric acid solution: 1 mole ℓ^{-1}

Ammonium hydroxide: concentrated

Barium nitrate solution: 0.5 mg mℓ$^{-1}$ Ba

Lead Nitrate solution: 40 mg mℓ$^{-1}$ Pb

Disodium ethylenediaminetetraacetate solution:
   0.25 mole ℓ^{-1}

Sulphuric acid: 50%

Indicator paper: pH ranges 1-12, 3-5.5

Acetic acid solution: glacial

Hydrofluoric acid: concentrated

Perchloric acid: 70%

Ammonium sulphate: Reagent Grade crystals.

Chemical Procedure

The materials received by this laboratory for ^{226}Ra analysis include specimens from mill effluent streams, leach liquors, washes and residues from uranium ore acid leach studies, and untreated uranium ores. The ^{226}Ra content of these samples is therefore wide-ranging, (< 5 pCi ℓ^{-1} to nearly 1 nCi g^{-1}). Investigations with the Elliot Lake ores and solid residues demonstrated that the large amount of silica invariably present must be completely eliminated to ensure the quantitative recovery of radium. Treatment with hydrofluoric acid was not sufficiently effective. Fusion with sodium peroxide in a Paar bomb was established as a convenient routine procedure for the treatment of solids. For the complete analysis of certain liquid samples, suspended matter was taken into solution with hydrochloric and hydrofluoric acids. Once complete dissolution of the sample had been effected, the isolation of radium on barium was essentially the same for all sample types. The method used is based on that of Goldin [6]. Radium is concentrated from the solution with mixed barium and lead sulphates; the barium (with radium) is then separated from the lead by precipitation from ammoniacal EDTA solution with acetic acid.

i)   Treatment of Solid Samples

1.   Weigh 0.5g of the dried sample and transfer to a Paar bomb nickel crucible. Add approximately 5g Na$_2$O$_2$ and 0.4g pulverized sugar charcoal. Mix well, assemble the bomb and ignite.

2. Place bomb crucible in a 600 mℓ beaker, slowly add 100 mℓ water and warm on the hot plate. After dissolution of the melt remove the crucible, wash with water, then <u>cautiously</u> add concentrated HCl until the solution is acid.

3. Slowly evaporate the solution to dryness on the hot plate, taking care to avoid spattering of the solids. Raise heat to completely dehydrate the silica.

4. Add 150-200 mℓ water, 5 mℓ concentrated HCl and boil for 5 minutes.

5. Cool and collect insoluble silica residue on a Millipore HABPO4700 47 mm diameter filter disc. Retain solution in the filter flask.

6. Transfer filter to a 50 mℓ flat-bottomed platinum dish, ignite and then heat over flame to destroy carbon.

7. Allow platinum dish to cool, add 4 to 5 mℓ concentrated HF and 1 mℓ concentrated HCl. Evaporate to dryness on the hot plate. (Any ^{226}Ra contained in the silica is thereby released on volatilization of the SiF_4.)

8. To remove traces of carbon and expel fluoride remaining in the dish, treat with a few drops of concentrated HNO_3 and 2 mℓ concentrated $HClO_4$, evaporate to dryness, then heat over flame.

9. Add 2 mℓ concentrated HCl and 10 mℓ water. Warm to dissolve solids, then cool and filter through a Millipore HABPO2500 25 mm diameter filter disc into the flask containing the filtrate from step 5.

10. If, at this stage, any residue is visible on the disc it should be tested for alpha activity using the flow counter system. Steps 6 through 9 should be repeated till all the activity is taken into solution.

11. Transfer the combined filtrates to a 600 mℓ beaker. Add 5 mℓ citric acid solution*, then adjust pH to 8-9 with concentrated NH_4OH. Add 1 mℓ $Ba(NO_3)_2$ solution, (0.5 mg Ba^{++}), 1 mℓ $Pb(NO_3)_2$ solution, (40 mg Pb^{++}) and 0.5 g $(NH_4)_2SO_4$.

12. Heat solution to boiling and precipitate Pb, Ba(Ra) sulphates by dropwise addition of 1:1 H_2SO_4 to pH 2. The solution should be stirred during precipitation and the stirring continued for about ½ hour while mixture cools to room temperature.

13. Collect precipitate on a Millipore HABPO4700 47 mm diameter filter disc and discard filtrate.

14. Transfer disc with the precipitate to bottom of a 100 mℓ beaker. Add ⁓20 mℓ water and adjust pH to 8-9 with concentrated NH_4OH. Add 5 mℓ EDTA solution to complex Pb, Ba and Ra. Remove then rinse filter with fine stream of water and add 1-2g solid $(NH_4)_2SO_4$ to the solution.

15. Stir solution using small Teflon-coated stirring bar and precipitate Ba(Ra) sulphate with glacial acetic acid to pH 4. Continue stirring for about 0.5 hour.

(*Note: Samples containing unusually high quantities of calcium, for example due to the addition of lime, may require correspondingly larger volumes to prevent its precipitation as hydroxide.)

16. Collect Ba(Ra) sulphate precipitate on a Millipore HABPO2500 25 mm diameter filter disc. Discard filtrate. (NOTE: It is important to obtain a uniform distribution of the precipitate over the filter; care should be taken to avoid creeping of the precipitate up the filter chimney wall.)

17. Dry the filter disc and mount on stiff adhesive coated paper for alpha counting.

ii). Treatment of Standard Samples

Radium-226 standards were prepared from a portion of a well-analysed Port Radium pitchblende known to contain ^{226}Ra in secular equilibrium with other members of the 4n + 2 radioactive series. An accurately weighed sample was completely dissolved and the volume adjusted to give 2ℓ of solution in 5% v/v HNO_3. The ^{226}Ra concentration of the standard solution was calculated from the known uranium concentration to be 4.26 pCi mℓ$^{-1}$. Aliquots of the standard solution containing from 85.3 pCi to 341 pCi of ^{226}Ra were then treated according to steps 11 through 17.

Counting Procedure

Samples were normally counted in vacuo at a distance of 1 cm from the detector surface; a bias of -100v was applied across the detector during the measurements. Counting times of 1000 seconds were adequate for most of the routine samples encountered. For samples containing very small concentrations of ^{226}Ra, (< 10 pCi ℓ$^{-1}$), longer times were required for good counting statistics (see Results section). Radium-226 concentra-

tions were computed from the net areas under the ^{226}Ra peak
(4.78 MeV). For the majority of spectra obtained by the procedure
described, the ^{226}Ra peak was found to be sufficiently well
resolved for computation even after the ingrowth of ^{210}Po. How-
ever, where samples submitted for analysis already contained added
barium, the resulting spectra were broad due to the alpha energy
degradation. It was necessary therefore, in instances such as
these, to count samples as soon after precipitation as possible
so that corrections necessitated by overlapping peaks could be
kept to a minimum.

Tracer Studies

The fraction of barium carrier recovered was estimated
by spiking a number of samples with ^{133}Ba tracer. An Amersham/
Searle solution containing 0.11 mCi mℓ$^{-1}$ ^{133}Ba (as chloride) was
diluted to 0.66 μCi mℓ$^{-1}$. Then 50 μℓ of the diluted solution
were added prior to any chemical processing to either the weighed
solids in the Paar bomb crucible or to the aliquotted solutions.
Barium yields were estimated by comparing the 356 keV gamma counts
from the final barium precipitates with those from 50 μℓ of
tracer solution dried directly onto 2.5 cm diameter glass fibre
discs. Measurements were made using a Harshaw 2 in. x 2 in.
NaI(Tl) scintillation detector, a NS 600 1024-channel analyser
and associated electronic equipment.

RESULTS AND DISCUSSION

Figures 1(a) and 1(b) compare spectra obtained for the Amersham/Searle Corporation mixed alpha source with and without the biased amplifier and serve to illustrate the high resolution capability of the equipment used. No real advantage was realized by including the biased amplifier in the case of the sources prepared by the barium precipitation technique. The efficiency of the counting system for a sample-to-detector distance of 1 cm and source diameter of 1.6 cm was found to be 32%. The drastic reduction in counting efficiency as a function of increasing distance is illustrated in Figure 2.

