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**FEASIBILITY STUDY FOR A MOBILE
LABORATORY TO EVALUATE
ENVIRONMENTAL RADIATION
CONTAMINATION**

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ARMED FORCES RADIOBIOLOGY RESEARCH INSTITUTE
Defense Atomic Support Agency
Bethesda, Maryland

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January 1966

FEASIBILITY STUDY FOR A MOBILE LABORATORY TO EVALUATE
ENVIRONMENTAL RADIATION CONTAMINATION

This report, prepared by
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ARMED FORCES RADIOBIOLOGY RESEARCH INSTITUTE
Defense Atomic Support Agency
Bethesda, Maryland

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ABSTRACT

A study of the laboratory requirements necessary for evaluating radioactive contamination of food, water and biological specimens has been completed. From the information obtained, a mobile laboratory facility with the capability of qualitative and quantitative analyses of millimicrocurie amounts of alpha, beta and gamma emitting radioisotope contaminants has been designed. The laboratory utilizes sufficient shielding so all analyses can be performed in a two milliroentgen per hour gamma radiation field and where the average gamma energy is one mev. The entire laboratory including all electronics, detectors, shielding, power supplies, motor generator, laboratory structure and hardware, and transport container weighs less than two thousand pounds and has a total power requirement (115 volts a.c. -60 cycle) of four kilowatts*. The laboratory in its cubic transport container, six feet on a side, is thus readily transportable either by helicopter or truck.

The entire laboratory is enclosed in a clean air plenum provided by an air-blower-supported, lightweight, nylon-and-rubber tent fifteen feet in diameter, with an additional area six feet square to serve as an air lock and personnel decontamination area. A rigid floor is provided by the opened transport container and this floor is integrated with an air tight seal to the tent walls. The tent can be set up on any level surface provided that fallout debris has been cleared from the area where the tent floor is to be located. Inflation of the tent requires less than

*Power is furnished by a self-contained gasoline driven motor generator.

thirty minutes. All the laboratory components are shipped within the tent structure in the confines of the transport container. A removable panel in the center of the laboratory floor provides access to the earth shield hole for the detector canister. Also included in the mobile laboratory is an air conditioning unit to provide temperature regulated filtered air to the laboratory interior.

From a review of the recent literature concerning radiation detection instruments, it is apparent that identification and quantitative evaluation of radioisotope contaminant constituents will require a standard sample size and counting geometry. All samples have thus been standardized to the dimensions and cylindrical geometry of a No. 10 food container. Reasonable detection efficiencies for alpha and beta counting are obtained by placing the detectors in intimate contact with the sample. (This also prevents the degradation of the alpha and/or beta particle energies due to ionizing interactions in an intervening air space.) The alpha and beta detectors are mylar-backed zinc sulfide and one-mil thick plastic scintillators respectively. These detectors are embedded in a plastic snap-on type throw-away lid, which for counting purposes replaces one end of a No. 10 container. The container is placed in a detector package, located in canister in such a way that the alpha and beta detectors are viewed at close range by photomultiplier tubes. Gross gamma counting is accomplished with a well-type plastic scintillator which is large enough to accommodate the entire No. 10 container. Alpha, beta and gamma counting are carried out simultaneously using standard, transistorized, modular, commercial nuclear counting apparatus including single channel pulse height analyzers for limited energy discrimination. Identification of the gamma emitting contaminants is performed with a sodium iodide scintillation

spectrometer and a commercial multichannel pulse height analyzer. All the detector systems are located inside the eight inch diameter, six feet long stainless steel canister. The end of the canister containing the detectors is lowered into a hole in the ground three feet deep to provide radiation shielding, and to eliminate the necessity of transporting heavy, shielding materials. Using this type of shielding and the aforementioned detectors, beta, gamma, and alpha contaminant concentrations of 3×10^{-3} μ curies, 1×10^{-3} μ curies, and 3×10^{-3} μ curies respectively, can be reliably measured in a counting time of less than one minute.

Radiochemical procedures require long times and rather elaborate equipment. Consequently, this form of isotopic analysis is not suggested for the purposes of the mobile laboratory. However, identification and quantitative estimation of contaminants in small volume biological specimens, such as blood, will require separation procedures, hence simple paper chromatographic analyses have been suggested with the ultimate alpha, beta, or gamma counting to be performed with the canister detection system.

It has been estimated that the first operational mobile laboratory unit would require a two man year program to design, develop and fabricate and would cost between 150 and 170 thousand dollars. It is also estimated that each additional laboratory unit would cost between 80 and 100 thousand dollars.

