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THE PREPARATION, PROPERTIES, AND USES OF AMERICIUM-241, ALPHA-, GAMMA-, AND NEUTRON SOURCES

J. E. Strain
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OAK RIDGE NATIONAL LABORATORY
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0.0 Abstract

This report deals with the preparation of alpha, gamma, and neutron sources using the long-lived radioisotope of Americium, Am^{241}. Americium-241 is an artifically-produced radioelement which has a half-life of 462 ± 10 years and decays to Np^{237} by alpha emission followed by low-energy gamma emission. The high specific activity of Americium-241 (7.0×10^9 d/m/mg) combined with its reasonably long half-life makes it ideally suited for the preparation of radioactive sources.

The chemical and physical properties of Am^{241} and the physical manipulations involved in fabricating alpha, gamma, and neutron sources are generally described in this report. Uses for each type of source are discussed and data are presented to indicate the respective properties and usefulness of each source type.

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1.0. Introduction

The adaptation of nuclear particles phenomena to the analysis of chemical systems offers many potentials for the development of unique analytical methods. As an outgrowth of experience in the Analytical Chemistry Division at Oak Ridge National Laboratory is promoting and devising analytical techniques based upon activation analysis, it was soon realized that many other nuclear methods of analysis were possible. It is expected eventually that many nucleonic methods of analysis will be common to the discipline of analytical chemistry.

The practicality of using a long-lived alpha- and gamma-emitting radionuclide in the preparation of intense sources of either alpha particles, low-energy gamma photons, or neutrons that can be utilized in a variety of analytical applications has been demonstrated. The results obtained from this study show that (1) high-intensity alpha sources, emitting equal to or greater than 10^{10} alphas per second, can be fabricated to investigate high-energy alpha reactions upon low Z number elements such as beryllium, fluorine, nitrogen, and boron; (2) low-energy gamma sources can be used in the radiography of low Z materials and in the quantitative determination of high Z elements in solution; and (3) low-intensity neutron sources, using α,n reactions on beryllium or boron, can be prepared and used in analytical techniques based upon radioactivation analysis, neutron absorptiometry, and neutron transmission.

Although a number of the radioactive isotopes of the transuranic series could have been used in an investigation of this kind, we found it convenient to consider the use of Am^{241}. Its radioactive decay characteristics, as described elsewhere in this report, are diverse enough to fulfill many of the general requirements of this investigation. Thus, this report shows that alpha, gamma, and neutron sources made from a radionuclide like Am^{241} can be utilized in a number of analytical techniques capable of practical use in solving analysis problems.

2.0. The Nuclear and Chemical Properties of Am^{241}

Americium, atomic number 95 and mass 241, was discovered in 1944 by Seaborg, et al,(1) when uranium-238 was bombarded with helium ions:

$$U^{238} + He^{4} \longrightarrow Pu^{241} + n \qquad (1)$$

Pu-241 decays with a 13.2 y half-life through negatron (β^-) emission to form Am-241.

$$Pu^{241} \xrightarrow[13.2\ y]{\beta^-} Am^{241} \qquad (2)$$

Currently, Am-241 is being produced in high-flux nuclear reactors in multigram quantities by successive neutron capture reactions upon Pu-239.

$$Pu^{239} + n \longrightarrow Pu^{240} + n \longrightarrow Pu^{241} \xrightarrow[13.2\ y]{\beta^-} Am^{241} \qquad (3)$$

Am-241 decays with a half-life of 462 ($\pm$ 10) years and with alpha and gamma emission to Np-237.(2) Figure 1 shows this mode of decay.

The chemical properties of americium are similar to those of the rare earth elements.(3,4) The only stable valence state in aqueous solution is the trivalent state. Am^{3+} shows typical rare earth reactions in that it will form insoluble precipitates of $Am(OH)_3$, AmF_3, and $Am_2(C_2O_4)_3$.(5) The quadrivalent state is observed chiefly in the form of the solid oxide, AmO_2,(4) a product of the pyrolytic decomposition of either $Am(NO_3)_3$ or $Am(C_2O_4)_3$. The Am^{5+} and Am^{6+} states are formed by oxidizing Am^{3+} with NaClO in a warm (90° C) solution of 2 M Na_2CO_3.(5) Both Am^{5+} and Am^{6+} are rapidly reduced in aqueous solution by the radiolytic decomposition products formed in the solvent by alpha-particle ionization.

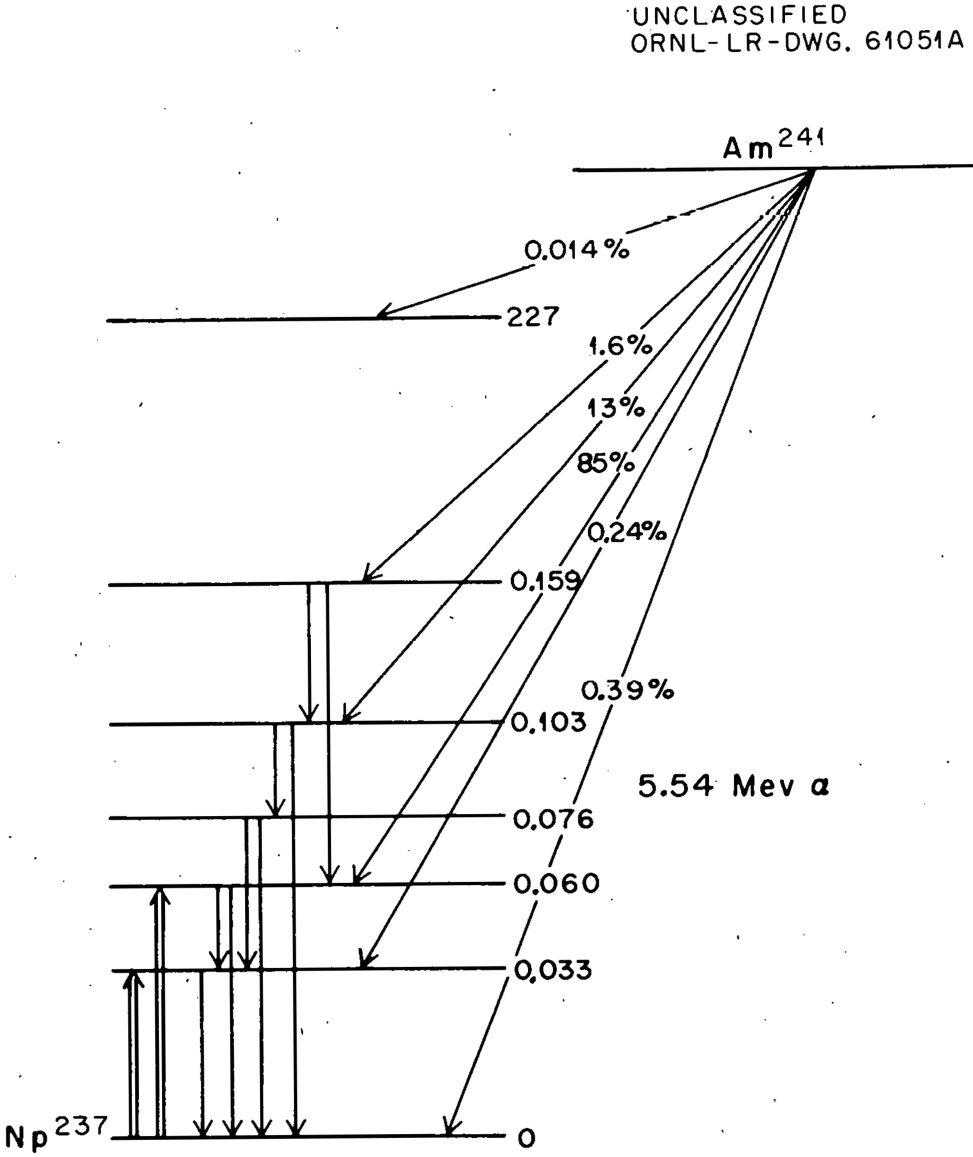

Fig. 1. Decay Scheme of Am^{241} (Phys. Rev. Vol 30, p 286, April 1958).

The nuclear properties of Am^{241}, i.e., its half-life and radiation characteristics,(2) were considered most favorable for its use in this particular investigation. The half-life of 462 years is sufficiently long to preclude frequent decay corrections and yet is short enough to yield a specific activity of 3.17 curies per gram, or 7.037×10^9 alpha disintegrations per minute per milligram of Am^{241}. The low-energy gamma rays of 0.0597 Mev* which accompany its alpha decay are ideally suited for radiographic or density measurements, yet they are sufficiently weak so that they can be completely shielded with any high Z absorber. The 5.49-Mev alphas serve as an excellent initiating energy source in (α,n) reactions to produce either neutrons or high-energy capture gammas in the light elements.

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*For convenience, a value of 60-kev is used throughout this report to express the energy of this gamma photon.

3.0. The Chemical Purification of Americium

The Am^{241} solution received for use in these studies contained 0.1% by weight of plutonium and 16% by weight of stable contaminants (9% rare earths, 3% iron, 2% chromium, and 2% sodium-nickel-copper). Although the radioactive purity was greater than 99%, it was necessary, in order to provide maximum specific activity, to purify the americium chemically prior to its use in gamma and alpha sources. The method which combined both ease of operation and acceptable purification was a modification of a process involving ion exchange with Dowex-1 resin and 5 $\underline{M}$ NH_4SCN elution.(6) In our purification method, the ion exchange separation was followed by extraction of the americium from the eluate into di-(2-ethyl hexyl)phosphoric acid (HDEHP). The procedure used was as follows:

1. An aliquot of the stock solution, containing approximately 50 milligrams of Am^{241}, was transferred to a 50-milliliter glass centrifuge cone and the Am^{241} precipitated as $Am(OH)_3$ by adding concentrated NH_4OH dropwise to the solution. The NH_4OH addition was continued until no further precipitation of the $Am(OH)_3$ occurred.

2. The mixture was then centrifuged and the supernatant liquid removed by decantation. The $Am(OH)_3$ precipitate was washed twice by stirring it in a small volume of 0.1 $\underline{M}$ NH_4OH solution. After each wash, the mixture was centrifuged and the wash liquid discarded.

3. The $Am(OH)_3$ was then dissolved in a few milliliters of purified 5 $\underline{M}$ NH_4SCN (NOTE: The NH_4SCN can be purified by passing it through a Dowex-1 resin column.)

4. The Am^{241}-NH_4SCN solution was then transferred to the top of a 2.5 x 25 cm Dowex-1 ion exchange resin column. (NOTE: 200-mesh Dowex-1 resin was conditioned with purified 5 $\underline{M}$ NH_4SCN.)

5. The Am^{241} was purified from other elemental species by eluting them from the column with the 5 M NH_4SCN solution. The flow rate used for elution was 1 milliliter per minute and the eluate was monitored for Am^{241} radioactivity (see Appendix A). As soon as the Am^{241} began to elute, the NH_4SCN elution was discontinued and the column stripped of the Am^{241} by eluting with a 2 M NH_4Cl solution at a flow rate of 0.5 milliliter per minute (NOTE: The use of 2 M NH_4Cl gave the most rapid and nearly complete elution of Am^{241} from the column. It proved superior to various molarities of nitric and hydrochloric acids as well as lower and higher concentrations of both NH_4Cl and NaCl. Weakly basic solutions of the complexing agent, Versene, also proved to be less effective).

6. At least 600 milliliters of the eluate were collected in a 1-liter glass separatory funnel. Three hundred (300) milliliters of 1 N di-(2-ethyl hexyl)orthophosphoric acid-hexane mixture was then added to the funnel and the mixture shaken for five minutes.

7. After shaking, the phases were allowed to separate and the aqueous phase drained off and discarded. The organic phase was then washed at least twice by adding an equal volume of water and shaking the mixture for five minutes. After each wash operation, the phases were allowed to separate and the aqueous phase drained off and discarded.

8. One hundred milliliters of 6 M HNO_3 were then added to the organic phase in the funnel and the mixture shaken for five minutes. After shaking, the phases were allowed to separate. The aqueous phase was drained into a new separatory funnel and the organic phase discarded.

9. An equal volume of hexane was then added to the aqueous phase in the separatory funnel and the mixture shaken for five minutes. (NOTE: The hexane wash will remove any traces of the di(2-ethyl hexyl)orthophosphoric acid from the 6 M HNO_3 solution.) After shaking, the phases were allowed to separate and the 6 M HNO_3 solution of Am^{241} (the aqueous phase) drained into a storage bottle. The organic phase was discarded. (NOTE: The Am^{241} solution is now essentially free of its original contaminants and may be evaporated or diluted to the concentration desired.)

The above separation procedure gives decontamination factors of at least 10^5 for the rare earths, plutonium, and sodium and results in a recovery of Am^{241} of the order of 90%. It is believed that the 10% loss occurs in the ion-exchange column separation because Am^{241} elutes only very slowly with any eluting agent. If the same column is used for processing additional amounts of americium, the retained americium is observed to slowly elute from the column at the rate of about 100 micrograms per liter of 5 M NH_4SCN. The behavior of stable Na^{23} and La^{139} in this system was studied by the use of 15 h - Na^{24} and 40 h - La^{140} radioactive tracers. Figure 2 shows this behavior.

Several other methods for americium purification are described elsewhere.(7-9) However, the method reported here for Am^{241} purification gives the best possible decontamination from plutonium and the rare earths and is easily adaptable to semi-remote glove box operation.

The elution of Am^{241} radioactivity from the resin column can be monitored in several ways. In our initial work, we used a Geiger-Mueller counting tube and a linear count rate meter to monitor the radioactivity of both the Am^{241} and the radioactive tracers used in the decontamination studies. In later efforts, a multichannel pulse-height analyzer and a NaI(Tl) crystal were used to observe the radioactivity of the solution and to identify the radionuclides. A third way, i.e., counting of the Am^{241} alpha radioactivity, was not considered. This particular analysis technique would have been laborious and time-consuming and the high NH_4SCN concentration in the solution would result in undesirable solids which could interfere in the gross alpha measurements. More details about the column monitors and radioactivity assay techniques are given in Appendix A.

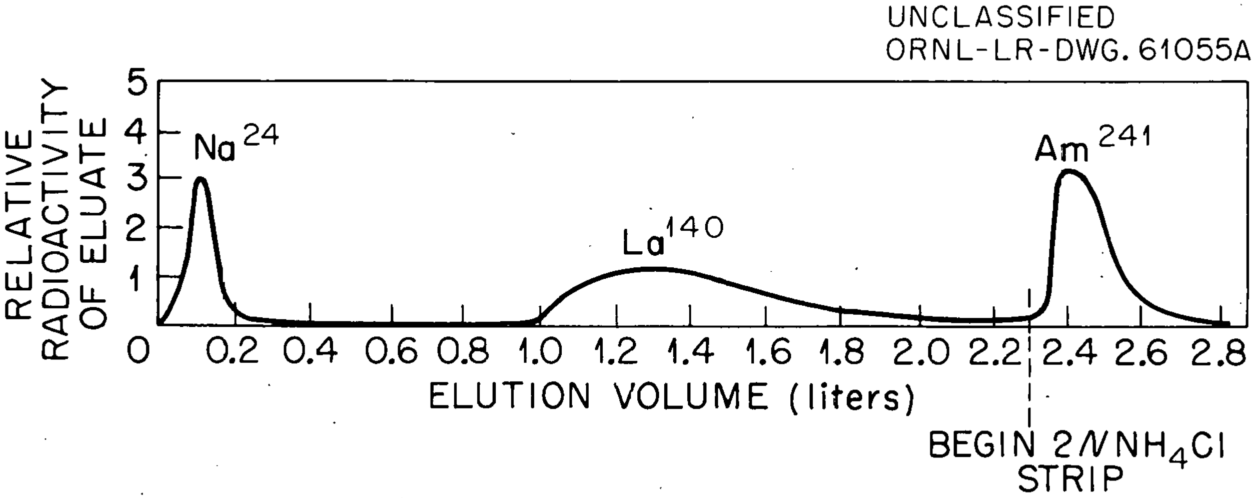

Fig. 2. Dowex-1 Ion Exchange Purification of Americium. (Elution with 5 N NH_4SCN, flow rate 50 ml/hr.)