A barium carrier mass of 0.5 mg was established as the smallest possible quantity consistent with good radium recovery. Adequate resolution of the ^{226}Ra peak in the presence of those of ^{223}Ra, ^{224}Ra and their descendent nuclides is then obtained. The effect of barium mass on resolution is illustrated in Figure 3. It can be seen that the barium content may be increased to 1mg without seriously impairing the definition of the ^{226}Ra peak. Where the barium content of the original sample results in a final barium mass of more than about 1mg the ^{226}Ra content may still be estimated by simple integration, providing counts are accumulated before appreciable ingrowth of the daughter nuclides and spectral interference from ^{223}Ra (5.70MeV) and ^{224}Ra (5.68 MeV) is minimal. Figure 4 illustrates the considerable increase in energy degradation for a barium carrier mass of only 2.5 mg. The sharp leading edge of the peak however still can be identified

with ^{226}Ra and the net peak area is in good agreement with that obtained using the prescribed 0.5 mg of barium. "Thick" precipitates containing appreciable amounts of ^{223}Ra and/or ^{224}Ra in the presence of ^{226}Ra result in spectra having overlapping peaks. If the overlap is not too pronounced it should be possible to estimate the contribution of these nuclides to the ^{226}Ra peak by fitting the curvature of the lower energy side of the interfering peak to a suitable equation.

The relatively large mass of lead carrier serves to ensure good barium recovery during its separation from the majority of the elements. Radium recovery was found to be high and independent of the mass of lead used over the range 10 to 100 mg. The ^{133}Ba tracer studies showed that, for a lead carrier mass of 40 mg and barium carrier mass of 0.5 mg, the overall barium yield for solid and liquid samples was close to 90%.

The standard curve for quantitative analysis, as described in the Experimental section, is illustrated in Figure 5. The ^{226}Ra recovery for the standards was established as better than 95% by counting under 2π geometry with the flow counter system. The counting efficiency of this system for the Amersham/Searle reference sources was established as 100%; this was assumed to hold for the larger-diameter standard sources. Alpha spectroscopy showed the standards to be essentially free of other radium nuclides and hence the theoretical number of alpha events from ^{226}Ra and its descendants could be computed.

The accuracy of the procedure was investigated by comparing experimental results with those computed from the known uranium content of well analysed ore samples. Table 1 compares data obtained for an Elliot Lake ore. Excellent radium recovery is therefore possible using the procedure described.

TABLE 1

Comparison of Measured and Calculated ^{226}Ra Concentrations

Sample	U conc.	Calculated ^{226}Ra Activity	Measured ^{226}Ra Activity	Ratio
Elliot Lake Ore	0.097%	324 pCig^{-1}	327 pCig^{-1}	1.01
Elliot Lake Ore	0.096%	322 pCig^{-1}	326 pCig^{-1}	1.01
Spiked Ore	–	422 pCig^{-1}	444 pCig^{-1}	1.05

The slightly lower yield indicated by the tracer experiments is consistent with the findings of Hallden et al.[8] who established that the recovery of ^{133}Ba added to bone samples was slightly poorer than recovery of ^{226}Ra.

The precision of the procedure was measured at three different radium levels. Standard deviations (S) and coefficients of variation (V) were calculated using the expressions,

$$S = \sqrt{\frac{\Sigma(\bar{n} - n)^2}{N-1}} \qquad \text{and} \qquad V = \frac{S}{n} \cdot 100$$

where

n = observed counts

$\bar{n}$ = mean of observed counts

N = number of replicate determinations

and V compared with the coefficient of variation for the average

total counts observed (V_c), where,

$$V_c = \frac{S_c}{\bar{n}} \cdot 100 \qquad \text{and} \qquad S_c = \sqrt{\bar{n}}$$

The data are summarized in Table 2. For the standard solutions,

TABLE 2

Precision Measurements for Different ^{226}Ra Concentrations

	5 pCi level[a] (counts/2000s)	150 pCi level[b] (counts/1000s)	1000 pCi level[a] (counts/500s)
Run #1	80	1080	3175
#2	62	1050	3265
#3	72	960	3225
#4	62	1000	3215
#5	76	1110	–
$\bar{n}$	70	1040	3220
S	8	51	37
V	11.4%	5.1%	1.2%
V_c	12.0%	3.1%	1.8%

a) Prepared from aliquots of standard solution.
b) Prepared from aliquots of residue from nitric acid leach.

the precision appears to be a function of the counting times used. A precision of ±10%, and hence a counting time of 2000 seconds, at the 5 pCi level should be adequate for the majority of samples likely to be received. The larger difference between S and S_c found for the solid samples could be attributed to sampling small aliquots (0.5 g) from an inhomogeneous material.

The Nitric Acid Leaching of Elliot Lake Ores

The determination of radium by alpha spectrometry was largely developed with a view to rapidly providing information concerning the distribution of ^{226}Ra in products resulting from the nitric acid leach studies of Elliot Lake ores. Some preliminary findings are discussed here to illustrate the value of this technique. Table 3 lists data for the feed material, a leach liquor, residue and water wash. The results are typical of

TABLE 3

The Distribution of ^{226}Ra in Nitric Acid Leach Products

Sample	Total Volume or Weight of Sample	Aliquot Sampled for Analysis	^{226}Ra content of Fraction
1. Residue	828 g	2.0 g	305 nCi
2. Leach Liquor	300 ml	10.0 ml	5 nCi
3. Water Wash	1200 ml	10.0 ml	3 nCi
1 + 2 + 3			313 nCi
Feed Material	1000 g	0.5 g	326 nCi

a number of samples which were reanalysed by the spectroscopic method several weeks after the acid leaching. More than 90% of the ^{226}Ra was found in the residue. Figures 6(a) and 7(a) show spectra recorded for the residue and leach liquor soon after the final barium (radium) precipitation step, (i.e. before significant ingrowth of those descendent nuclides having a half-life longer than a few minutes). Figures 6(b) and 7(b), (c) and (d) show subsequent measurements and illustrate the ingrowth

of alpha-emitting daughter nuclides followed by decay of the shorter-lived ^{224}Ra and ^{223}Ra families. Measurement times are included in the Figures together with the peak assignments made by reading the energy at which the extrapolated leading edge of the peak intercepts the energy axis[7]. The apparent deterioration in resolution of the decayed samples (Figures 7(c) and 7(d)) occurs because of an increasing contribution, and hence overlap, from the relatively longer-lived ^{223}Ra; (11.43 days compared with 3.64 days for ^{224}Ra).

The spectroscopic analyses therefore demonstrate clearly the almost complete separation of ^{226}Ra from ^{223}Ra and ^{224}Ra into the residues. A situation such as this might be anticipated through the conventional sulphuric acid leaching of uranium ores where radium would be largely retained in the residue as sulphate. The separation of the ^{226}Ra series can then be explained by considering the relative half-lives of ^{223}Ra, ^{224}Ra and ^{226}Ra. On completion of the acid leaching the radium nuclides are present in the residue as sulphate; their parent nuclides, ^{224}Th, ^{227}Th, and ^{230}Th accompany uranium in the leach liquor. The ^{223}Ra and ^{224}Ra then grow back into solution from thorium with half-lives of 11.43 and 3.64 days respectively. Radium-226 will not be detected for a much longer time because of its long half-life, (1620 years). In the case of the residue the shorter lived ^{223}Ra and ^{224}Ra nuclides will essentially disappear after approximately six weeks leaving only the ^{226}Ra family. Analyses of nitric acid leach products soon after completion of the leaching verified that the above situation obtains in this system,

(Figures 8 (a) and 9 (a)). Sampling of the residue and liquor at a later date showed the predominant presence of the ^{226}Ra and ^{224}Ra series respectively, (Figures 8 (b) and 9 (b)), as found previously. The reason for the retention of radium in the residue in a nitric acid system is not easily explained. Formation of sulphate ion by the oxidation of sulphide present in the ore has been demonstrated [9,10] and could result in radium precipitation in the presence of naturally occurring barium or lead. Work to elucidate this mechanism is continuing.