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INTRODUCTION

GENERAL

This feasibility study has determined that a mobile laboratory facility capable of on-site evaluation of radioactive contamination in food, water and biological specimens could be constructed using, for the most part, commercially available instruments and operated through use of established analytical techniques.

As requested in the scope of work, the facility is self-contained, and includes power supplies, radiation detection instruments, and radiation shielding. The laboratory weighs less than 2000 pounds, including its transport container, and should present no transportation difficulty for helicopters such as those models listed in Appendix A. The suggested laboratory equipment permits detection of millimicrocurie amounts of radioactivity in the presence of a high gamma radiation background. Isotope identification and the quantitative estimation of isotopic concentration are also possible.

The development of this final report has been arranged to present immediately the physical details of the laboratory, describe the laboratory capabilities, and estimate the approximate development costs. Part I describes the general design and operating characteristics of the laboratory, including a discussion of the laboratory environment, detector shielding concepts, instrumentation, sample handling and preparation, and analytical capabilities and limitations. Block diagrams of a typical laboratory instrumentation system are shown. The description of systems components, and the cost estimate for the entire laboratory facility follows.

Part II covers radioactive contaminants, instruments for measuring radiation, and radioisotope analysis. This information has been used as the basis for designing the mobile

laboratory. The three sections review the state of the art in radiation detection systems and analytical techniques for separating and identifying radioisotopes. They include a review of the instrumental methods of detecting and analyzing alpha, beta, and gamma emitting radioisotopes, plus analytical schemes for gamma energy analysis and radioisotope identification. The sensitivities of several of the detection systems are included. Basic methods of radiochemical analyses are also reviewed, and a brief description of simple paper chromatographic and electromigration techniques, as they apply to radio-analyses, are also given.

The appendices list the capacities of several U. S. helicopters, typical commercial equipment, and developmental hardware for the laboratory.

NOTE

Numbers in parentheses (1) indicate references.

PROBLEM DESCRIPTION

The hazards created by radioactive contamination of a life-supporting environment are difficult to assess. An overall evaluation must consider both the immediate and long term effects, and this necessitates determining both qualitatively and quantitatively the composition of the contaminants and their transport vehicles (dust, water, etc.). Half-life data and knowledge of chemical activity and biological effects will then define the appropriate countermeasures.

Contamination confined to the laboratory requires immediate isolation of the affected area, and health physics monitoring and decontamination of the personnel involved. Personnel control and decontamination procedures can be established within a well-defined and controlled health physics program.

Radioactive contamination occurring because of a reactor excursion or nuclear weapon detonation presents an entirely different situation. The unconfined contaminants can be spread over a large area, as in the case of weapon fallout, with the distribution a function of the prevailing weather conditions, altitude, soil conditions and soil composition, etc.⁽¹⁾ Consequently, a much more elaborate effort is required to care for the immediate hazards and the long term effects. Events occurring in, or subsequently affecting, areas far removed from a completely equipped radiological health physics laboratory present hazards which are difficult to assess because of the logistics. The problem involves performing on-site, low-level (highly sensitive) qualitative and quantitative determinations of the type and degree of contaminant-uptake in humans from the atmosphere, food, and water, etc., under conditions of a high radiation background. To carry out low level measurements requires well-shielded radiation detectors and this may require transporting heavy shielding materials to a remote area. On the other hand, to transport what may amount to thousands of samples (food, water, biological specimens) from widely separated areas to an adequate health physics laboratory each time regular monitoring is required is no mean task.

A solution to these monitoring and analyses problems would be the development of completely portable health physics laboratories which could be readily transported (by helicopter, if necessary) to an affected area shortly after a nuclear event and at a time when the bulk of fallout or reactor-vented contaminants are already down. These laboratories would have to be capable of operating in a high gamma background environment while performing radiological studies and analyses of biological systems (blood, urine, and fecal materials), food and water, animal feeds, etc. Routine operation of the laboratory would be carried out by a trained technician. Additional help could be provided by untrained personnel.

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PART I

MOBILE LABORATORY CONCEPT

- Design
- Component Description
- Cost

CONCEPTUAL DESIGN OF A MOBILE ENVIRONMENTAL RADIATION EVALUATION LABORATORY

GENERAL LABORATORY SPECIFICATIONS AND OPERATING CONCEPTS

The Mobile Environmental Radiation Evaluation Laboratory (MEREL) includes measuring capabilities for alpha-, beta-, and gamma-emitting radionuclides. The minimum detectable concentration of these nuclides corresponds to the NCRP standards for total body burden and is of the order of milli-microcuries⁽²⁾. The maximum sample size corresponds to a number 10 food container.