4.0. Safety Requirements for Am^{241} Source Preparation

Due to the high specific ionization effects of alpha radiation, the processing of solutions containing large amounts of alpha radioactivity presents a serious handling and containment problem. It is essential that all operations, involving the purification of alpha-containing materials and the subsequent use of the alpha radioactivity in such investigations as chemical properties, source fabrication, etc., be carried out within a sealed-glove box in which a slight negative pressure is maintained. In addition to the alpha hazard, the 60-Kev gamma radiation necessitates shielding of the separation equipment if appreciable quantities of Am^{241} are to be handled. In this investigation, all of the proposed types of sources - alpha, gamma, and neutron - had to meet rigid standards of safety based on the following criteria:

1. A positive containment of the radioactive material. Any leakage of the large quantities of alpha radioactivity present in the source could result in a serious health hazard.

2. A rugged containment vessel to preclude accidental rupture of the source through rough handling.

3. A simple method of source sealing in order to avoid excessive radiation exposure to personnel during source fabrication.

4. The development of storage, use, and monitoring procedures which would minimize gamma or neutron exposure to personnel as well as minimize the possibilities of alpha contamination (see Appendix B).

Preparation of the alpha and gamma sources presents special problems, because the source "windows" must be thin enough for efficient radiation transmission and still meet safety requirements. Detailed information on the preparation of each type of source follows.

5.0. Preparation and Uses of Am^{241} Alpha Sources

Since Am^{241} decays with the emission of 5.49-Mev alpha particles, these radiations may be used in a number of analytical applications.

Source Preparation: At least four techniques for alpha source preparation were evaluated in this investigation. The first method, i.e., by electrodeposition,(10,11) was considered because of its potentially high alpha particle yield and uniform deposition. Although this method has proven superior for fabricating low-level sources, it was found that it was impossible to electrodeposit greater than 100 micrograms of Am^{241} per cm^2 of area with acceptable physical properties. Our second choice involved the evaporation of $Am(NO_3)_3$ solutions on a stainless steel plate. In the alpha radioactivity levels desired, this procedure resulted in a nonuniform spongy deposit. Attempts to ignite and compact the oxide resulted in large mechanical losses and additional nonuniformity. The third method attempted was a slurry technique in which an ether or alcohol slurry of $Am_2(C_2O_4)_3$ was allowed to settle on a stainless steel plate and air dried. A spongy but uniform layer was formed; however, it was also nonadherent. The fourth method requires a reduction of AmF_3 at a high temperature in the presence of barium metal and the subsequent evaporation of the americium metal onto a tantalum or tungsten metal plate. For our reduction of the AmF_3, we have modified the equipment described by Westrum and Eyring(12) to make use of an induction furnace rather than a tungsten heater filament. This has allowed us to use existing equipment and also to handle larger quantities of Am^{241} and barium metal. This method, while requiring special equipment, can result in the production of an alpha plate of high specific alpha activity per cm^2 of plate area with excellent adherence and freedom from nonradioactive contaminants.

Encapsulation of Am^{241} Alpha Sources: Since safe encapsulation of alpha sources had to be considered, several tests were carried out to determine the feasibility of various materials for use as source covers. For these tests, low-level sources were prepared by aliquotting one microgram or less of Am^{241} in a HNO_3 solution onto a stainless steel plate. A heat lamp was used to evaporate the solution and then the plate flamed to a dull redness in order to destroy the nitrates. These sources were counted on a gas-flow proportional alpha counter to measure their radioactivity, then covered with a plastic film or a metal foil and recounted to determine the percent alpha transmission. Organic films of materials, such as mylar, zapon, and polyethylene, were considered first. While they gave excellent transmission (> 90% transmission for 0.1-mil polyethylene film), they were found to be unstable to high intensity alpha radiation. One source containing 0.20 gram of AmO_2 covered with 0.1 mil polyethylene film was found to develop large holes in the covering in less than eight hours after its preparation; eventually, the polyethylene was completely destroyed through radiation damage. With the failure of organic materials, a search was begun for a metal covering which was absolutely leak tight yet had the optimum transmission characteristics. The data presented in Figure 3, obtained by using a Frisch Grid Chamber for the radioactivity measurements, compares the observed alpha energy and alpha particle emission rate for covered and uncovered Am^{241} sources. The transmitted integral alpha count through the 0.1-mil thick nickel foil covered mount was 72% of

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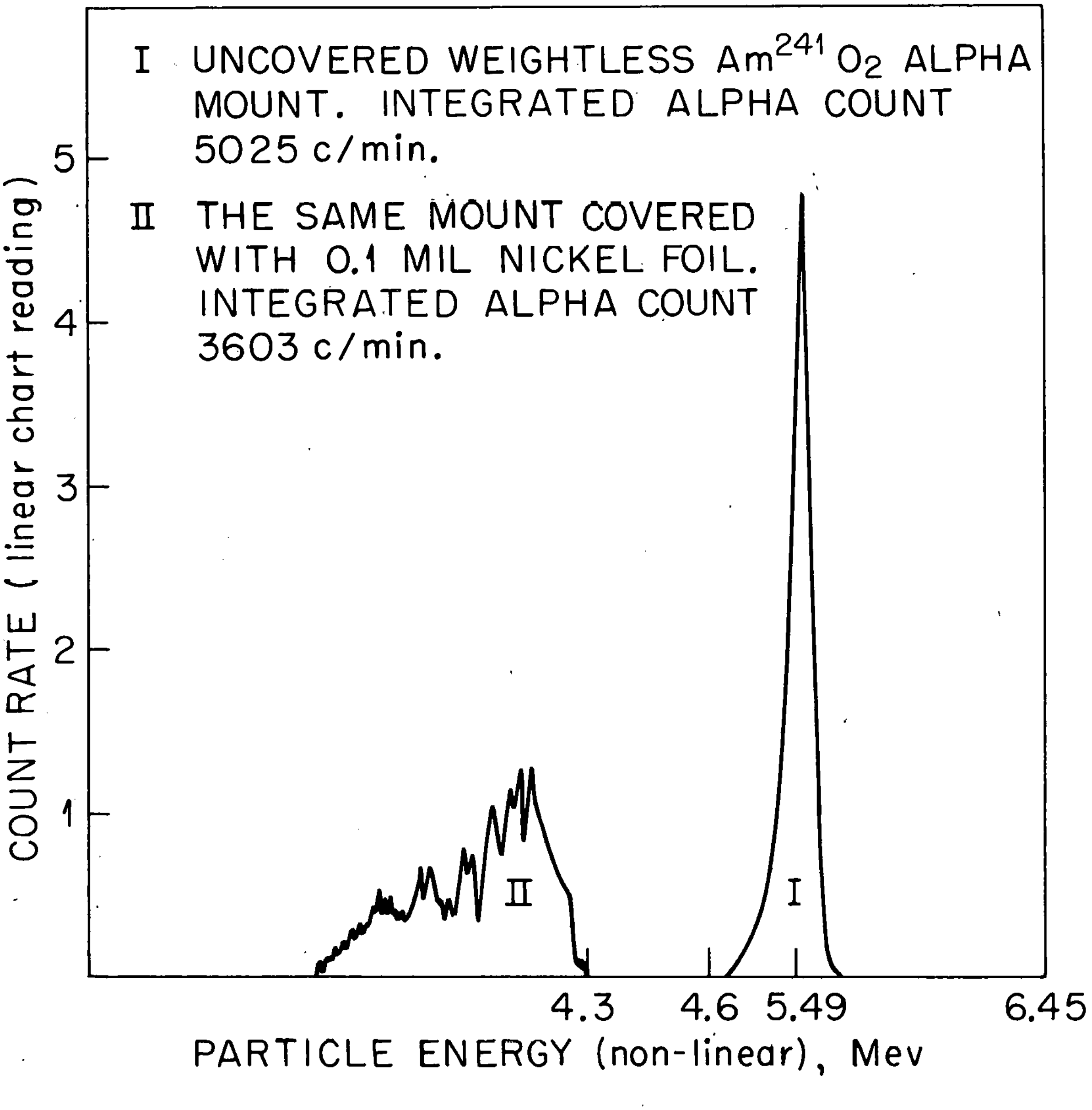

Fig. 3. Frisch Grid Chamber Measurement of Emitted Alpha Particle Energy from a Covered and Uncovered Mount.

the uncovered mount, while the average alpha energy decreased from 5.49 Mev to approximately 4 Mev due to the energy loss in the covering material. However, this is still sufficient energy to initiate alpha-capture reactions in elements below nitrogen (i.e., reactions involving alphas of 4.4 Mev or less); therefore, it was concluded that thin nickel foils could be used to cover high intensity alpha sources.

Am^{241} Alpha Source Applications: One objective of our work with alpha sources was to produce high specific activity sources that could be used in highly sensitive, safe, continuous air monitors for the detection of air-borne beryllium dusts. Reiffel(13) has already demonstrated the feasibility of using a Po^{210} alpha source and the following reactions in such an application.

$$Be^{9} + \alpha \longrightarrow C^{12*} + n^{o} \qquad (4)$$

$$C^{12*} \longrightarrow C^{12} \text{ (stable)} \qquad (5)$$

where C^{12*} is a highly excited state of stable carbon, C^{12}, that instantaneously emits prompt-gamma radiations following its formation. The radiation energies of these prompt gammas are 3.2, 3.6, and 4.5 Mev, respectively. Unfortunately, the use of 138-day Po^{210} necessitates frequent decay corrections and limits the usefulness of the method. The use of long-lived Am^{241} instead of Po^{210} would greatly enhance the stability of an alpha source for use in monitors of this type. It is anticipated that Am^{241} alpha sources will be used in equipment such as that shown in Figure 4 and 5 to continuously monitor air-borne beryllium content.

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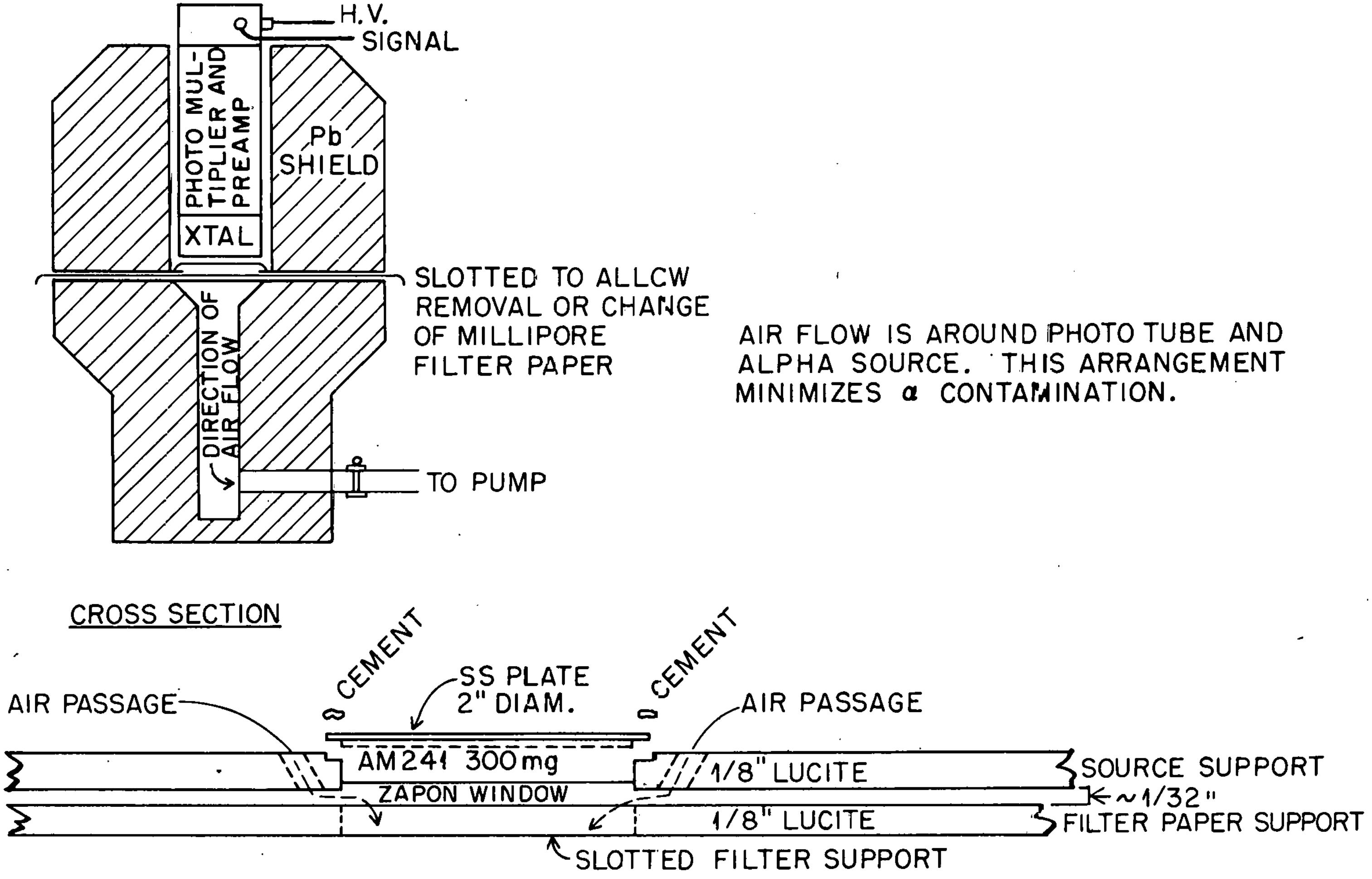

AIR FLOW IS AROUND PHOTO TUBE AND ALPHA SOURCE. THIS ARRANGEMENT MINIMIZES α CONTAMINATION.

Fig. 4. Source Mounting Detail for Air Borne Be Monitor.

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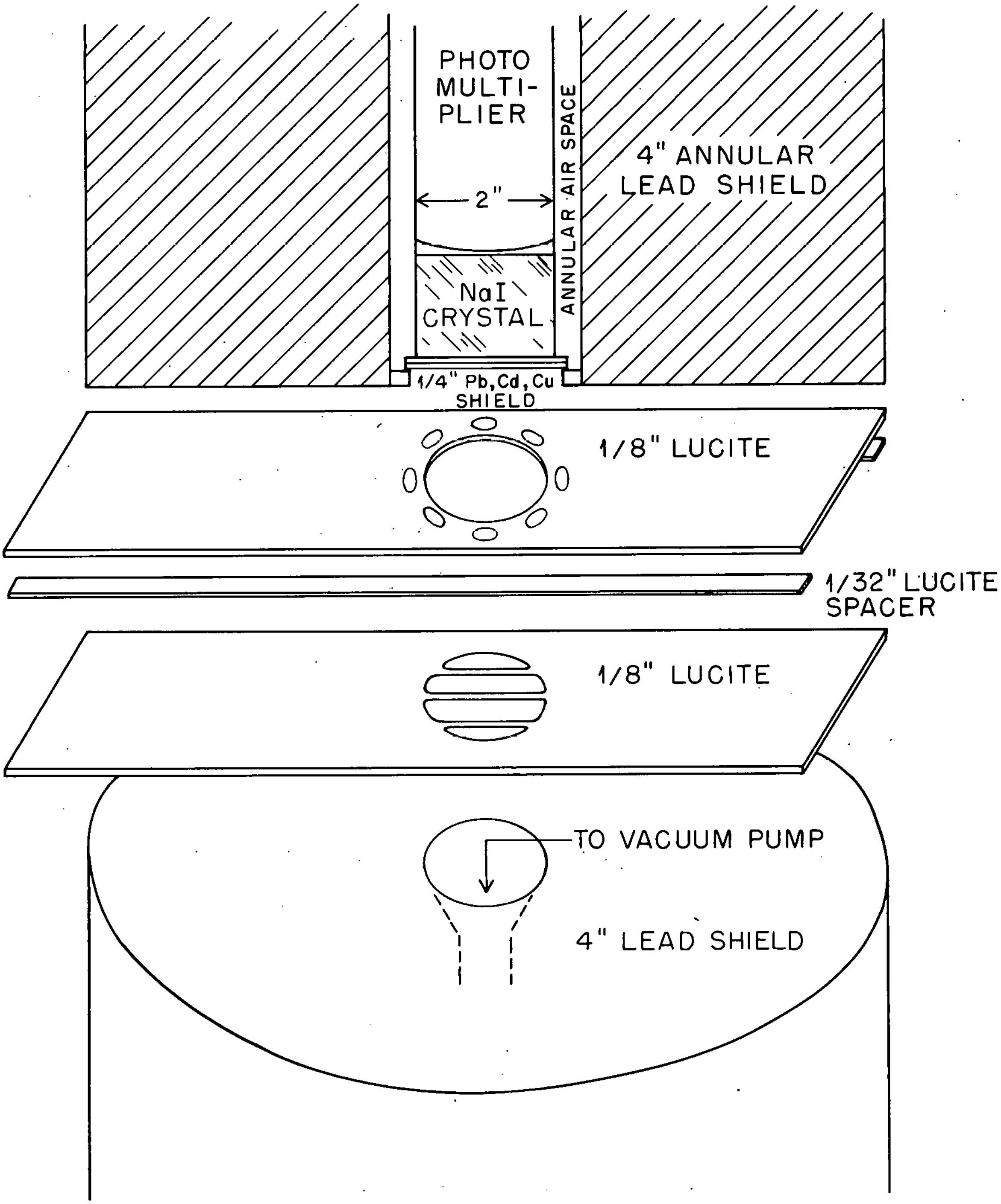

Fig. 5. Exploded View of Counter Source Holder and Filter Paper Guide and Support.