ACKNOWLEDGEMENTS

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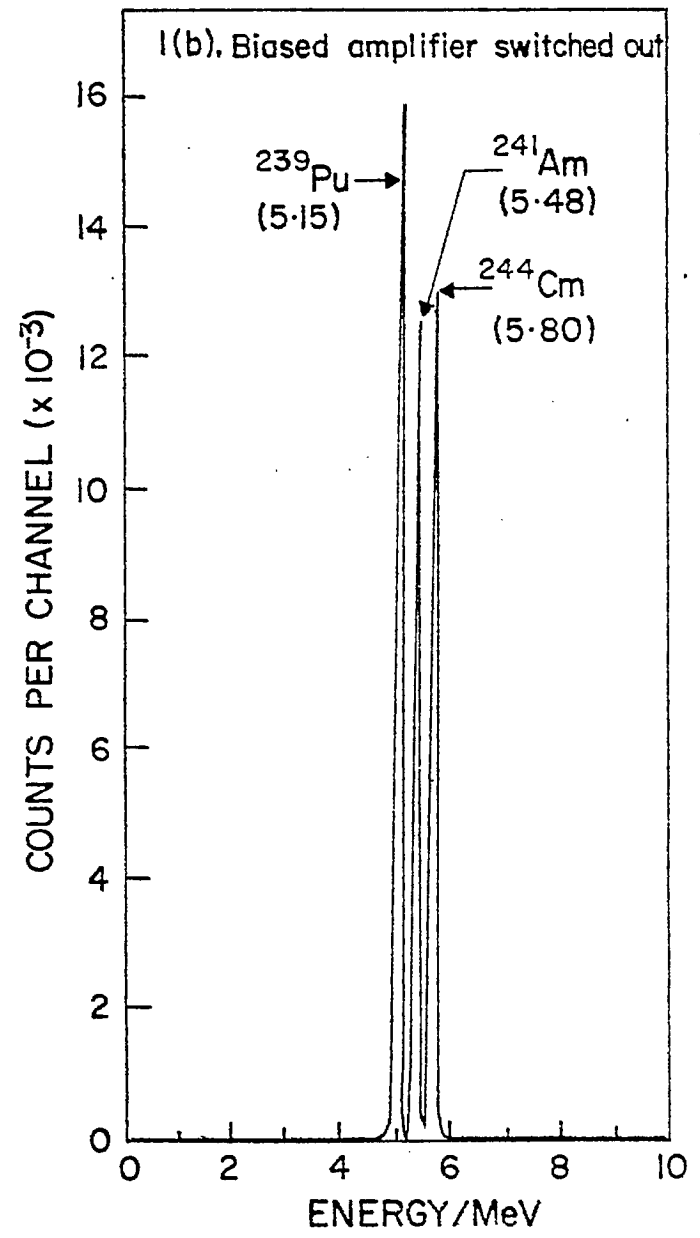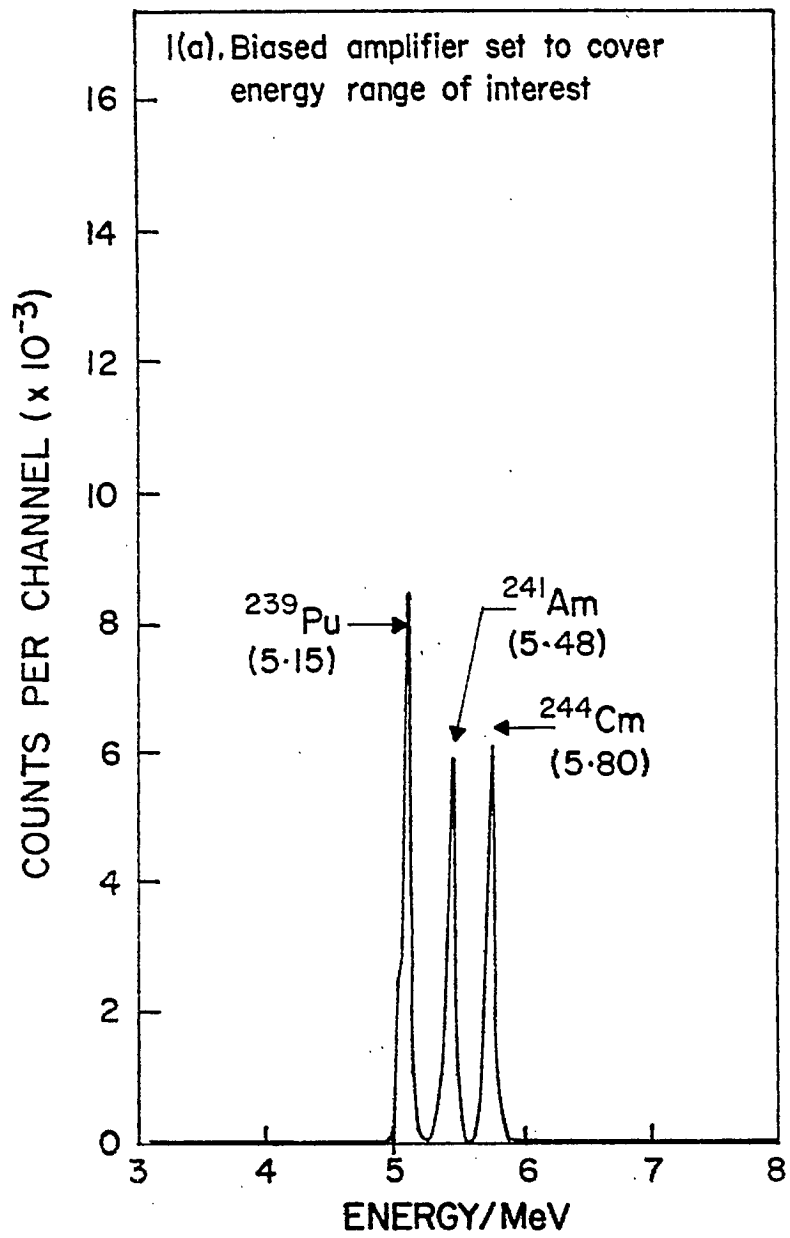
Figure 1.   Spectra of Mixed Alpha Reference Source.
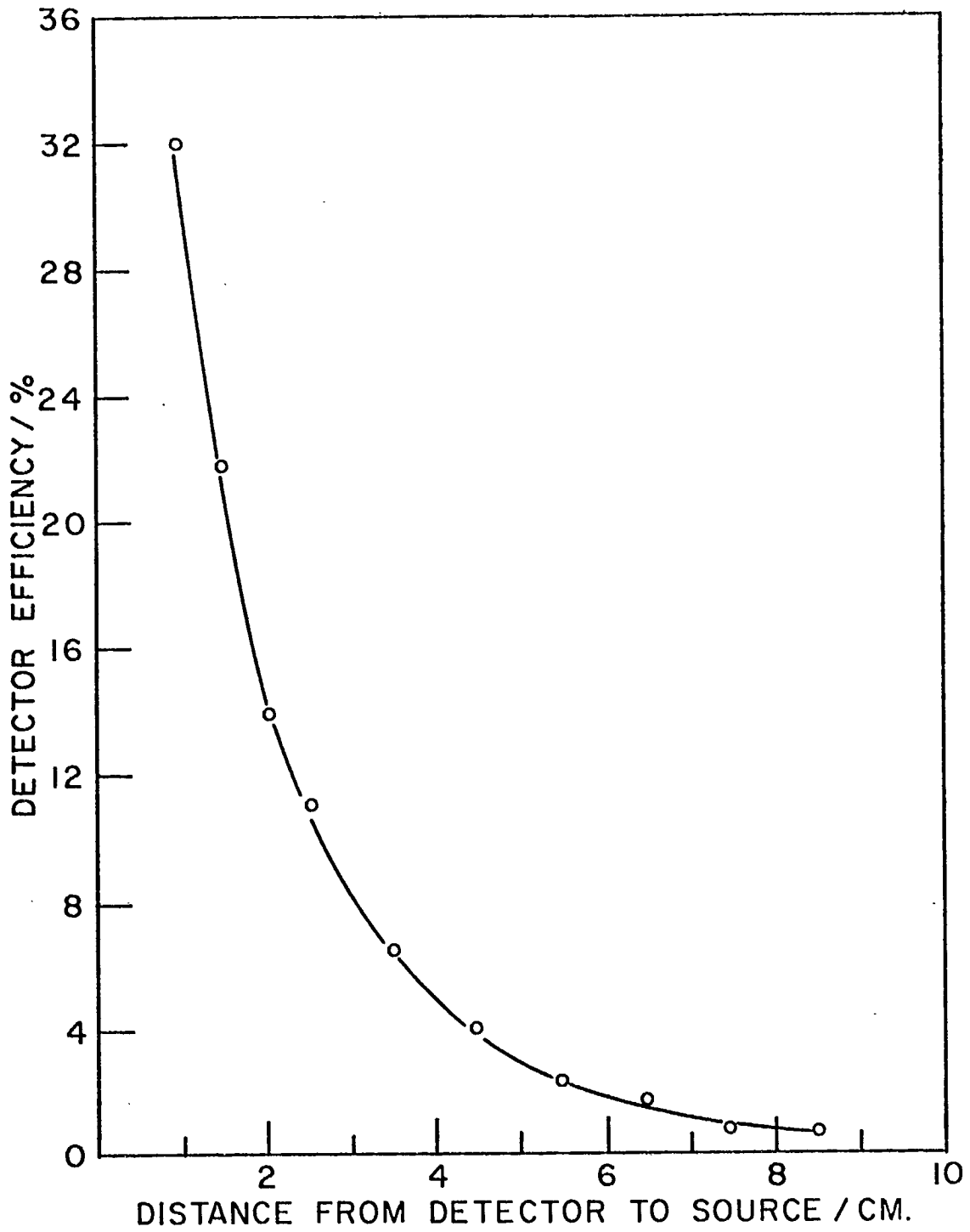