The mobile laboratory facility has the capability of operating in a 2-mr/hr gamma background whose average energy is 1 Mev. The origin of the background may be in the form of a distributed source such as a fallout field.

The laboratory derives its power from a built-in power generating system consisting of a 4 KW gasoline driven motor generator.

The services of one trained laboratory operator are required. Under his guidance, untrained assistants can be used to perform most of the manual operations required.

The laboratory weighs about 2000 pounds including the laboratory shelter, motor generator equipment and the 6 ft x 6 ft x 6 ft transport container. Thus, it can be readily transported by helicopter.*

The laboratory must be located on a level site large enough to accommodate a fifteen-foot diameter clean air plenum (air-supported tent) and a 6-ft by 6-ft airlock. If the ground has a fallout cover, the area must be cleared before the tent is set up.

*See Appendix A.

A three-foot-deep, 8-inch diameter hole must be dug in the center of the cleared area. This hole is used for shielding the radiation detectors. Two men, using a hand-operated post hole digger with a special oversized blade, can dig this opening in almost any terrain.

The center of the transport container bottom (Figure 1), is located over the hole, using the centering guide shown. The container is lowered via wheel jacks and the wheels removed and stored. At this point the container sides are opened (Figure 2), and the extra triangular sections located as shown. Thus the transport container provides a rigid base for supporting the floor of the laboratory.

The clean air plenum is provided by an air-blower-supported nylon-and-rubber tent. This tent has an air lock area (Figure 2) with access to the main part of the lab. The air lock serves as the decontamination area for personnel entering the laboratory and as sample storage area. The walls of the tent, including the air lock, are integrated in an air-tight seal, with a floor covering of material similar to that used in the tent walls. This fabric floor covering is securely attached to the rigid floor. (See the cross-hatched area of Figure 2.) Inflating the tent requires only start up of the internal motor generator system.* Less than thirty minutes is necessary to completely inflate the laboratory shelter. To prevent damage to the laboratory structure due to high winds, it is necessary to secure the tent to the ground by means of ropes and stakes located around the tent periphery.

*The process of applying power to the air blower also turns on the high voltage power supplies, and after an appropriate time delay supplies power to the photomultipliers, so that they are stabilized by the time the shelter has been inflated. Heat dissipation problems are minimized by the use of solid state electronics.