As part of this work, we have demonstrated the use of these reactions in the determination of microgram amounts of beryllium. In these experiments, solutions containing 12 milligrams of Am^{241} were mixed with solutions containing varying quantities of beryllium. In each test, the final solution volume was 3.0 milliliters. This was placed in a sealed polyethylene vessel and the vessel mounted on the top of a solid 3" x 3" NaI(Tl) scintillation crystal for the radioactivity measurements. The count rate of the instantaneous gamma radiations being emitted as a result of the $Be^9(\alpha,n)C^{12*}$ reaction was recorded for four minutes by means of a 20-channel pulse height analyzer. In each test, only the gamma radiations above 3 Mev were recorded. Figure 6 shows the relationship of the count rate of the gamma radioactivity above an energy of 3 Mev to the quantity of beryllium added to the $Am(NO_3)_3$ solution.

In a series of other tests, it was shown that when the Be-Am^{241} solutions were evaporated to dryness, the specific activity per milligram of beryllium increased by at least a factor of 50. For example, solutions containing <u>10 micrograms</u> of beryllium when evaporated with solutions containing <u>12 milligrams</u> of Am^{241} gave a count rate of 50 counts per minute. However, the evaporation technique is very sensitive to impurities in that the alpha interactions with beryllium can be decreased by the presence of non-evaporable solids. Thus, it is considered necessary to minimize these interferences by performing at least a partial separation of beryllium on any sample to be analyzed by this method.

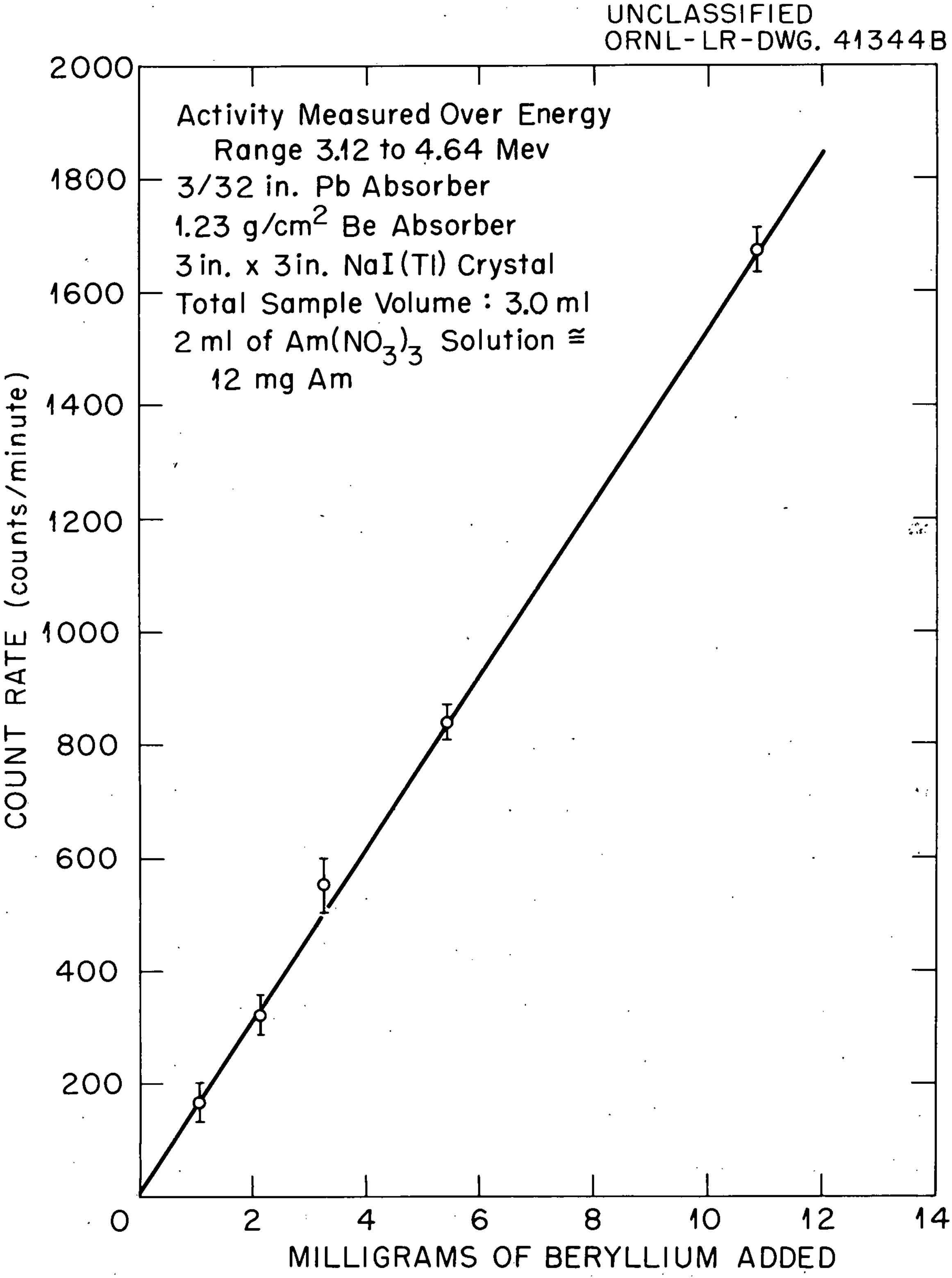

Fig. 6. C^{12*} Prompt Gamma Radioactivity vs Beryllium Concentration.

6.0. Preparation and Uses of Am^{241} Gamma Sources

In this part of our investigation, Am^{241} gamma sources were prepared in order to use the 60-kev gamma radiations emitted by Am^{241} in its decay for analysis applications.

Source Preparation: Since radioactive purity is a primary factor influencing the performance of a gamma source and chemical purity is of only slight consequence, no further purification of the original Am^{241} solution was necessary. In preparing a gamma source, an aliquot of the stock solution containing the desired quantity of Am^{241} was transferred to a 50-ml centrifuge cone and $Am(OH)_3$ precipitated by the addition of concentrated NH_4OH. The $Am(OH)_3$ was removed from the mixture by centrifugation, washed with water, and finally ignited in a platinum crucible to AmO_2. The AmO_2 was then ground in a mortar to a fine powder. Prior to its encapsulation, the amount of Am^{241} in the powder was determined by measuring its specific activity by means of an alpha counter.

Encapsulation of Am^{241} Gamma Sources: In order to determine the intensity of the 60-kev gamma photons necessary for a particular analysis application, one must consider the geometry of the source arrangement, the abundance and internal conversion of the desired photon, and the degree of absorption of the photons in the source materials. Using thin sources and thin windows, the total number of gamma photons emitted from the source will approach 2π. The abundance of the 60-kev gamma is seen from the decay scheme (Figure 1) to be only about 40% and of this number nearly half of the photons are internally converted. Measurements on a 3" x 3" NaI(Tl) crystal and multichannel gamma spectrometer indicate that the effective 60-kev gamma to alpha ratio is 0.34 (see Appendix C).

Curves showing the differential absorption of the Am^{241} gamma radiations in various thicknesses of stainless steel are presented in Figure 7.

The Am^{241} gamma source container found to be most satisfactory from the standpoint of ruggedness, ease of sealing, and freedom from leakage is shown in Figure 8a. Fabricated from 347 stainless steel, it is 0.69 in. high by 1.0 in. in diameter, and it can be used to contain up to 0.25 gram of Am^{241} as the oxide. At this point, the self-absorption of the 60-kev radiation in the AmO_2 causes no increase in photon emission when additional amounts of Am^{241} are placed in the capsule. A source window of 0.010-inch thick stainless steel (347) is used to contain the AmO_2 powder. This window thickness will cause a 37% decrease in the available photons (theoretical yield $\sim 5 \times 10^8$ photons/sec). In order to get higher photon yields, a larger stainless steel (347) source holder 0.32-inch x 1.75-inch with a 0.020-inch thick stainless steel window was designed. This source capsule, as shown in Figure 8b, has a calculated maximum capacity of 3 grams of AmO_2 with an available 60-kev photon emission rate of approximately 1×10^{10} photons/sec.

Both source capsules are loaded in the same manner. The AmO_2 is weighed into the capsule to the nearest 0.1 milligram, after which the capsule is closed mechanically, cleaned thoroughly, and sealed by welding. The capsule sealing is checked by a smear technique and monitored frequently during storage by counting on an alpha counter.

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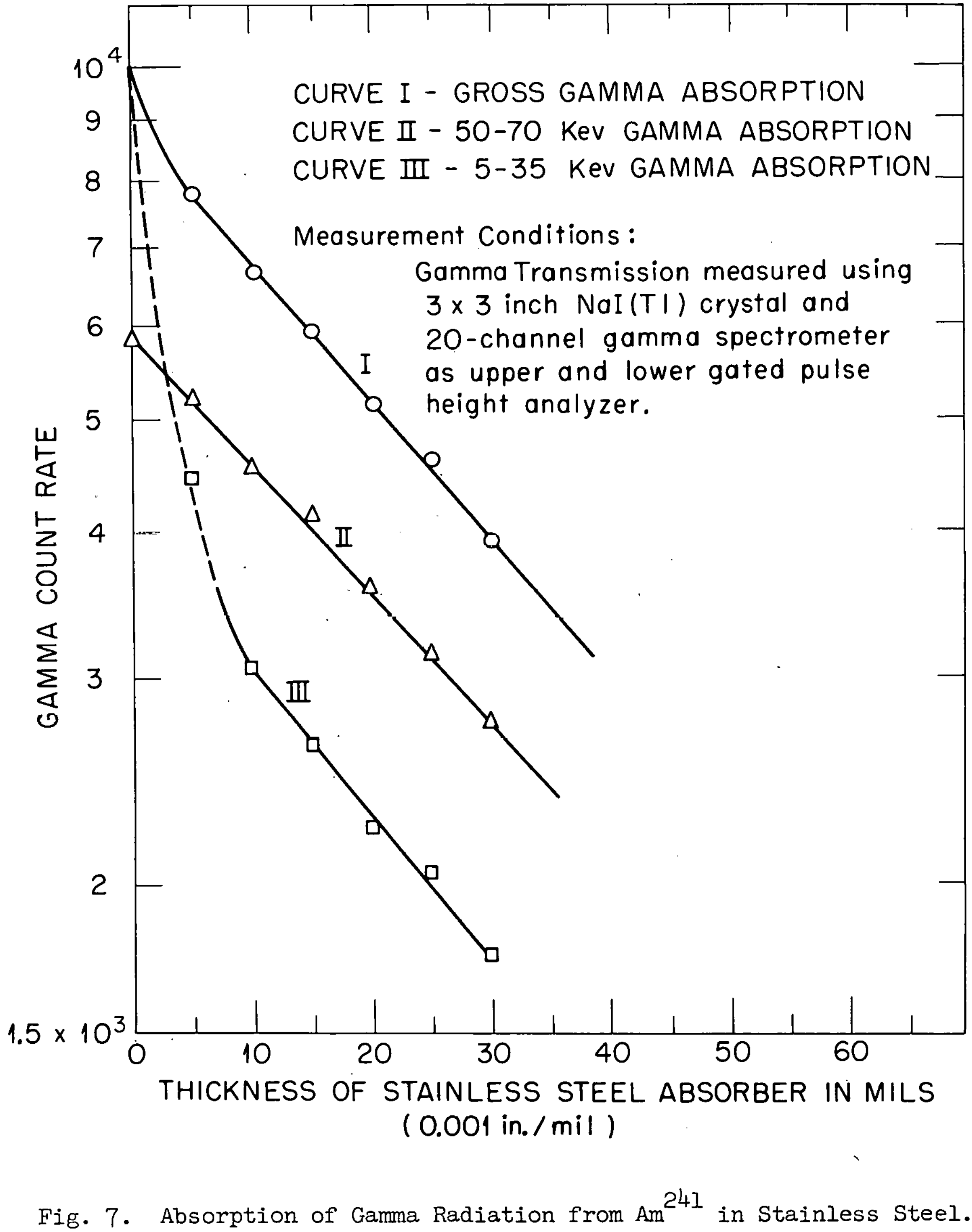

Fig. 7. Absorption of Gamma Radiation from Am^{241} in Stainless Steel.

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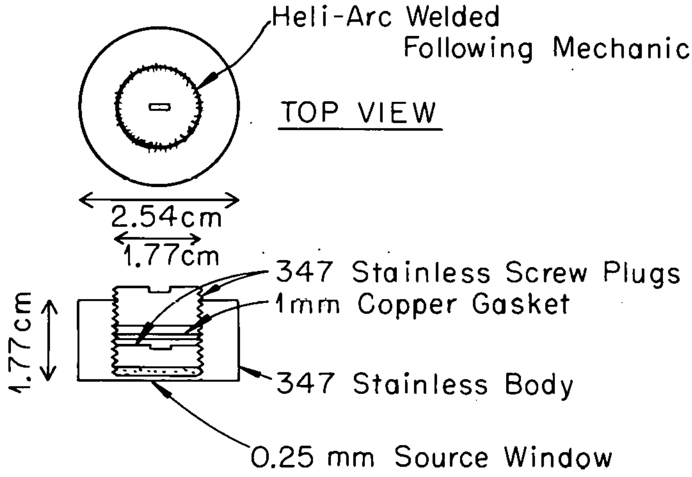
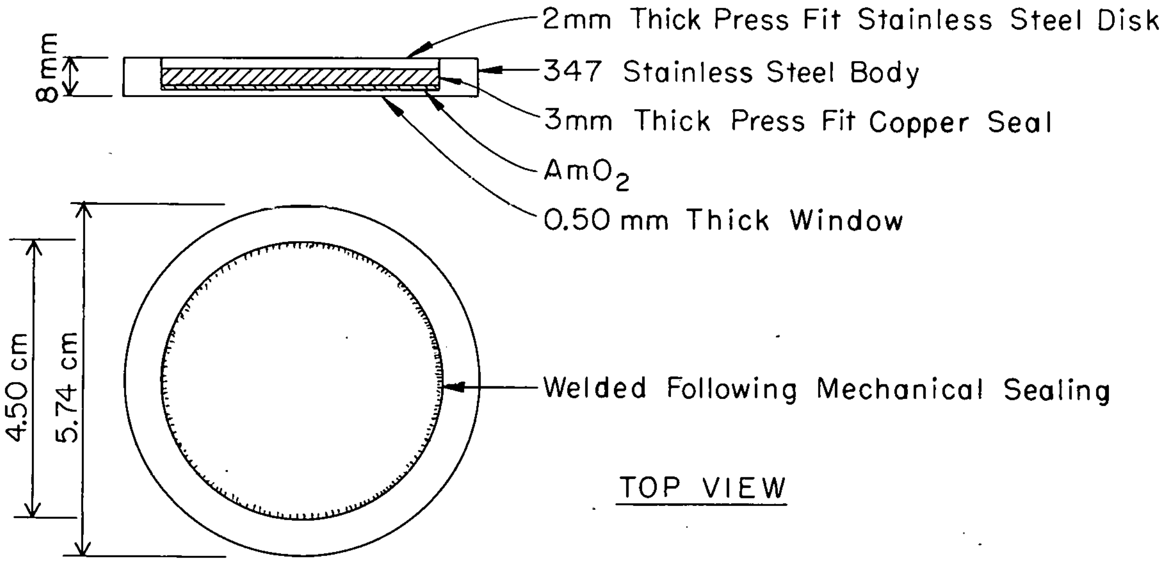


(a) Source holder for AmO_2 up to 0.25 gram.