Figure 2. Variation of Detector Efficiency with Source-to-
Detector Distance.
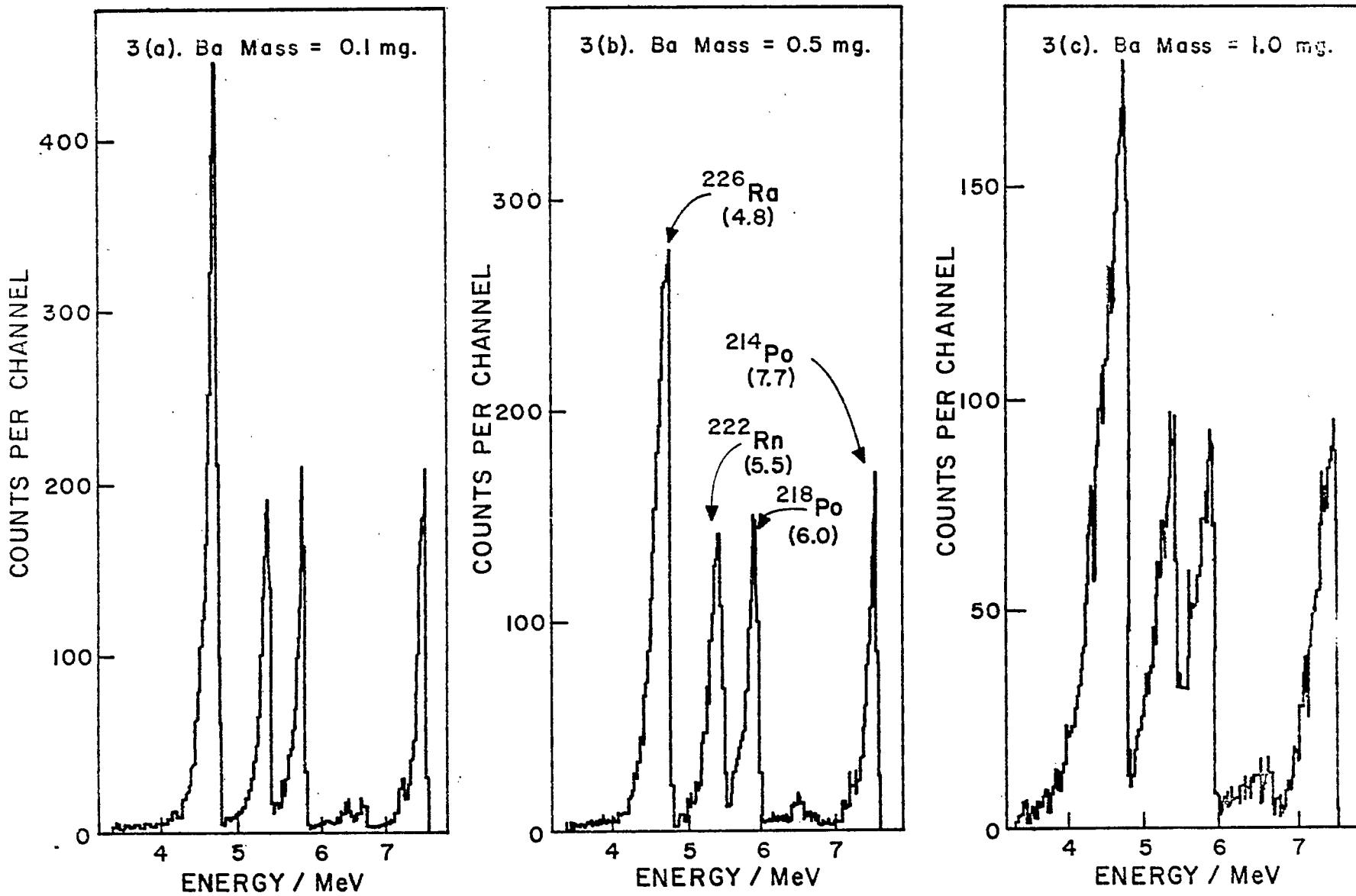
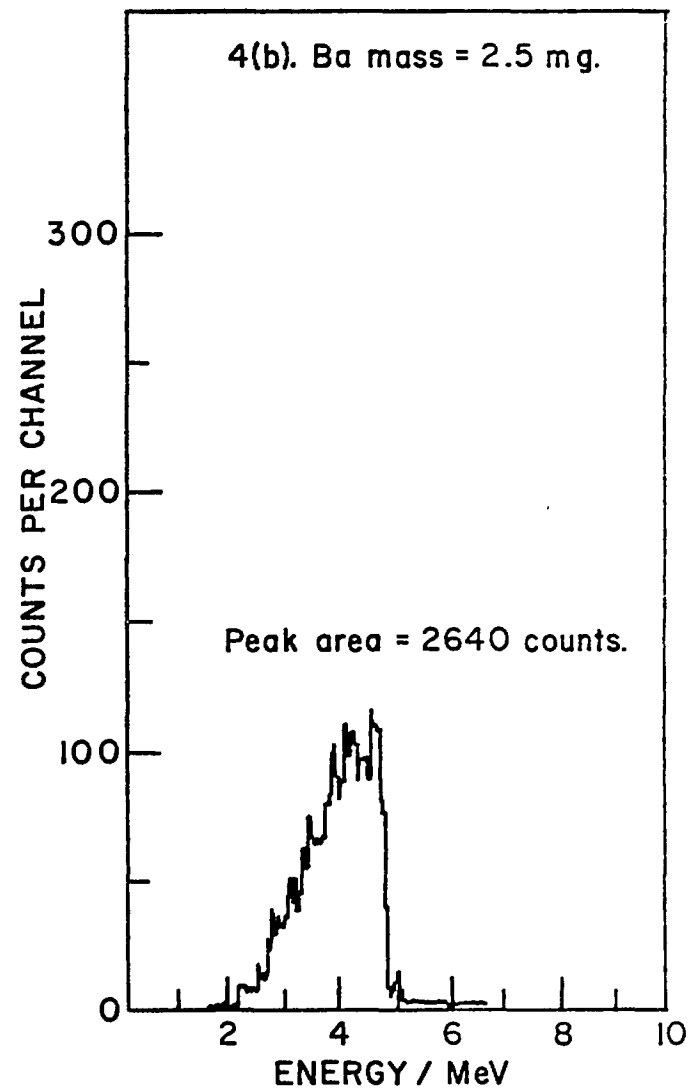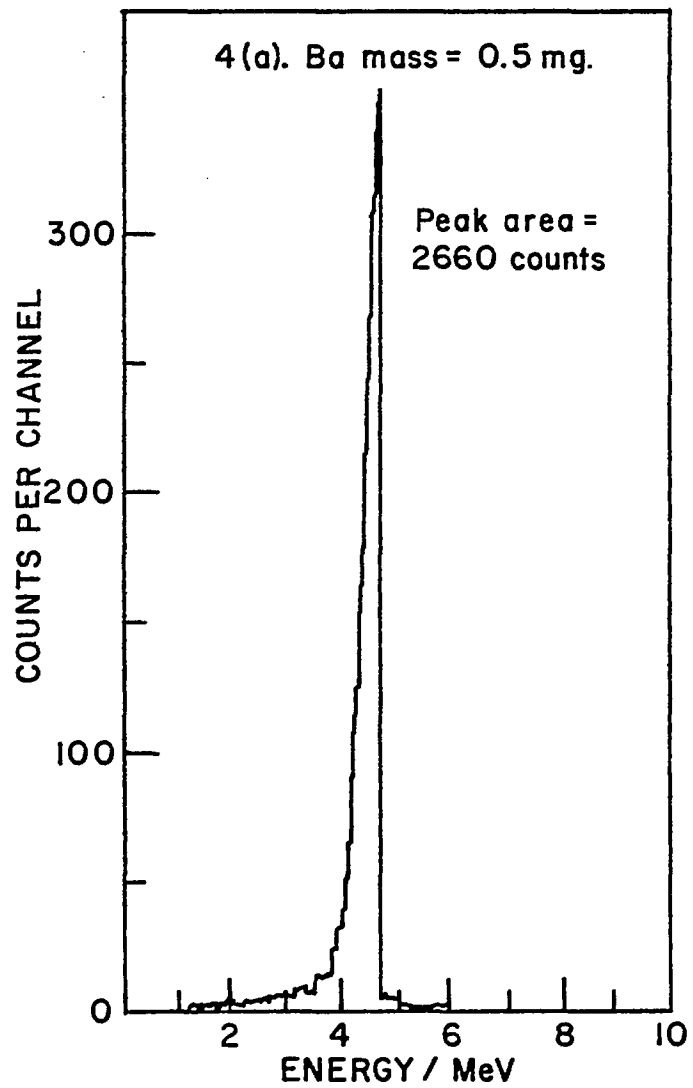Figure 3. Spectra Showing Effect of Barium Carrier Mass on Resolution.