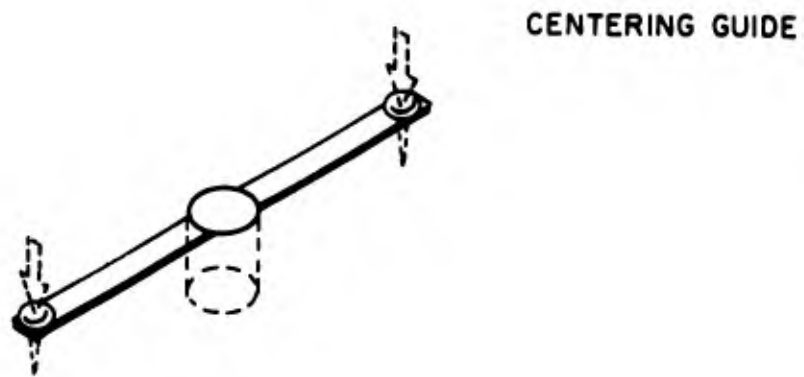
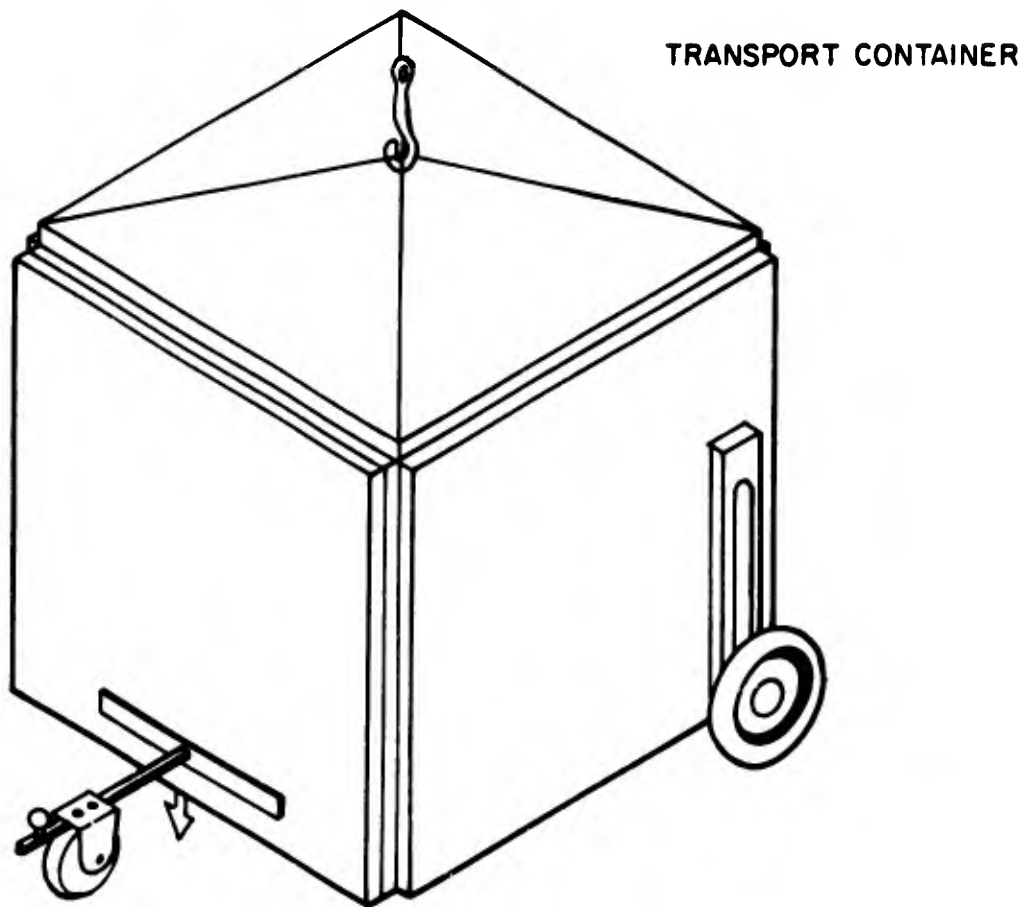