(b) Source holder for AmO_2 up to 3 grams.

Fig. 8. Source Holders Used in the Preparation of $Am^{241}O_2$ Gamma Sources.

Americium gamma sources can be prepared from either AmF_3, $Am_2(C_2O_4)_3$ or AmO_2. In our work, AmO_2 was employed for several important reasons. It does not react with alpha particles to produce high-energy gamma photons and neutrons. For example, although AmF_3 is stable to radiation and produces no gaseous products, the fluoride atoms have an appreciable cross-section for alpha-particle capture to form radioactive 2.58 y sodium-22 and to emit neutrons by the reaction

$$F^{19} + \alpha \longrightarrow Na^{22} + n^1 + \gamma. \qquad (6)$$

The neutron emission rate from a 0.010 gram Am^{241} gamma source prepared as AmF_3 was found to be 5×10^3 neutrons per second; a 0.020 gram Am^{241} source as $Am_2(C_2O_4)_3$ has a neutron emission of less than 80 n/s. Also, as shown in Figure 9, AmF_3 sources will emit prompt gamma radiations with energies up to approximately 7 Mev (probably due to alpha capture).

AmO_2 does not form gaseous radiolytic decomposition products as does $Am(C_2O_4)_3$. For example, an Am^{241} gamma source containing 0.020 gram of $Am_2(C_2O_4)_3$ sealed with plastic cement (see Figure 10) generated in a period of two months sufficient gas pressure to bulge the 20-mil thick aluminum window. During a reencapsulation operation in a glove box, the source window ruptured with an audible explosion and the Am^{241} was scattered about the inside of the glove box. An AmO_2 source encapsulated in stainless steel (see Figure 8a) has shown no observable distortion of the source window nor has any leakage been observed after 1 year.

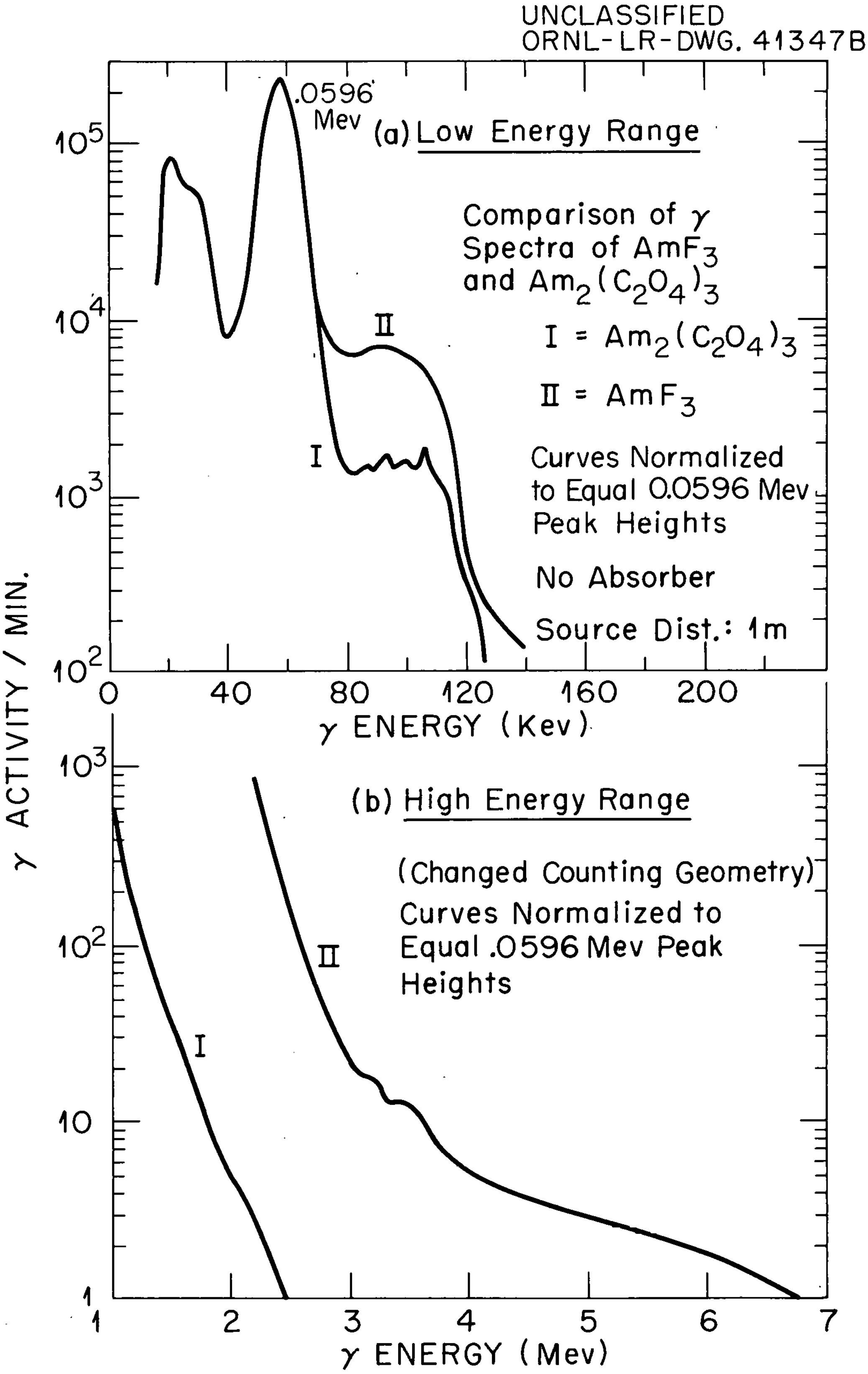


Fig. 9. Gamma Spectra of AmF$_3$ and Am$_2$(C$_2$O$_4$)$_3$ Sources.

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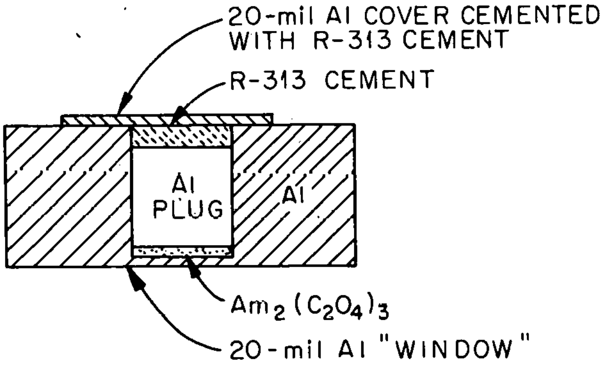

Fig. 10. Cross-Section of Experimental Am^{241} Gamma Source.

Figure 9 also shows a comparison of the gamma spectrum from an $Am_2(C_2O_4)_3$ source with an AmF_3 source. AmO_2 has a similar spectrum to that for $Am_2(C_2O_4)_3$.

Am^{241} Gamma Source Applications: The usefulness of an Am^{241} gamma source lies in the purity of the emitted gamma radiation and its low energy. The low-energy radiations emitted have good sensitivity for radiographic film as well as an absorption coefficient in materials which is a function not only of their density but also of the atomic number of the absorber. The atomic number influence is caused by the fact that at a gamma energy of 60-kev a very high percentage of the gamma absorption is through the photoelectric process which is proportional to the fifth power of the atomic number.(14)

In Radiography: Figure 11a demonstrates a radiographic technique using Am^{241} gamma sources. It is a radiograph of an obliquely cut cube of polyethylene approximately 1-inch thick with a sloping ramp cut in one side. In the upper surface it is possible to see two holes in the block. The one adjacent to the ramp is drilled vertically 3 mm deep and is 1 mm in diameter. The second hole is drilled at a 45° angle at the top rear of the block. Figure 11b shows a radiograph of a human hand and a mechanical pencil. Both of these exposures were made using Ilford high contrast Industrial X-Ray film, a 100 mg Am^{241} source, a source to film distance of 25 centimeters, and an exposure time of 10 minutes. Figure 12 shows the use of this same source in making a radiograph of a simulated luggage container weighing 5.1 pounds and having the dimensions of 14" x 15" x 6". Royal blue medical x-ray film, a

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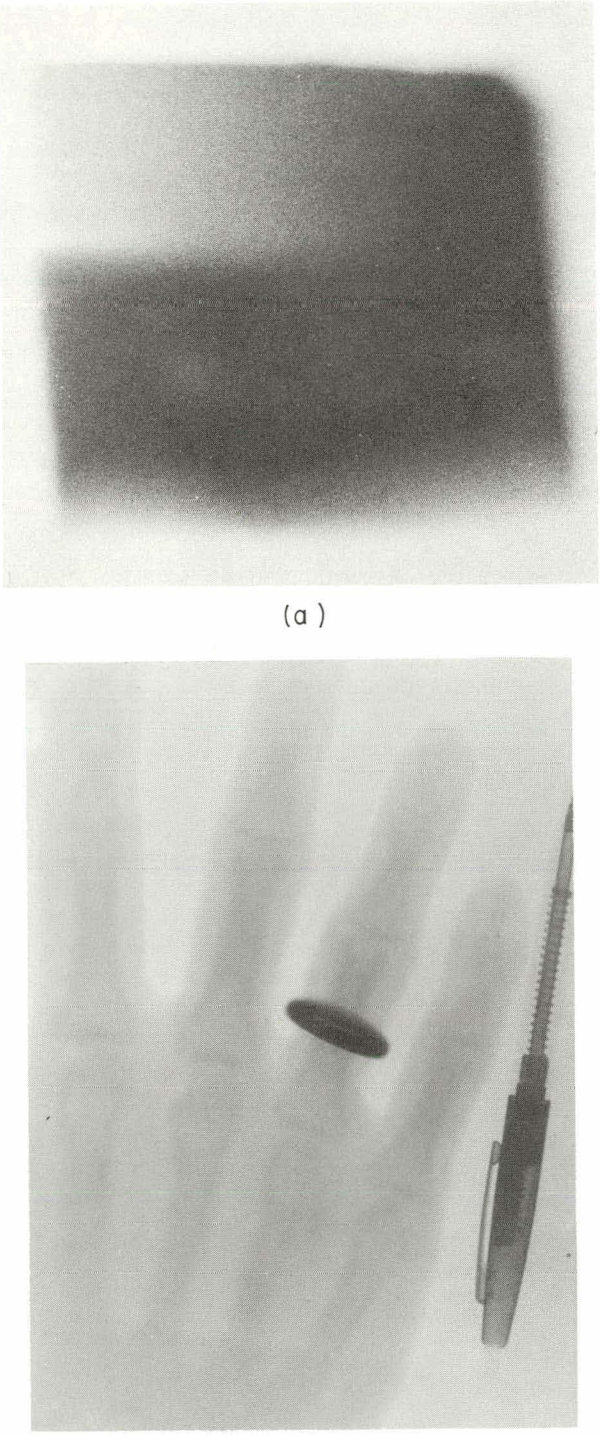

(a)

(b)

Fig. 11. Am^{241} Radiograph Examples.

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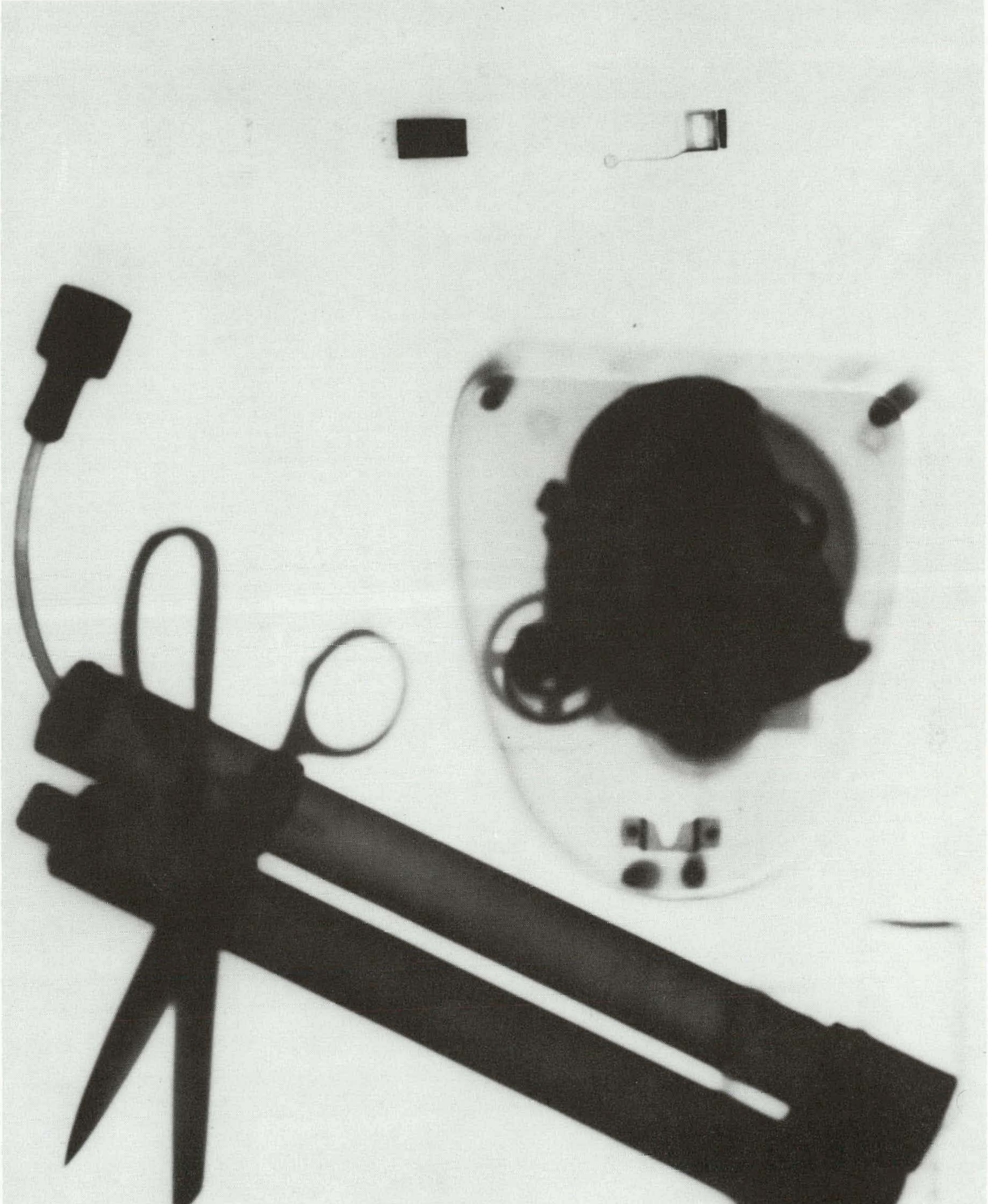

Fig. 12. Americium-241 Radiograph.

source to film distance of 20 inches and an exposure time of 10 minutes were used to make this radiograph.