(Count Times = 2000 s).

Figure 4.   Spectra of ^{226}Ra Obtained Using 0.5 mg and 2.5 mg of Barium Carrier.

(Count Time = 1000 s).
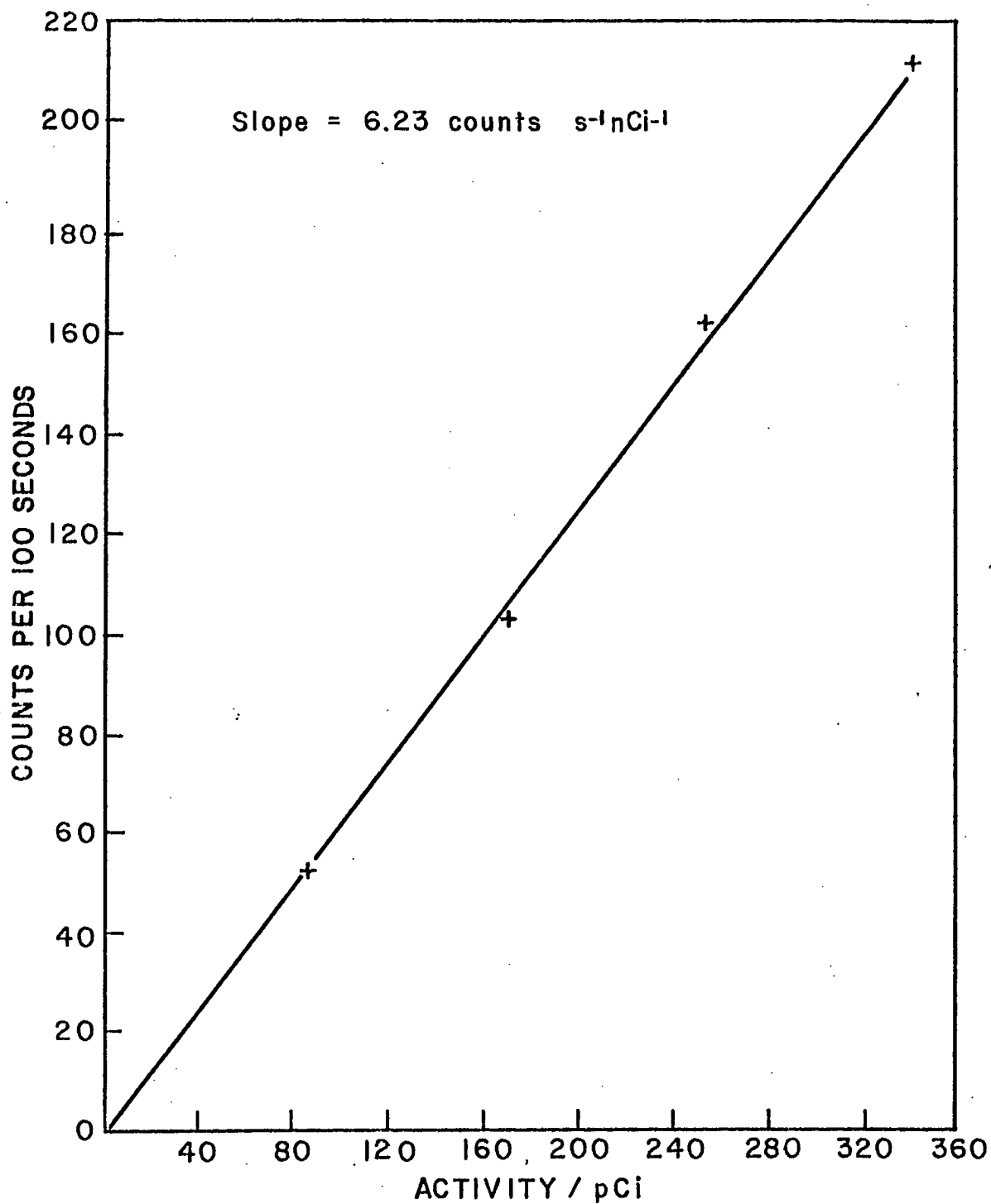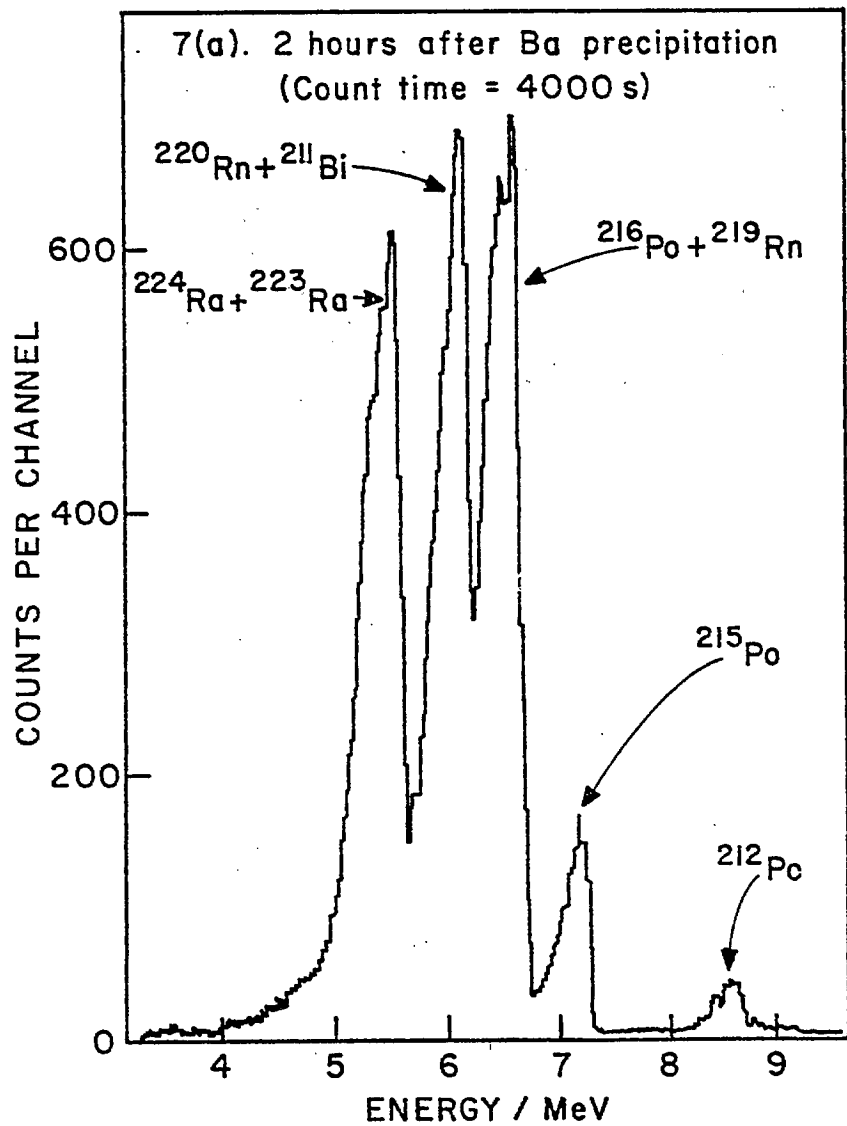
Figure 5. Calibration Curve for Alpha Activity.
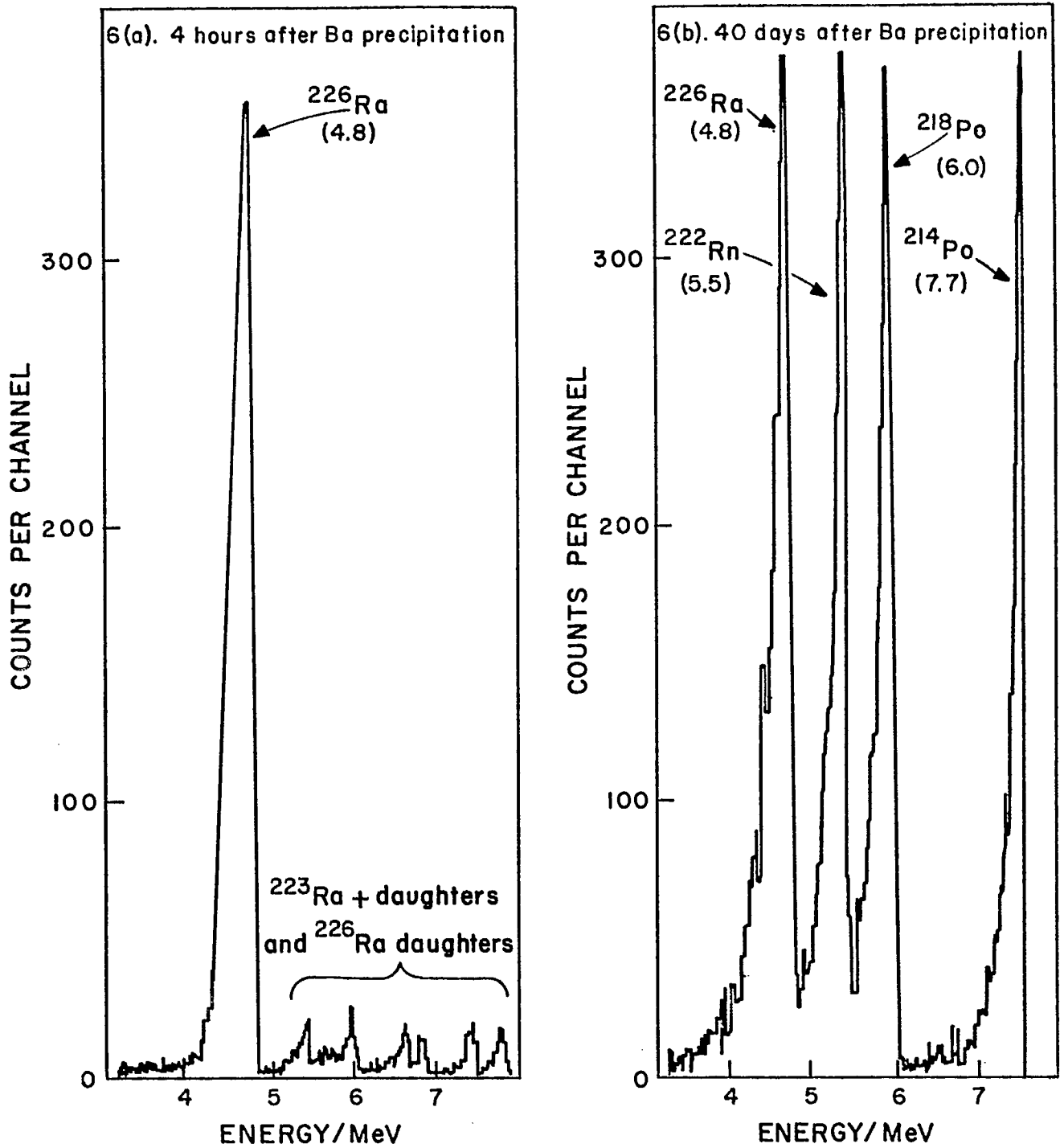
Figure 6. Spectra Obtained by Analysis of Old Residue from Nitric Acid