FIGURE 1. MEREL TRANSPORT CONTAINER AND CENTERING GUIDE

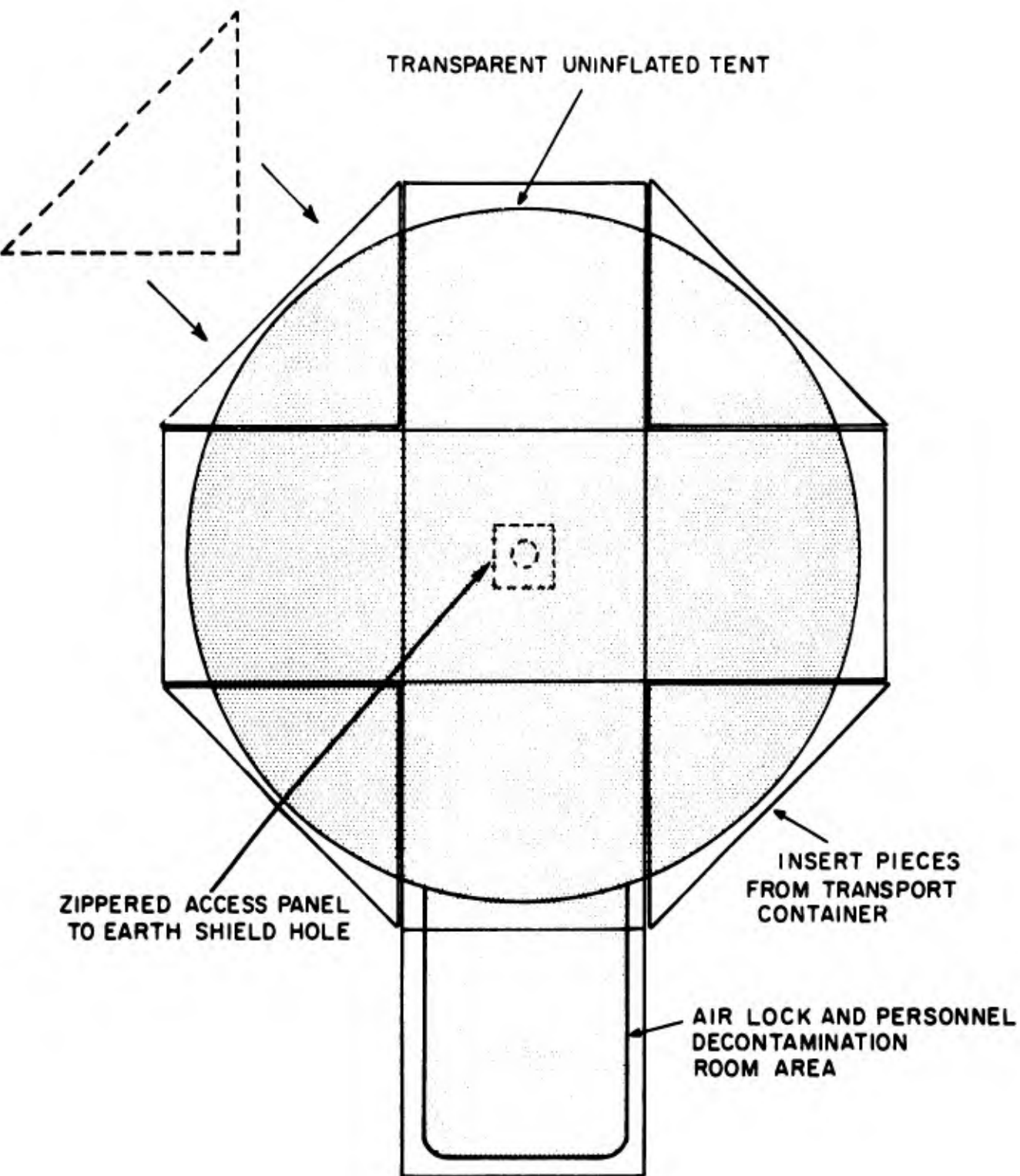


FIGURE 2. UNINFLATED LABORATORY SHELTER SHOWING RIGID FLOOR PROVIDED BY TRANSPORT CONTAINER

With the shelter inflated, Figure 3, the laboratory is ready for operation. A trained technician enters the airlock, changes to uncontaminated garments and then enters the central laboratory area. The detector package as well as the electronic components and laboratory supplies are stored at all times in the central section of the shelter.* The detector package, Figure 4, is lowered through the access panel into the hole and is ready to use. The electronic components associated with the detector package are located as shown in Figure 3. The instrument racks are rigidly mounted to the transport container via vibration and shock mounts. Support structures, such as benches, tables, storage racks, and laboratory supplies can now be assembled. The hardware is all light-weight aircraft-type and is easily stored when dismantled.

PRELIMINARY GAMMA RADIATION MONITOR FOR NO. 10 CONTAINERS

Initial monitoring of unopened containers and samples is necessary so that those having very high counting rates will be eliminated from further analysis. Initial monitoring is carried out using a scintillation-type ratemeter, because its response is inherently faster than ionization chamber ratemeters. Plastic scintillators are coupled to photomultiplier tubes and used with standard ratemeter circuitry. By using two detectors, one for monitoring background only, a net activity count rate for each sample is determined; i e , the current from the background monitor is automatically subtracted from the container monitor current. Rise times of scintillation ratemeters are typically less than five seconds, so that the total monitoring time for each container is less than ten seconds.

* It should be noted that at no time during the transporting and setting up of the laboratory has the internal environment of the laboratory been disturbed or exposed to fallout contamination, water, dirt, etc