In Absorptiometry: Gamma absorptiometry has been successfully used to determine high atomic numbered elements.(15-18) Figure 13a was constructed from data obtained by Stelzner,(18) who used one of the gamma sources prepared as part of this investigation to determine lead in a flowing stream; Figure 13b illustrates typical calibration curves obtained by Miller and Connally(17) in the in-line process analysis of plutonium and uranium by means of Am^{241} gamma sources.

In our work, both gamma absorption and radiographic techniques were combined in an experimental Am^{241} gamma source fluoroscopy unit similar to that shown in Figure 14. Using a 0.400-gram AmO_2 source in the 1.75-inch diameter source holder described earlier (see Figure 8b) and a standard ZnS x-ray fluoroscopy screen, it was possible to detect changes in the density of low Z materials such as aluminum, plastic, beryllium, etc., from the image intensity variations. A very simple and inexpensive method of obtaining quantitative data on absorption measurements is to use a small piece of ZnS screen taped to a light sensitive phototube. The phototube output may then be read on any sensitive microammeter or galvanometer.

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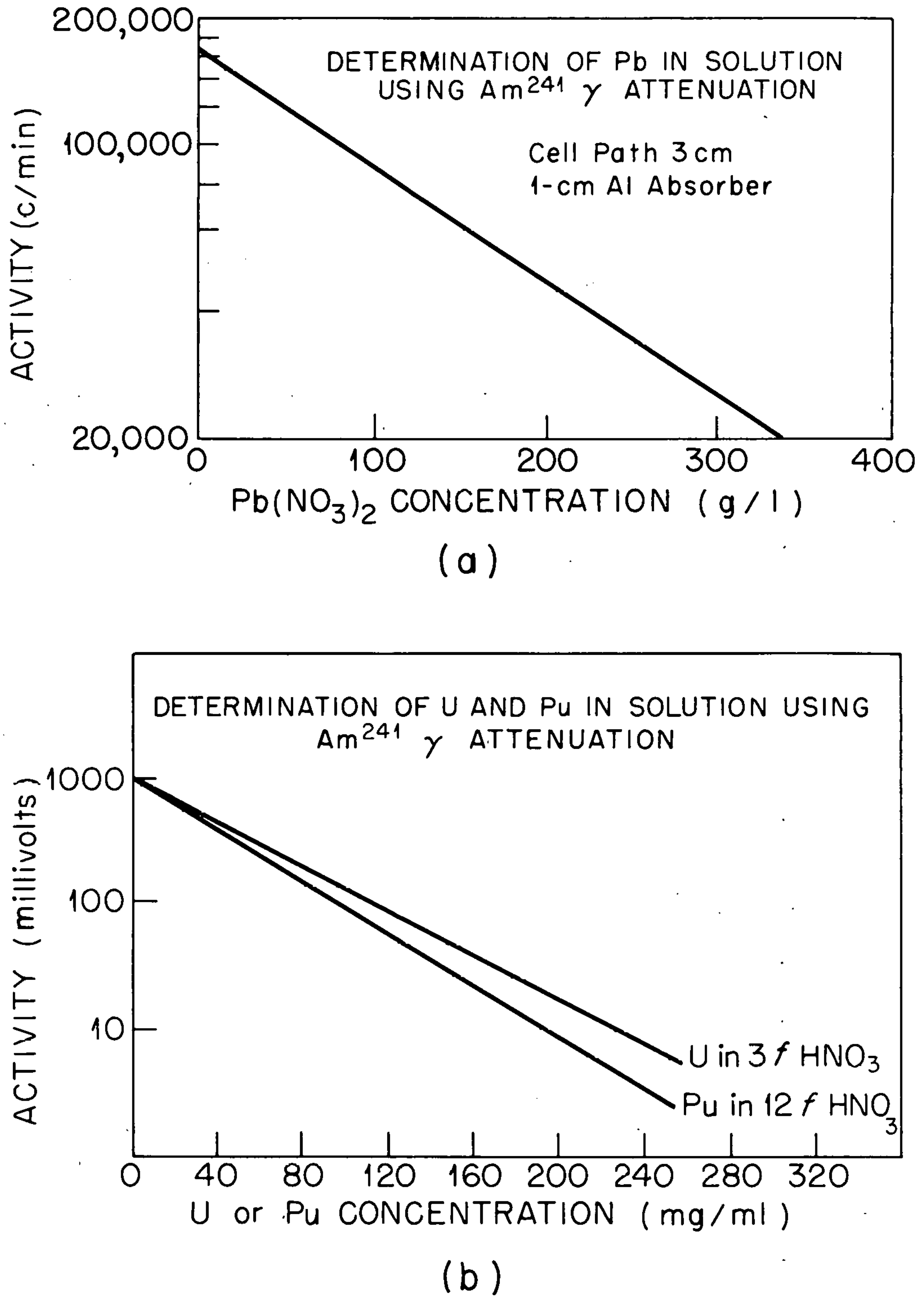

Fig. 13. Determination of Heavy Metals in Flowing Stream.

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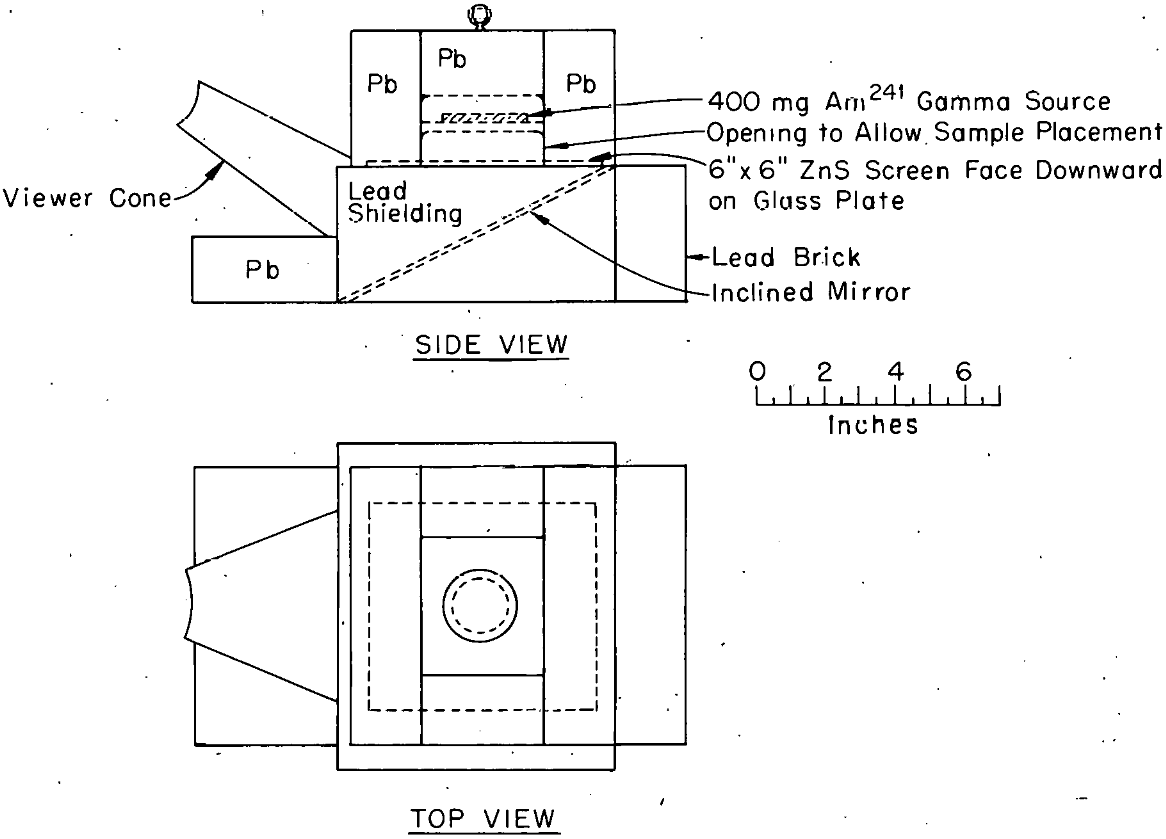

Fig. 14. Experimental Am^{241} Gamma Fluoroscopy Unit.

7.0. Preparation and Uses of Low-Intensity Neutron Sources

Low-intensity neutron sources prepared from a mixture of an alpha-, or high-energy gamma-emitting radioisotope and beryllium produce neutrons by the following reactions:(19)

$$Be^9 + \alpha \longrightarrow n^1 + C^{12} \quad Q = 5.70 \text{ Mev} \qquad (7)$$

$$Be^9 + \gamma \longrightarrow n^1 + Be^8 \quad Q = 1.63 \text{ Mev} \qquad (8)$$

Both reactions will take place in a Ra-Be source, since Ra^{226} decays by 4.78-Mev α emission to short-lived daughters which emit gamma radiations with an energy of greater than 1.63 Mev. Pu-Be and Po-Be sources produce neutrons only by the (α,n) reaction. The Sb^{124}-Be source is a pure (γ,n), or photoneutron, source. Each of these sources have several disadvantages which make them relatively undesirable. Sources made from Ra^{226} and Sb^{124} have a very high gamma level relative to neutron productivity. The low specific activity of Ra^{226} and Pu^{240} necessitates physically large sources for comparable neutron output relative to the size of Po^{210} initiated sources. However, the short half-life of Po^{210} (138 d) requires frequent recalibration of the source. The same is true of sources made from 60-day Sb^{124}.

Although neutron sources may be prepared by mixing an alpha emitter with boron,(19) the alpha reaction upon beryllium is the more advantageous since a greater yield of neutrons results (77 n/s per 10^6 alphas for beryllium *vs*. 22 n/s per 10^6 alphas for boron). In addition to neutron yield, the emitted neutron energy distribution, the gamma radiations associated with a source, and the half-life of the alpha-emitter will effect the choice of source materials. The neutron-energy distributions for polonium-beryllium(20) and polonium-boron(21) are shown in Figure 15. Wattenberg(22) reports on the neutron-energy distribution from the (γ,n) reaction. The neutron-energy distribution from either Am-Be or Am-B sources is expected to be very similar to that shown in Figure 15.

For purposes of evaluation as to neutron yield, gamma intensity, physical size, etc., experimental data from Am^{241}-Be, Am^{241}-B, and Ra^{226}-Be neutron sources has been combined in Table I with information available in the literature on Po^{210}-Be, Pu^{239}-Be, and Sb^{124}-Be sources.

The neutron emission rate of the Am^{241}-Be and Am^{241}-B sources was determined using a 4π graphite sphere detector(23) which had been calibrated using a National Bureau of Standards Ra-Be (γ,n) source of known emission rate. Assuming a theoretical neutron yield of 77 neutrons per 10^6 alpha particles,(24,25) the neutron emission rate of the Am^{241}-Be source (Table I) is only about 80% of the expected yield. Previous work(25) has shown that an americium reduction technique using AmF_3 and Be metal powder at high temperatures will result in theoretical yields and that the emission rate of a two-curie Am-Be source will be 5.7×10^6 n/second.

The gamma spectra of the Am-Be and Am-B sources were measured using a 3" x 3" NaI crystal and a multichannel analyzer to demonstrate the production of high-energy capture-gamma radiations produced in the (α,n) reaction on Be. High-energy gamma radiations in Figure 16 shows the distribution of these high-energy gamma radiations.

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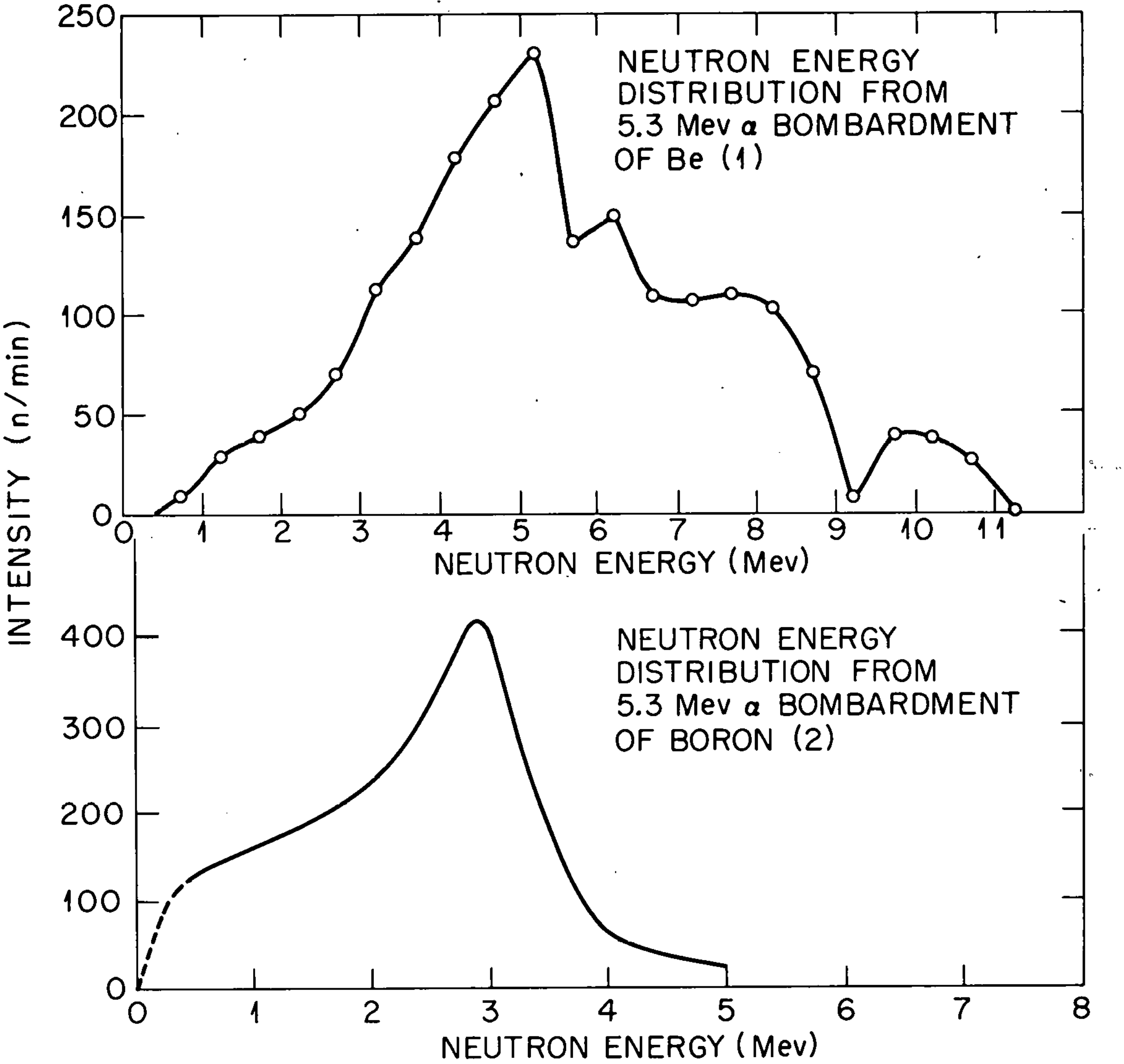

(1) MP-74 Pierre Demers (1949)
(2) MDDC-1490, H. Staub (1947)

Fig. 15. Neutron Energy Distribution from Po^{210}-Be and Po^{210}-B Neutron Sources.