Leach. (Count Times = 2000 s).

7(a). 2 hours after Ba precipitation
(Count time = 4000 s)

$^{220}Rn + ^{211}Bi$

$^{224}Ra + ^{223}Ra$

$^{216}Po + ^{219}Rn$

^{215}Po

^{212}Pc

COUNTS PER CHANNEL

600

400

200

0

4   5   6   7   8   9

ENERGY / MeV

7(b). 3 days after Ba precipitation
(Count time = 5300 s)

$^{223}Ra + ^{224}Ra$

^{220}Rn

^{212}Bi

$^{211}Bi + ^{216}Po + ^{219}Rn$

^{212}Po

^{214}Po

COUNTS PER CHANNEL

600

400

200

0

4   5   6   7   8   9

ENERGY / MeV
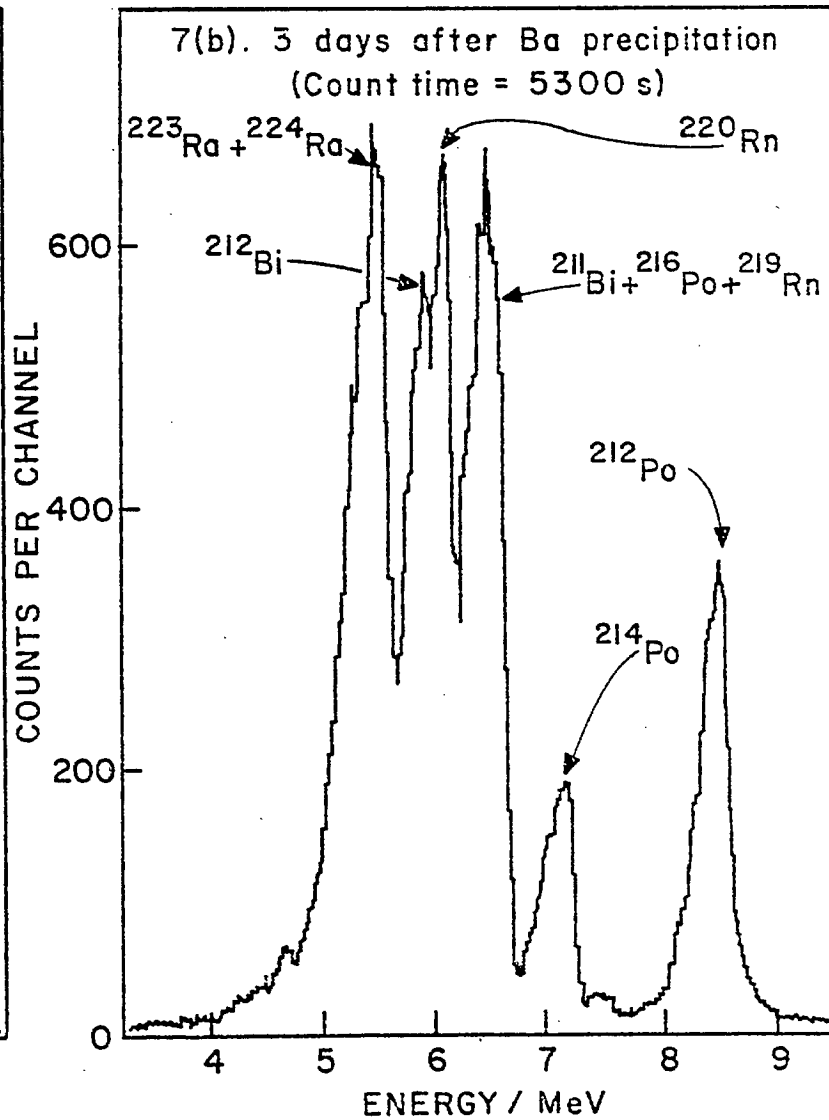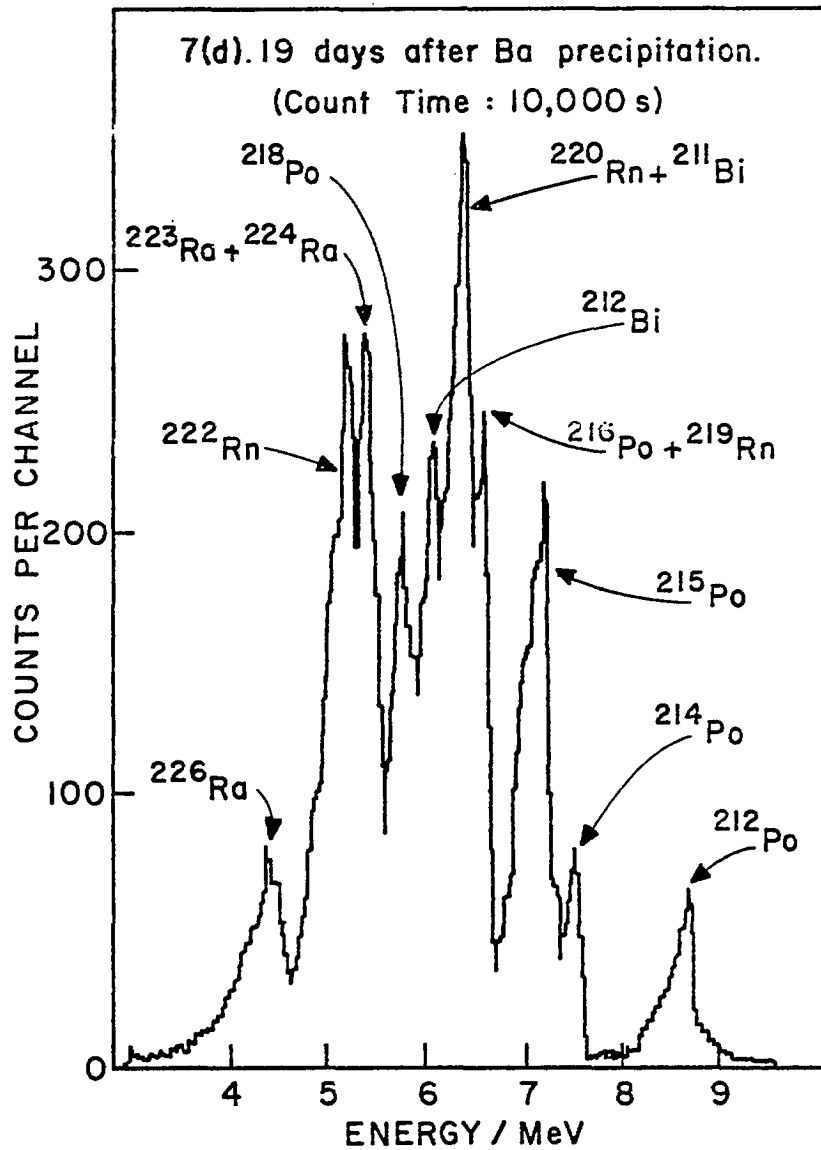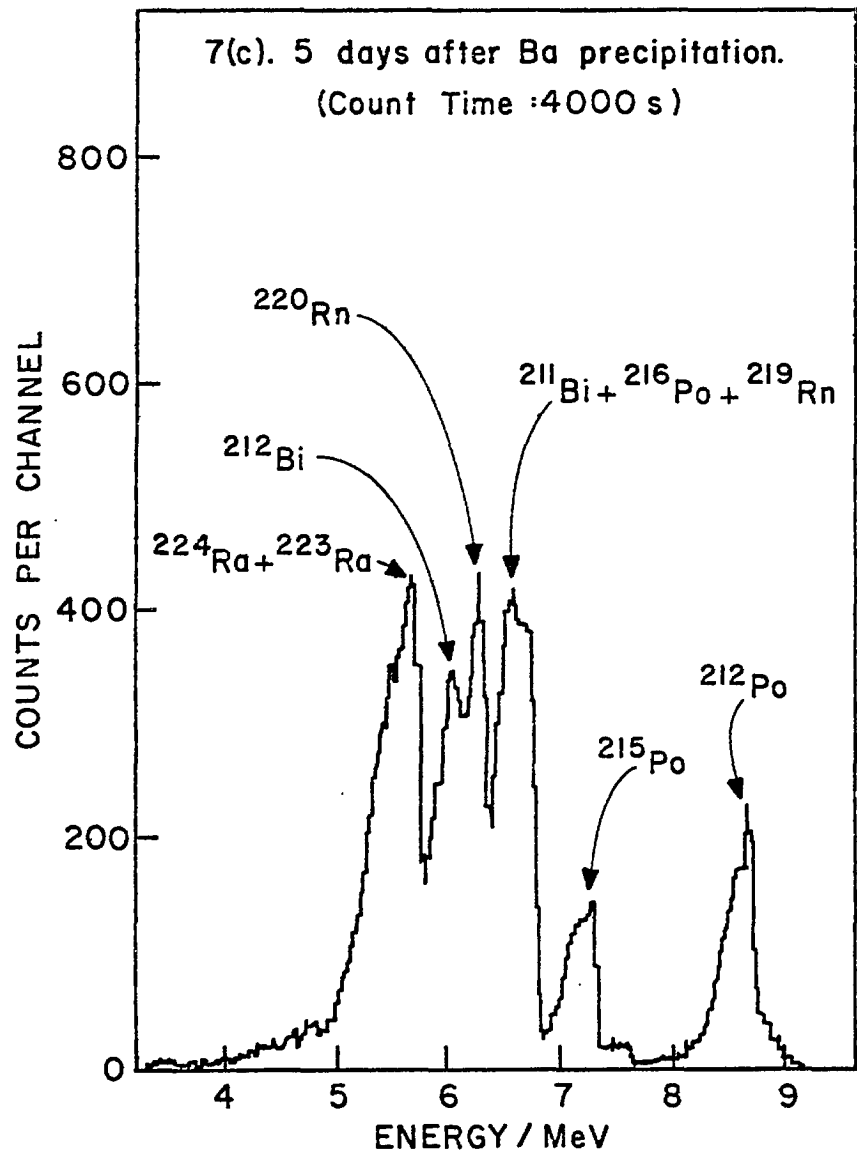
Figure 7. Spectra Obtained by Analysis of Old Liquor from Nitric Acid Leach.

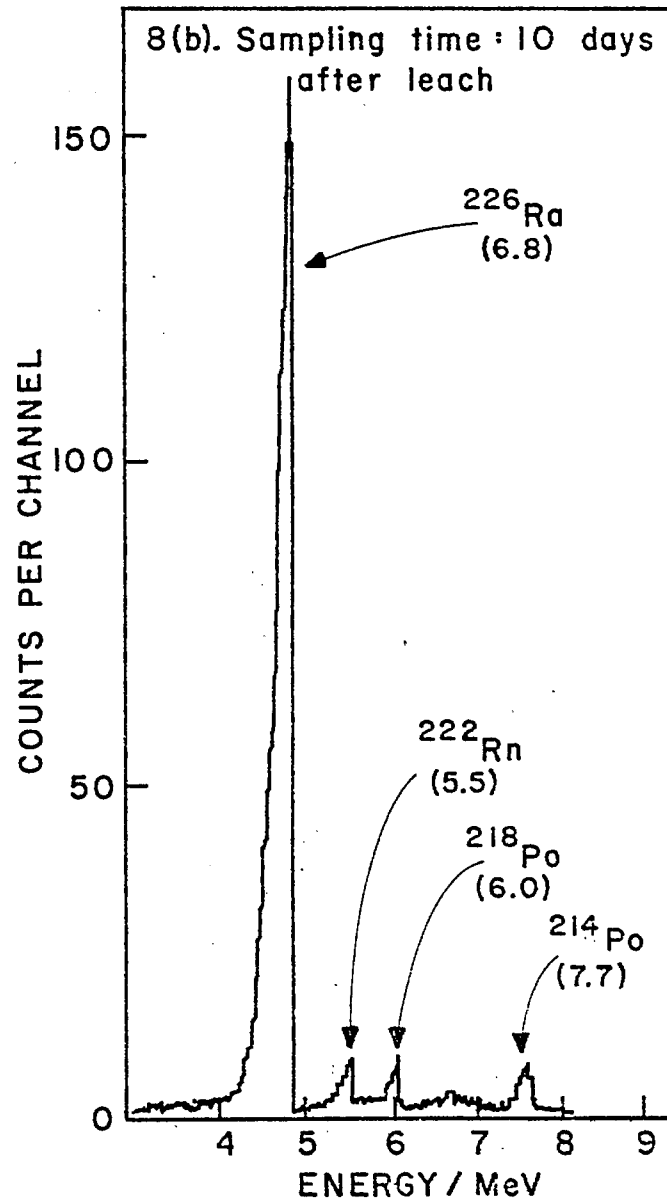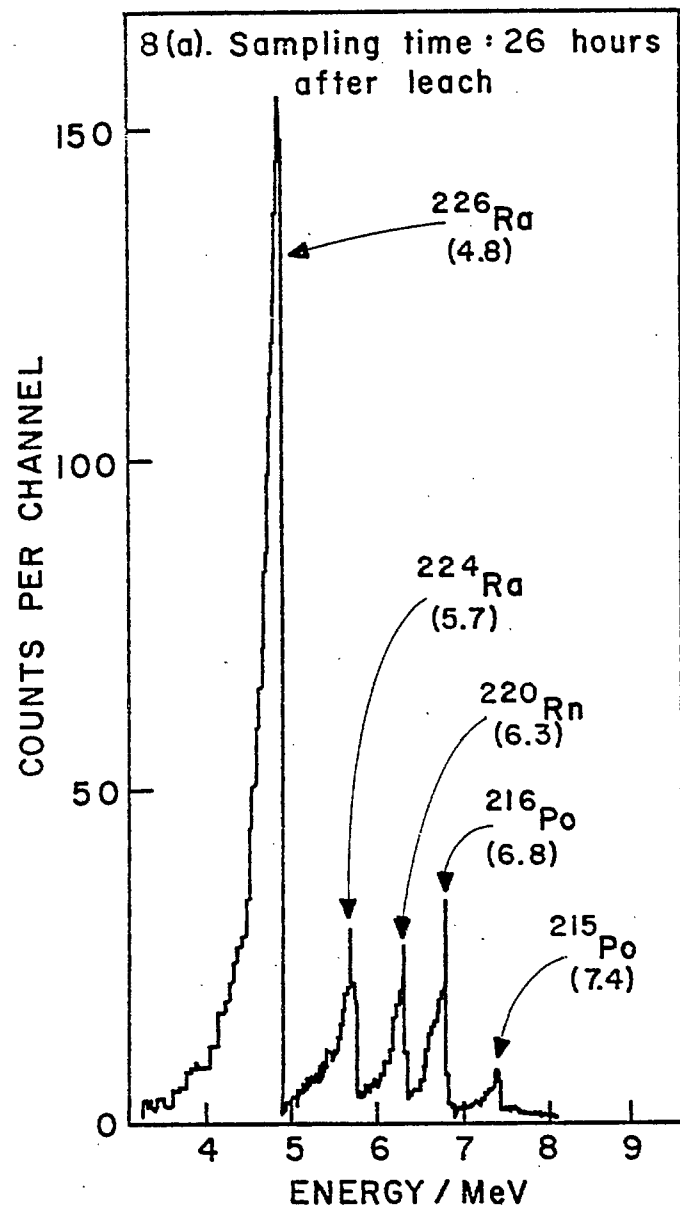(N.B. Different Count Times as shown with Spectra).