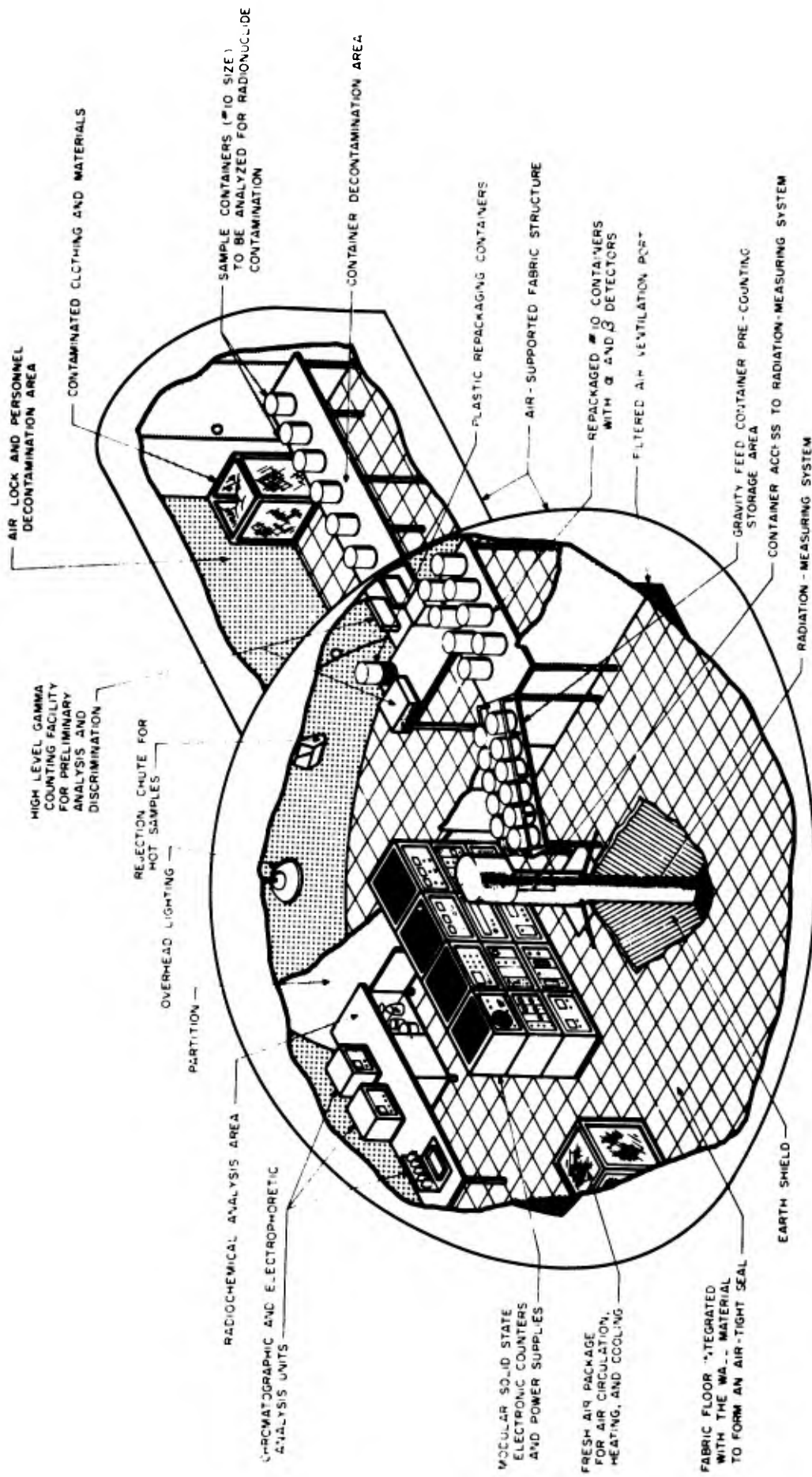


FIGURE 3. MOBILE ENVIRONMENTAL RADIATION EVALUATION LABORATORY

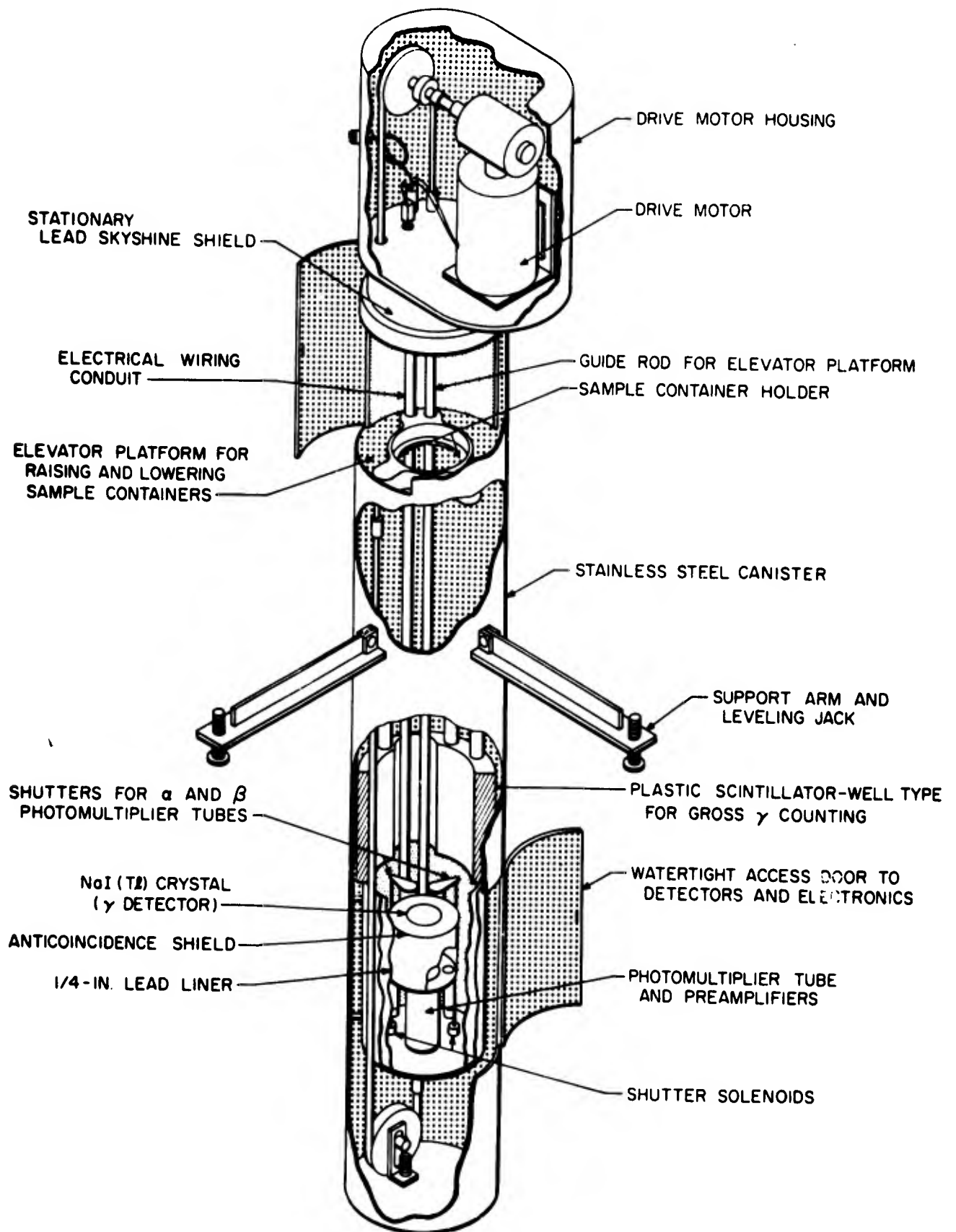


FIGURE 4. RADIATION-MEASURING SYSTEM

If a high count rate is observed, a sign indicating this fact is illuminated. The actual count rate is indicated on a meter and can also be recorded on a chart. If an acceptable level of radioactivity is indicated, the container is advanced along for further processing and analysis.*

* An acceptable level here means one which will not have a counting rate higher than can be handled by the very sensitive counting equipment used for qualitative and quantitative alpha, beta, and gamma determinations.

PLASTIC PACKAGING CONTAINERS WITH INTEGRATED ALPHA AND BETA DETECTORS

In order that radioactive contaminants residing on the exterior surfaces of the No 10 containers or other sample containers do not contaminate the nuclear counting apparatus, these containers must be completely repackaged in a durable container. A dual purpose may be served by this procedure; not only is the spread of contamination limited, but also by opening and recapping the No 10 container with a snap-on type plastic cap in which alpha and beta detectors are imbedded, a geometrical configuration is created in which 50 per cent and 30 per cent counting efficiencies should be realizable for the alpha and beta detectors respectively. The alpha detector is mylar-backed zinc sulfide.