TABLE I

A COMPARISON OF THE CHARACTERISTICS OF NEUTRON SOURCES

Source Material	Radionuclide Half-life	Source Dimensions[b]	Neutron Emission, n/s[c]	Average Neutron Energy, Mev.	Gamma Intensity[d]
Po^{210}-Be	138 d	0.5 x 0.5 in.	5.7×10^6	4.5	< 100 mr/hr
Pu^{239}-Be	24,360 y	1.5 x 1.5 in.	3.4×10^6	4.5	< 100 mr/hr
Am^{241}-Be[a]	462 y	1 x 1 in.	4.8×10^6	4.5	100 mr/hr
Am^{241}-B[a]	462 y	1.2 x 1.2 in.	0.93×10^6	2.5	10 mr/hr
Ra^{226}-Be[a]	1,622 y	1 x 2 in.	20.2×10^6	4	> 100 R/hr
Sb^{124}-Be	60 d	1.6 x 1.6 in	0.4×10^6	0.035	> 100 R/hr

---

[a] The values given for these sources are actual values experimentally determined from available sources. All others are calculated based on the best available information.(22,25)

[b] Overall dimensions using standard containers with 0.2-inch wall thickness for double encapsulation.

[c] Normalized to 2 curies of radioactive material.

[d] Contact measurement made using a radium calibrated "cutie-pie" survey instrument.

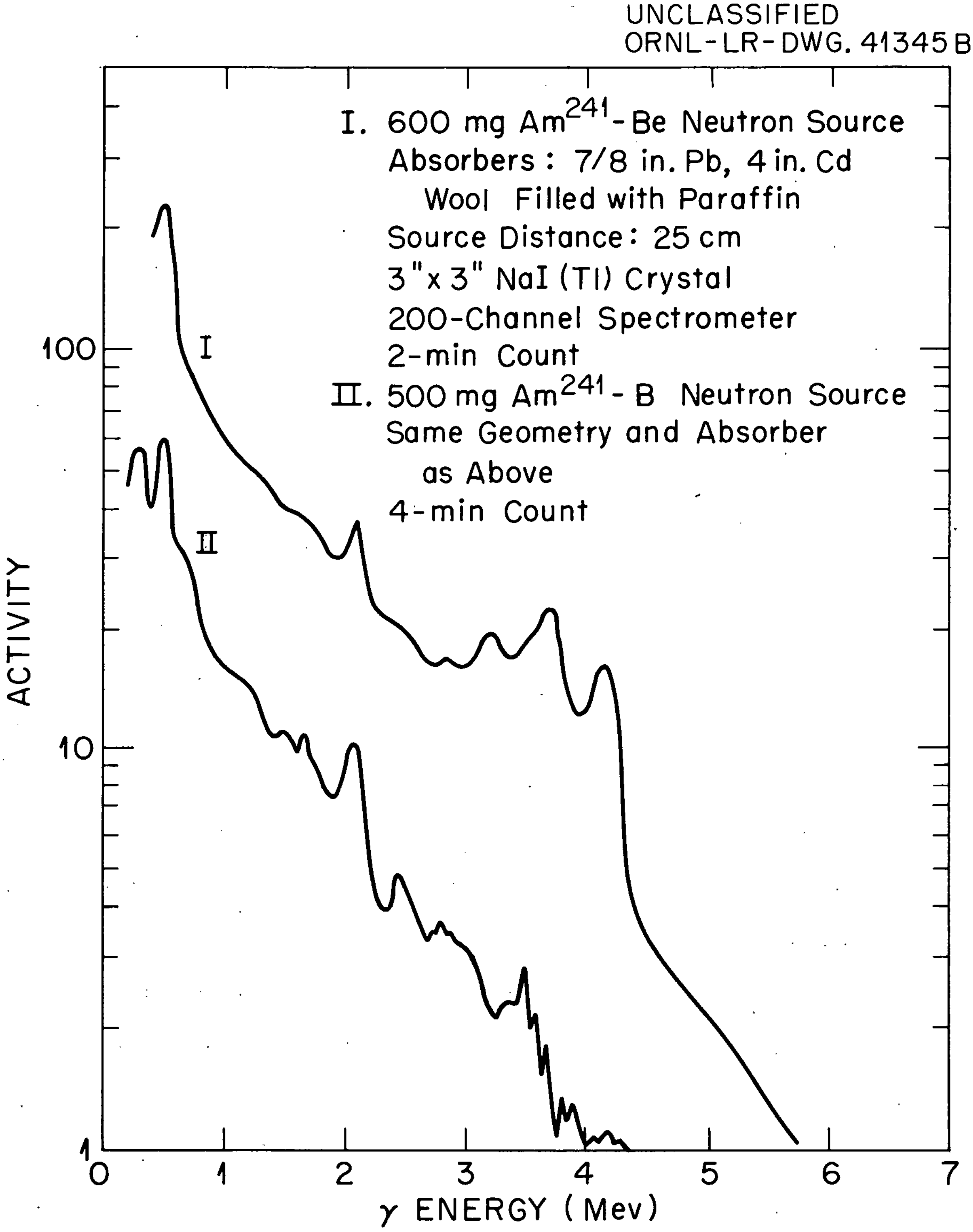

Fig. 16. Gamma Spectra of Am^{241}-B and Am^{241}-Be Neutron Sources.

Source Preparation and Encapsulation: In our investigation of neutron sources, two types of americium-initiated neutron sources were produced: Am-B and Am-Be sources. The Am^{241}-Be sources were prepared simply by thoroughly mixing $Am(NO_3)_3$ in weak HNO_3(pH-2) with > 325 mesh beryllium powder and evaporating the mixture to dryness and firing at 500° C to expel NH_4NO_3 and convert the americium to AmO_2. The Am^{241}-B sources were prepared in a similar manner. However, following the firing, the boron-AmO_2 powder was combined with 0.050 gram of paraffin dissolved in CCl_4 and allowed to dry. The mixture was then ground and transferred to a press to convert the loose powder into a 0.72 in. x 0.73 in. right cylinder under a static load of 10 tons. Following removal from the press, it was again heated to 500° C to remove the paraffin binder.

Both types of pellets were then placed in source containers similar to that shown in Figure 17 and sealed. The sealing was done in several stages. First, the pellet of pressed AmO_2 and boron or beryllium metal was placed in the nickel (or inner) container and heated to 275° C on a hot plate. Using Ruby flux and standard solder, a uniform film of solder was placed on the threads of the cap of the nickel container and the cap then screwed onto the body of the container. After a cooling period, the sealed container was cleaned with acid (1 $\underline{N}$ HNO_3), dried and checked for alpha contamination on its surface. When a smear technique removed less than 75 α counts/min. from the outer surface, the source was placed in the outer stainless steel container, its lid screwed on, and the container sealed by heli-arc welding. (NOTE: Such sources as these are

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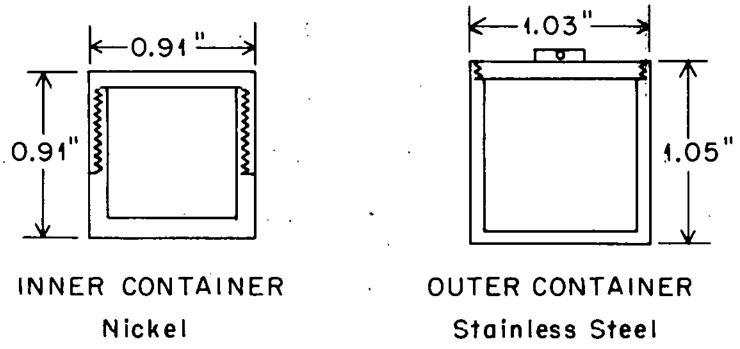

Fig. 17. Am^{241}-Be Neutron Source Container

used in our laboratory and are checked monthly by a smear technique for alpha leakage. No contamination has been detected to date.)

Low-Intensity Neutron Source Applications: The investigations of the Analytical Chemistry Division at Oak Ridge National Laboratory concerned with low-intensity neutron sources are divided into three phases: activation analysis, neutron absorptiometry, and neutron transmission.(26)

Neutron Activation Analysis

Neutron activation analysis has been carried out using an Am^{241}-Be source with a total neutron emission of 4.54×10^6 n/s. When enclosed in a paraffin moderator, the highest neutron flux attainable is 2.36×10^4 $n/cm^2/sec$ as measured by Au, Mn, and Co foil activation. Using a series of pure oxides, nitrates, or carbonates, the sensitivities of a number of elements have been tabulated for various irradiation times using a 3" x 3" NaI detector coupled to a 20-channel pulse height analyzer for discriminatory counting. Table II gives a partial listing of these elements.

It can be seen from these data that while these neutron sources are not sufficiently intense to allow trace analysis for more than a few isolated elements, it does offer a rapid and specific analysis method for macroelement concentrations in a wide variety of materials. As spontaneous neutron emitters become available, i.e., Cf-252, the portable neutron source will play an increasing role in neutron activation analysis.

TABLE II

SENSITIVITIES OF A FEW ELEMENTS FOR NEUTRON ACTIVATION USING A Am^{241}-Be NEUTRON SOURCE

Thermal Flux of 2.4×10^4 n/cm^2/s

Element	Isotope Produced	t 1/2	Gamma Energy, Mev	Irradiation Time	Energy Range of Counting, Mev	Integral c/min/mg
Hf	189 m	19 s	0.161	1 m	0.02 -- 0.21	2
Sc	46	19.5 s	0.14	1 m	0.02 -- 0.21	3
W	183	5.5 s	0.105	1 m	0.02 -- 0.21	.01
Al	28	2.3 m	1.78	5 m	1.6 -- 1.88	.04
I	128	25 m	0.455	30 m	0.72 -- 1.48	.08
Ag	108	2.3 m	0.44, 0.60	5 m	0.36 -- 0.74	.2
Mg	27	9.4 m	0.84 1.02	10 m	0.72 -- 1.48	.002
Mn	57	2.58 h	0.845	60 m	0.80 -- 1.18	.08
Au	198	2.70 d	0.412	2.6 d	0.34 -- 0.53	3
U^{238}	U^{239}	23.4 m	0.074	40 m	0.04 -- 0.42	1
U^{235}	FP's	Short	Many	40 m	0.04 -- 0.42	1

Neutron Absorptiometry

The basis of neutron absorptiometric analysis is a measurement of neutron flux depression due to neutron absorption by high cross-section materials in the sample. The flux depression is directly porportional to the number of atoms present and their thermal absorption cross-section.

The mathematical relationships of the flux depression to an element concentration and absorption cross-section may be derived in the following manner:

Assume that a thermal neutron flux of intensity of q is generated in a paraffin block moderator by a neutron source. If a $B^{10}F_3$ counter is located at some point in the moderator to measure the neutron flux, it will produce a number of pulses or counts per second which may be expressed by

$$C/S = \frac{(K)(q)(N_B\sigma_B)}{N_B\sigma_B + N_M\sigma_M} \qquad (1)$$

where C/S = the number of counts per second recorded by the scaling circuit

K = a detection constant of the system

$N_B\sigma_B$ = the number of atoms of B^{10} in the detector multiplied by the isotopic absorption cross-section.

$N_M\sigma_M$ = the number of atoms of other elements present which may absorb neutrons multiplied by their cross-section

If the count rate is again determined with a neutron absorbing sample surrounding the $B^{10}F_3$ detection, the count rate will be lowered and may be expressed by:

$$C_1/S_1 = \frac{(K)(q)(N_B\sigma_B)}{N_B\sigma_B + N_M\sigma_M + N_X\sigma_X} \qquad (2)$$

where $N_X\sigma_X$ is the number of atoms of absorbing element present times its cross-section.

Dividing equation (1) by equation (2), we obtain:

$$\frac{C/S}{C_1/S_1} = 1 + \frac{N_X\sigma_X}{N_B\sigma_B + N_M\sigma_M} \qquad (3)$$

Since $N_B\sigma_B + N_M\sigma_M$ is a constant, we may let:

$$1/K = N_B\sigma_B + N_M\sigma_M \qquad (4)$$

and

$$\frac{C/S}{C_1/S_1} = 1 + K\ (N_X\sigma_X) \qquad (5)$$

Expressing the number of atoms of the absorbing element in terms of molar concentration, (X),

$$\frac{C/S}{C_1/S_1} = 1 + \frac{K}{NA}\ (X)\ \sigma_X = 1 + K^1\ (X)\ \sigma_X \qquad (6)$$

where N = Avagadro's number

A = the molecular weight of the absorbing element.

Equation (6) is linear and is normally used in this form.

Using the apparatus of Figure 18, which provides neutron thermalization, shielding, and a rigid geometry, and standard solutions of known concentration and total absorption cross-section, the value of the constant may be determined. Figure 19 presents a series of curves showing

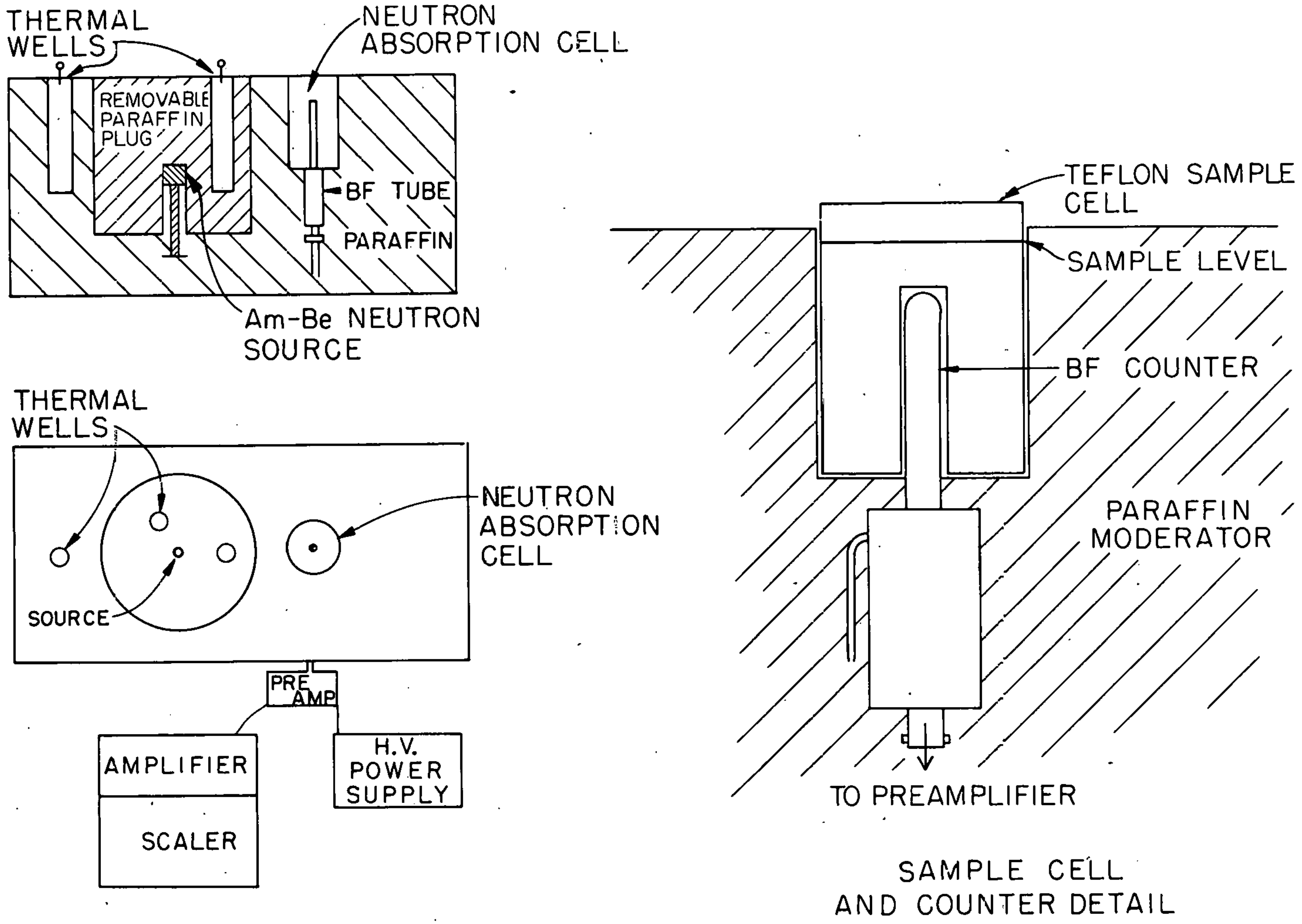

Fig. 18. Neutron Absorption Apparatus.