Figure 8. Spectra Obtained by Analysis of Fresh Residues from Nitric Acid Leach.
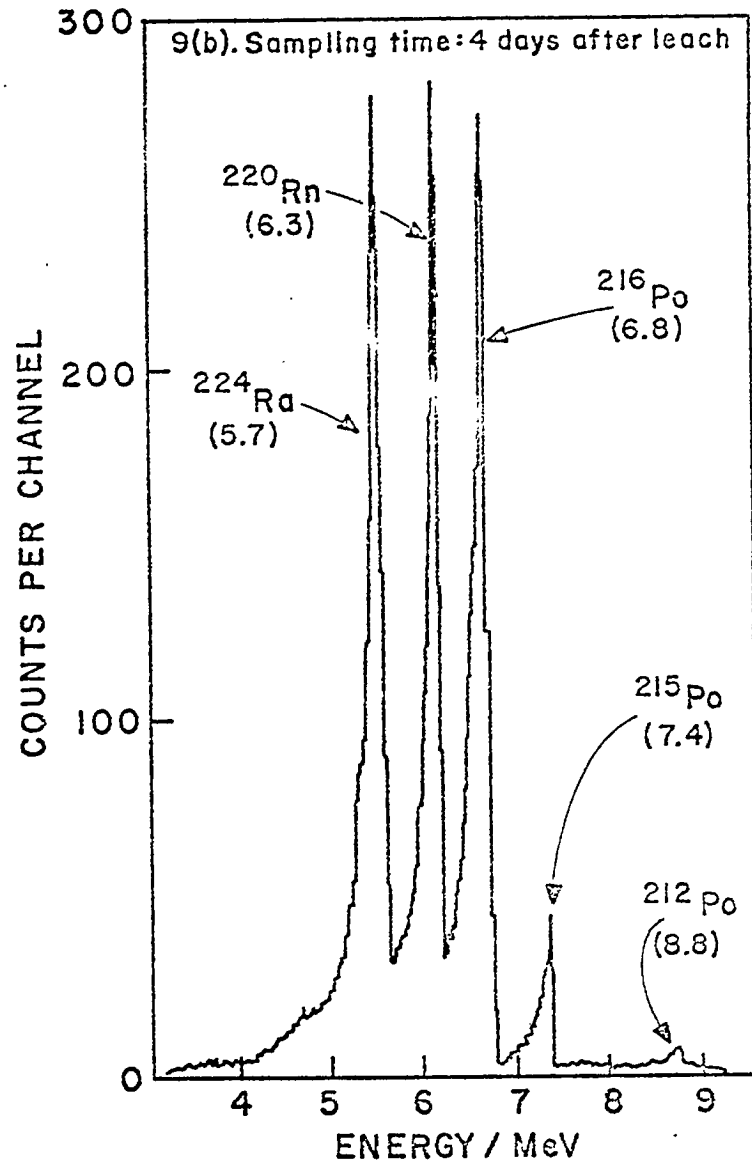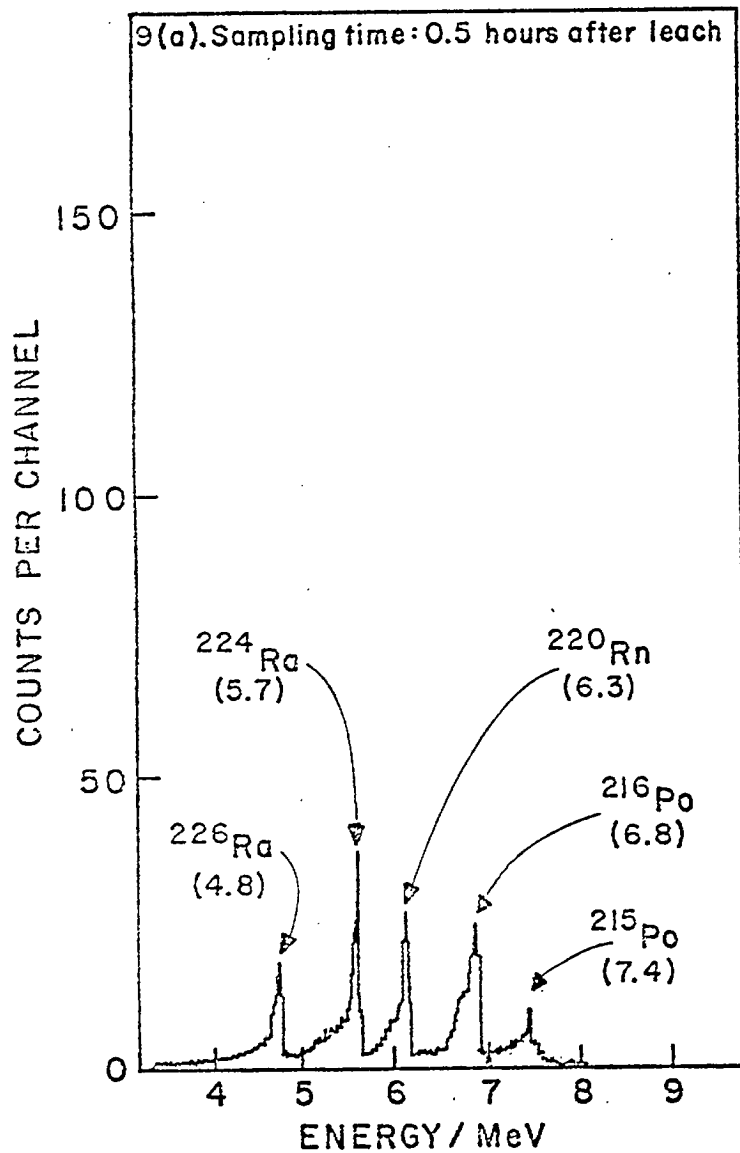
(Count Times = 2000 s).

Figure 9. Spectra Obtained by Analysis of Fresh Liquors from Nitric Acid Leach. (Coutn Times = 2000s).