* A one-mil thick plastic scintillator, such as NE102, serves as the beta detector.** Both detectors are too thin to have any substantial gamma sensitivity. The plastic cap is integrated into a plastic bag and thus forms the encapsulating container (Figure 5). Inverting the container so that the cap with the detectors is on the bottom allows the open end of the plastic bag section to be heat-sealed. It also brings the container's contents into direct contact with the alpha and beta detectors.

A second type of plastic container (dimensionally the same diameter as a No 10 container) is necessary to repackage the contents of other containers and to package all biological specimens so that a standard counting geometry will be established. This container is fabricated of polyethylene and has a snap-on, water-tight lid. The alpha and beta detectors are located in the bottom of the container and are again in direct contact with the container's contents. Pliable specimens of irregular shape

* Available from William B Johnson and Associates, Mountain Lakes, New Jersey

** Available from Nuclear Enterprises Limited

- A. NO. 10 CONTAINER
- B. CONTAINER WITH LID REMOVED
- C. ALPHA AND BETA DETECTORS MOUNTED ON LID OF WATER-TIGHT PLASTIC PACKAGE IN POSITION TO ENCLOSE CONTAINER (D.)
- E. PLASTIC BAG BEING INSTALLED TO ENCLOSE CONTAINER
- F. ENCLOSED CONTAINER INVERTED AND SEALED
- G. CROSS SECTION OF BOTTOM OF DETECTOR CONTAINER