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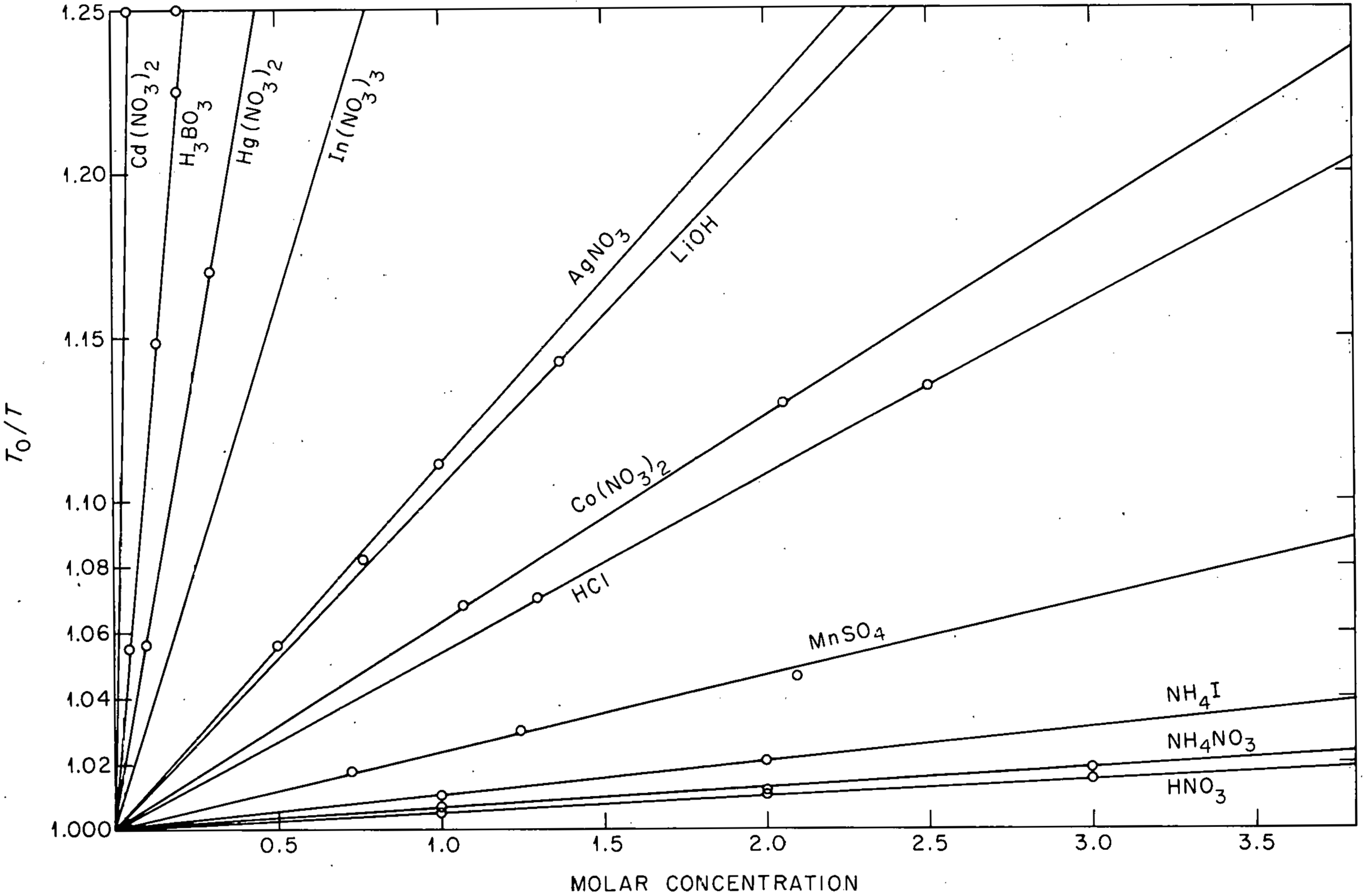

Fig. 19. Neutron Absorption Measurements (T_0/T vs Molar Concentration for Various Aqueous Solutions of the Elements in 35mm Annular Cells).

the behavior of high absorption cross-section elements or compounds in this neutron absorption system. Table III presents experimental data in tabular form in order to show the limits of detection for this method.

As an extension of the same principle of analysis, neutron absorption has been used to determine the concentration of selected elements in flowing streams. The apparatus used consisted simply of a stainless steel vessel having a capacity of about one liter in which were provided wells to contain the source and a $B^{10}F_3$ detector (Figure 20a). The vessel is connected into the process stream and the concentration of the absorbing element determined continuously. Calibration curves of the apparatus are shown in Figure 19b.

Isotopic analysis of such elements as B, U, and Li, which have one principle neutron absorbing isotope, is possible using neutron absorption if an independent method of concentration determination is used. From equation (6), if the constant, K, and the molar concentration, (X), are known, the cross section, σ, is readily determined. In the case of boron, since B^{10} has an absorption cross-section of 4.0×10^3 barns and B^{11} has a cross-section of less than 0.05 barns, its isotopic ratio is simply calculated from the observed cross-section:

$$\% \, B^{10} = \frac{\text{Experimentally derived Cross-Section, } \sigma}{4.0 \times 10^3 \text{ barns}} \times 100 \qquad (7)$$

Using this same principle, it has been possible to continuously determine the isotopic ratio of uranium, lithium, and boron in process streams.

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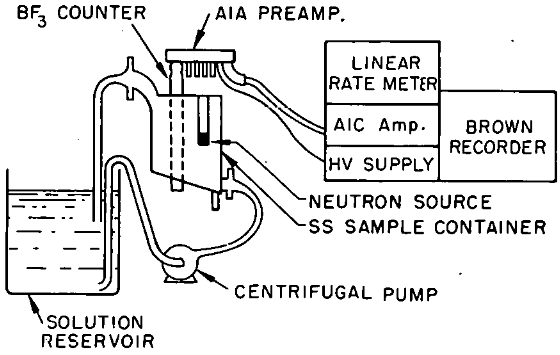
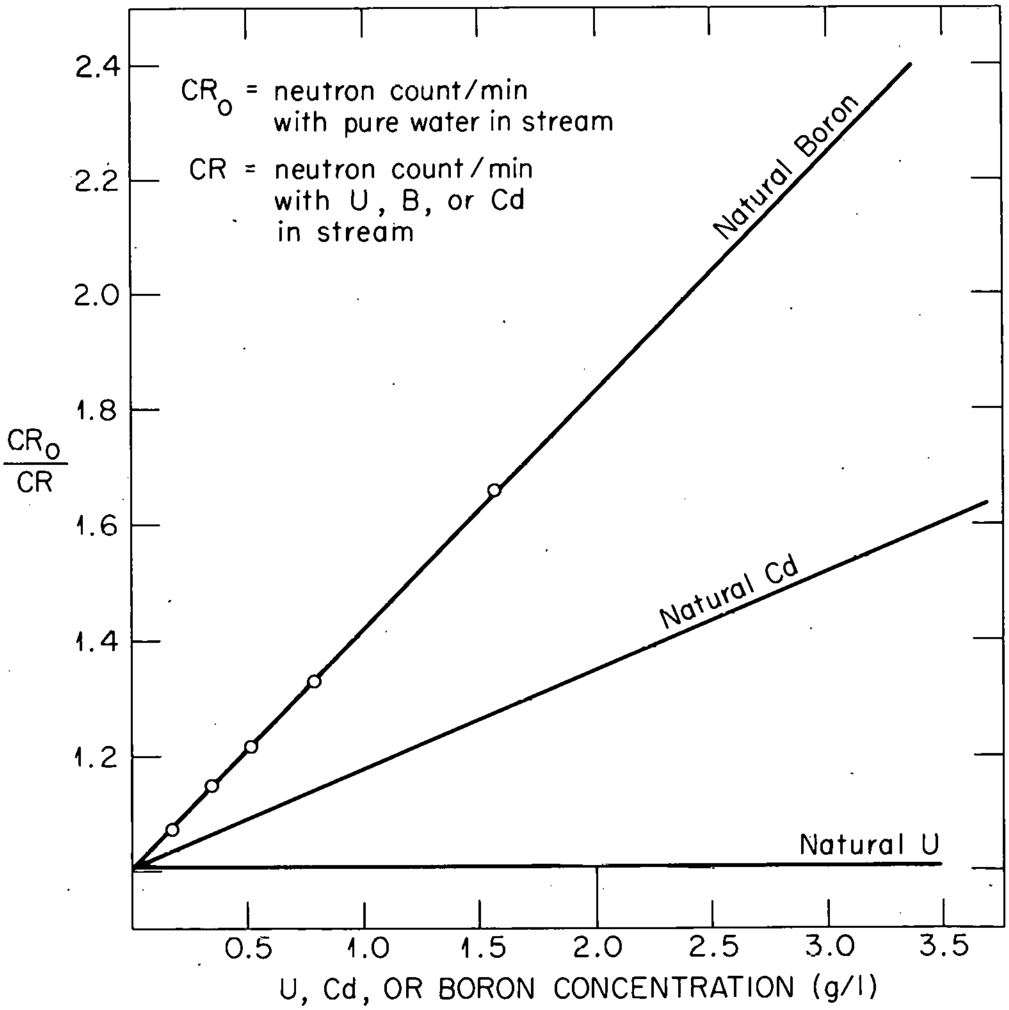


(a) Block diagram of equipment used for evaluation and calibration.



(b) Calibration curves obtained from pure solutions.

Fig. 20. Neutron Absorption in Flowing Streams.

TABLE III

APPARENT LIMITS OF DETECTION USING NEUTRON ABSORPTION ANALYZER

Compound	Apparent Molar Cross Section*	Lower Limit of Detection** As Molar Concentration	By Weight
LiCl	100	0.02	0.85 mg LiCl/ml
HNO_3	1	0.74	46.5 mg HNO_3/ml
H_2SO_4	1	0.74	72.5 mg H_2SO_4/ml
HCl	37	0.067	2.48 mg HCl/ml
LiOH	69	0.035	0.84 mg Li/ml
$AgNO_3$	72	0.034	5.78 mg Ag/ml
$In(NO_3)_3$	213	0.012	1.38 mg In/ml
$Hg(NO_3)_2$	380	0.0065	1.30 mg Hg/ml
H_3BO_3	750	0.0033	0.036 mg B/ml
$Cd(NO_3)_2$	3250	0.0008	0.087 mg Cd/ml
$UO_2(C_2H_3O_2)_2 \cdot 2H_2O$	28.5	0.052	12.4 mg U/ml

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[*] Observed experimental cross section of solutions based on the assumption that the total absorption cross section of natural boron is 750 barns.

[**] Using 40 ml of sample in the cell. Sensitivity may be increased by using a larger sample cell.

Neutron Transmission

Another application of a radioisotopic neutron source is the nondestructive analysis of plastics and stainless steels for boron content. The apparatus is pictured in Figure 21a and is simply a cadmium-shielded paraffin block to moderate and collimate a thermal neutron beam for transmission measurements.

Neutron transmission measurements may be determined either by electronic counting using a $B^{10}F_3$ detector, metal foil activation, or by boron-loaded photographic emulsions.(27) Due to the speed and sensitivity inherent in electronic counting, it was employed in our investigations.

Figure 21b shows the single calibration curve obtained by measurement of neutron transmission through varying thicknesses of indium foils. In this system of analysis, it is necessary to experimentally determine the effective cross-section of the various elements to be analyzed. Some of the values which have been determined are given in Table IV. Both the solid sample holder and the liquid sample cell, shown in Figure 21a, were used in these experiments.

These values of cross section, when used in the equation

$$\frac{CR_o}{Cr} = 1 + K\left(\frac{mg/cm^2}{\text{molecular wt.}} \times \sigma_T\right) \tag{8}$$

where $\frac{CR_o}{CR}$ = count rate with no absorbing element present divided by the count rate with the sample containing the unknown quantity of absorber in place,

K = slope constant determined from the standard curve,

σ_T = the experimentally determined cross-section value, will result in a direct determination of the amount of absorber present in mg/cm^2. This apparatus has been used to determine the quantity of boron in thin samples of stainless steel and polyethylene as low as 0.13 weight percent boron.

A modification of this apparatus, as shown in Figure 22a, allows one to determine the quantity of boron in large sheets of boron-loaded polyethylene up to one-inch thick and boron concentrations up to 6% by weight. A typical calibration curve is shown in Figure 22b.

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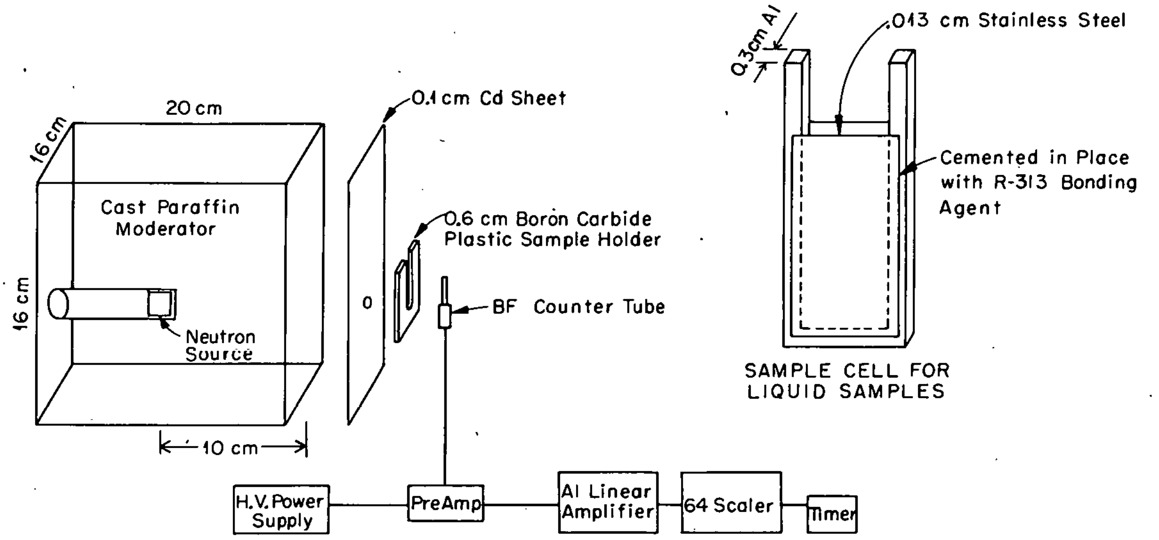
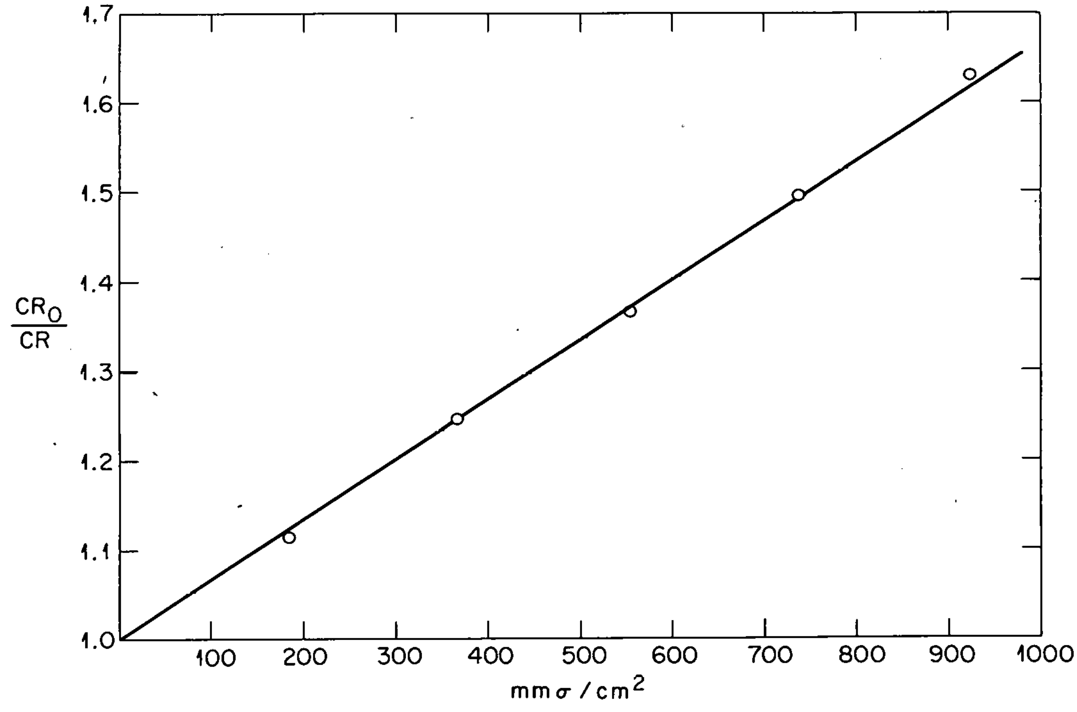


(a) Experimental neutron transmission apparatus with liquid sample cell and electronics.



(b) In calibration curve for neutron transmission; 600 sec count, in primary standard.

Fig. 21. Neutron Transmission Apparatus and Calibration.

TABLE IV

MEASUREMENT OF EFFECTIVE CROSS-SECTIONS BY NEUTRON TRANSMISSION

Element	Cross Section(29) Absorption, Barns	Scattering, Barns	Effective Transmission Cross Section, Barns
In	190	2.2	192*
Ag	62	6	65
Cd	2.55×10^3	7	3400
B	755	4	800
H	0.33	38	10.3
O	< 0.0002	4.2	1.2

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*Used as point of reference; all other values have been determined relative to this cross section.

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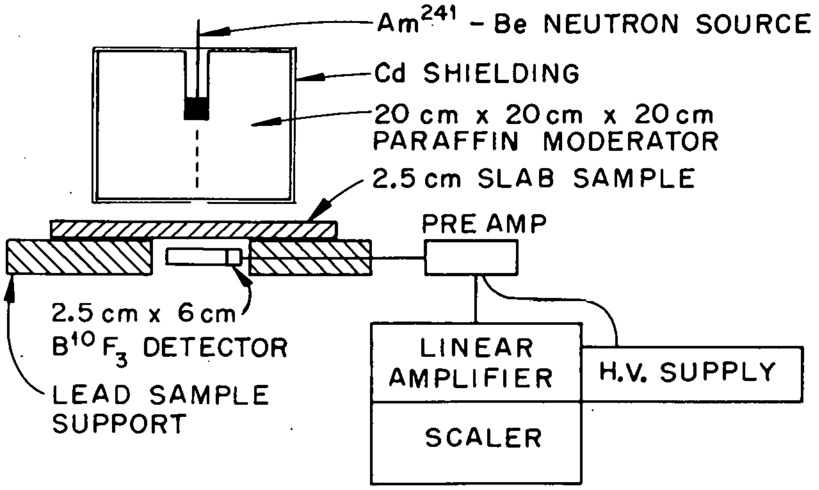
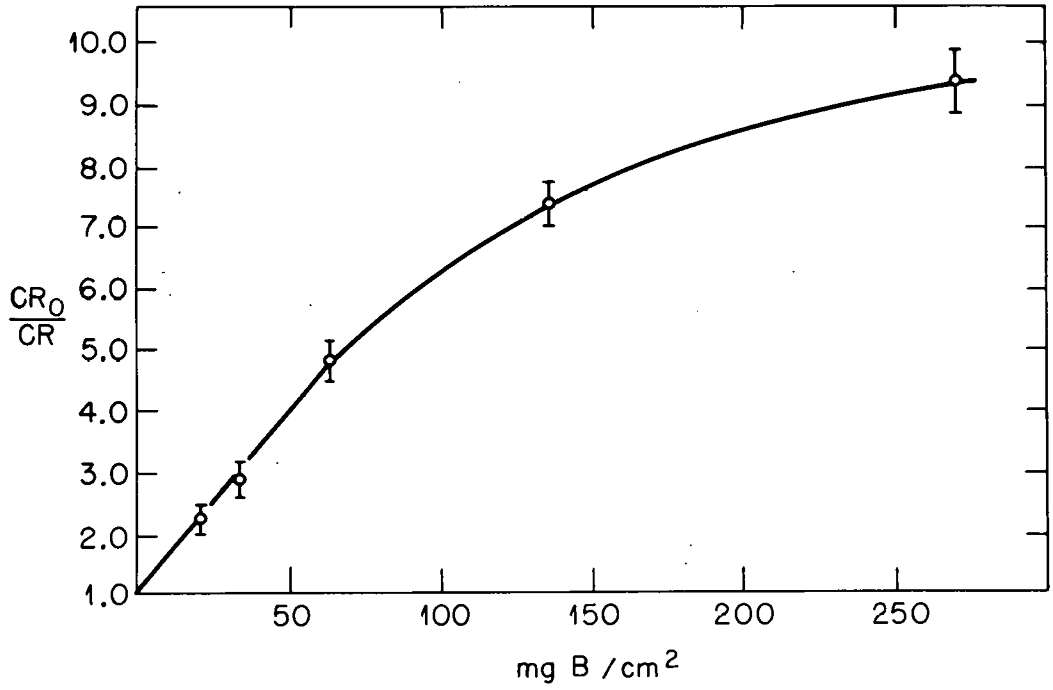


(a) Thick slab neutron transmission analyzer.



(b) Experimental result of neutron transmission measurement in synthetic boron loaded samples.

Fig. 22. Thick Slab Neutron Transmission.

8.0. Conclusions

Americium, atomic number 95, mass 241, is a valuable starting material for the preparation of long-lived alpha, gamma, and radioisotopic neutron sources. Its chemistry is simple and straightforward. The 60-kev gamma radiation associated with its decay enables one to detect its presence easily and yet is sufficiently weak to present no problems of shielding or collimation.

The potential usefulness of the portable neutron source, low-energy gamma sources, and alpha reactions have yet to be fully developed. The expanded use of americium as a substitute for the short-lived Po^{210}, low specific activity Pu^{239}, or the high gamma Ra^{226} should greatly facilitate the research in these fields.

APPENDIX A

Column Monitors and Radioactivity Assay Techniques

Since all work with milligram or greater amounts of Americium must be carried out in a glovebox maintained at a slight negative pressure, a monitoring system was necessary whereby the eluate from the ion-exchange separation column (see 4.0 Purification of Americium-241) could be monitored for gross radioactivity and yet maintain glovebox integrity. In order to accomplish both goals, a 1/2" diameter lucite window was placed in a stainless steel glovebox at a point near its base. A beta-gamma, end-window proportional G-M counter was then fastened to the outside of the window on the outside while a spiral of small-bore plastic tubing held in place by Wood's metal was placed against the inside wall of the window. The tubing was connected to the base of the column and the G-M counter was connected to a standard porportional counter and a linear rate meter to yield a Brown chart record of the activity flowing through the column.

In order to evaluate the decontamination factors effected by the column separation, radioactive isotopes of sodium (Na-24), lanthanum (La-140), and iron (Fe-59) were added to the column with the amount of Am^{241} to be purified. These added radioactive tracers marked the elution of the stable and radioactive contaminants from the column and it was possible to determine quantitatively the percent of the respective elements removed. Identification of each radioactive component was made by measuring aliquots of the eluate by means of gamma spectrometry.

Both sodium and lanthanum elute from the column prior to the americium (and iron) stripping. After extracting the strip solution with di(2-ethyl hexyl) phosphoric acid solution, the iron was separated from the americium by extracting the HDEHP phase with water.

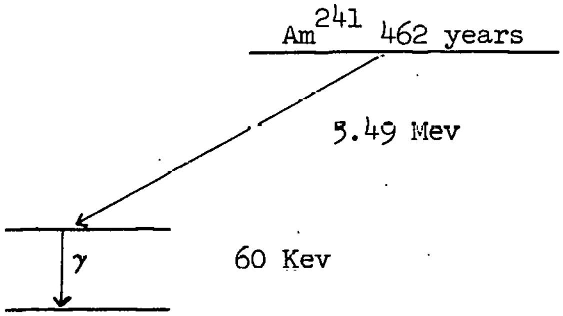
All of the americium radioactivity measurements were made by gamma counting. Assay was performed by calibrating a 20-channel gamma spectrometer with a standard volume of known Am-241 content. Since the effluent monitor recorded only gross radioactivity, more specific information was required as to the radiochemical purity of the Am^{241} fraction. In order to accomplish this, the Am^{241} radioactivity measurements were made by means of gamma spectrometry (in this instance, a scintillation counter equipped with a 3" x 3" NaI crystal and a 20-channel pulse height analyzer). The spectrometer was calibrated for Am^{241} by use of standard samples of known Am^{241} content. The standard solution was prepared from $Am(NO_3)_3$ solutions and assayed by alpha counting. To minimize the effect of an increased 60-kev gamma count due to the Compton scatter contribution of a more energetic gamma, the 20-channel spectrometer was used as a discriminatory counter, and samples of pure La-140, Na-24, and Fe-59 were counted in the 60-kev region as well as at their major photo peaks. These experimentally determined ratios were than used to mathematically correct for counts recorded in the 60-kev region due to Na, La, or Fe radioactivities.

This system of radiochemical analysis allows one to transfer the sample to be assayed from the glove box in polyethylene bottles, counted, and returned to the system without any of the dilution, evaporation, and solid removal treatments necessary with alpha counting. The values routinely determined using gamma spectrometry are found to agree within 5% of the alpha counting techniques.

APPENDIX B

Handling and Monitoring Procedures for Americium-241 Sources

All users of Am^{241} sources, whether as alpha, gamma, or neutron (with Be or boron) sources, should follow a regular schedule of inspection and monitoring of the sources in order to minimize personnel radiation hazards and the possibility of serious contamination of the environment. Am^{241} decays with a half-life of 462 ± 10 years with the emission of alpha particles (average alpha radiation energy, 5.49 Mev) and 60-kev gamma radiations. The decay scheme as shown in Figure 1, may be simplified to show that most of the decay of Am^{241} occurs in the following manner:



Simplified Decay Scheme

The total permissible body burden is approximately the same as that of plutonium in microcuries although the relatively short half-life gives americium a specific activity of 50 times that of Pu^{239} or 3.17 curies per gram.

In order to establish a practical and safe usage of Am^{241} sources, it has been found advisable to handle and monitor these sources in the following manner:

Alpha Sources of Am^{241}: There are presently two types of alpha sources in use:

1. The uncovered source which is a constant contamination hazard and is always kept in a sealed container.
2. The foil covered source used for deionization application and alpha counting standards.

The open source is always handled as a source of contamination. Depending upon the activity of the source, more stringent measures must be taken to assure its controlled use. The source can continually flake off radioactive material through thermal cycling or recoiling masses of atoms. All equipment associated or used with this type of source must be considered contaminated until thoroughly monitored.

The sealed sources consist of an active deposit of Am^{241} covered with a thin metal foil which will transmit alpha particles but contain the active material. The most satisfactory material found to date is 0.1 mil (0.0001 inch) Ni foil. This transmits approximately 72% of the alpha activity from Am^{241} with a maximum emergent energy of 4 Mev in a continuum to zero energy. This covered source, while still fairly delicate, can stand more rough treatment than the open source with less danger of contamination to the apparatus in which it is used.

Although the quantity of radioactivity in an alpha source is generally much lower than that in a gamma or neutron source, the storage and monitoring techniques are the same as those described below for use with an Am^{241}-neutron source.

Gamma sources of Am^{241}: These sources can contain up to 1 curie of Am^{241} and for our purposes have been encased in a welded stainless steel container having a "window" thickness of 10 mils (0.010 in. or greater). The source is rugged and the puncturing of the thin stainless steel window is the only real hazard. There is no measurable gamma emission from the back or sides of the source.

Sources of this type are monitored in a manner similar to that used for the Am^{241} neutron sources. A method of storage has been used in which a 1/4-inch thick lead sheet is used to cover the active face (or source window). This assembly is enclosed in a plastic container which may be sealed. A 1-inch thick lead plate is usually machined to accept the source container so it is immobilized in the well. A "contact smear" or small circle of filter paper is placed in the container in contact with the source. Prior to each use, the source should be visually inspected and the "contact smear" counted for alpha contamination. At least once a month, if the source is not used more frequently, the contact smear should be monitored.

Neutron Sources of Am^{241}: The sources prepared in this study have been doubly sealed in a nickel inner container sealed with soft solder within a welded stainless steel outer container. The inner container wall thickness is 80 mils (0.080 inch) and the outer container has a 60 mil (0.060 inch) wall thickness so that the source is fairly rugged and will take a great deal of rough treatment, i.e., temperatures to 500°C and hydraulic pressures of up to 100 atmospheres.

Since the sources may contain up to several grams of Am^{241}, it is imperative that any evidence of source leakage be discovered at the earliest possible date and measures taken to control the spread of contamination. This is best accomplished using a series of regular checks for physical damage and smearing to detect leakage.

1. The source is stored in a cadmium-shielded or boron-impregnated paraffin shield sufficiently large to reduce fast neutron leakage to a safe level (this will depend on proximity of personnel and source output) in an exhaust hood area.

2. A "contact smear" or small circle of filter paper is placed in the storage container in contact with the source which is counted for alpha activity before source use or on a monthly basis, whichever is the more frequent.

3. The gamma emission from the source is no problem as the thickness of paraffin used to reduce neutron hazard will more than control the gamma dosage received.

Summary: All radioactive sources are a potential hazard, as are all electrical appliances, fuel tanks, etc., and must be inspected regularly.

Always assume that a source is contaminated until a check of the contact smear is made.

If a source is found to be leaking, seal and double seal it at once in any gas tight container and survey to determine the extent of contamination.

Regular checks for leakage or physical damage can pay big dividends in saved time, money, and difficulties.

APPENDIX C

Determination of the 60-kev Photon Emission From Sealed Americium-241 Sources

The preparation of a gamma source of known photon emission involves several parameters which make it rather difficult to calculate in advance exactly the quantity of radioactive material which should be used. A few simple counting experiments to determine the various effects of source materials will, however, allow one to calculate the final emission with reasonable accuracy.

The first factor to consider is the decay scheme of the radioactive isotope which will be used and a decision as to what gamma photon will be the basis of measurement. In order to combine all nuclear parameters for Am^{241}, i.e., branching ratio and internal conversion, into one value, it was decided to experimentally determine the ratio of 60-kev gamma photons to alpha particles. This was done simply by evaporating a few micrograms of Am^{241} onto a stainless steel plate (since the source holders are fabricated from stainless steel) to equalize any radiation backscatter. This plate was then counted in a gas flow proportional alpha counter of known efficiency and geometry and also directly on the top of a NaI(Tl) crystal of the 20-channel gamma spectrometer. Correcting the number of counts collected in the 60-kev peak due to losses by means of the iodine x-ray escape(28) which takes place at the surface of the NaI crystal, one has the ratio of 60-kev gamma to alpha particle ratio.

A sample calculation is of the form:

Alpha count/4 min = 41,087 c/4 min. at 50% geometry

60-kev gamma counts/4 min. = 11,462 c/4 min. at 50% geometry

Iodine x-ray escape factor = 0.22

Corrected 60-kev photons/4 min. = 11,462 x 1.22 = 13,984 photons/4 min.

$$\text{Ratio of } \frac{\text{gamma}}{\text{alpha}} = \frac{1.4 \times 10^4}{4.1 \times 10^4} = 0.34.$$

This value can be used for calculation of the number of 60-kev gamma photons emitted by a given quantity of Am^{241} and encompasses correction for both branching ratio and internal conversion.

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