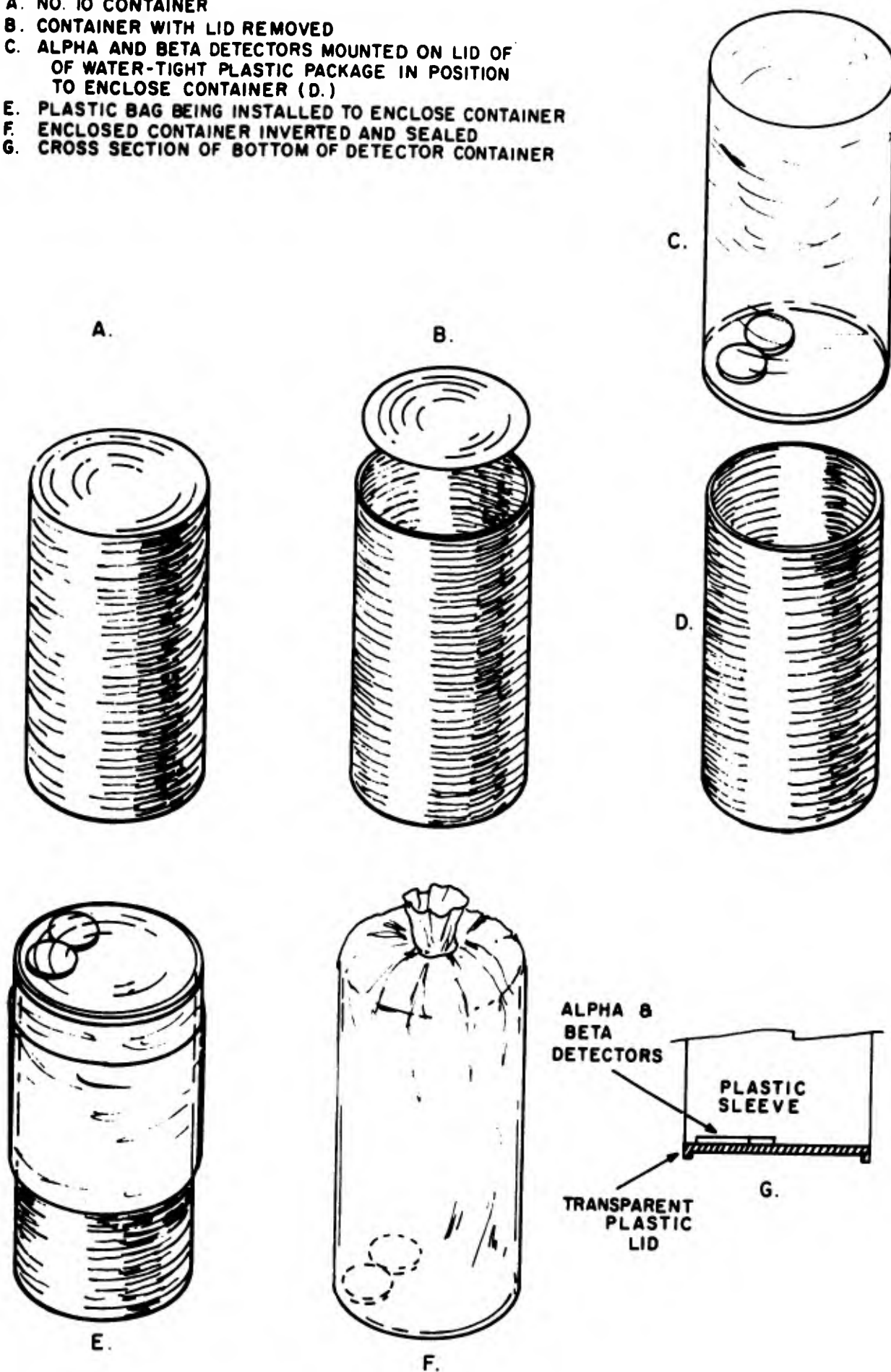


FIGURE 5. SEQUENCE FOR PACKAGING A RADIOACTIVE CONTAINER

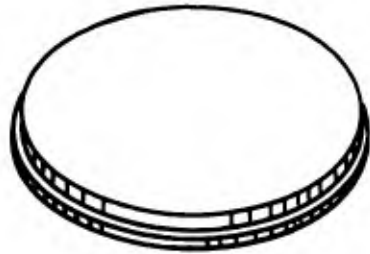
can be compressed into a standard geometrical configuration, using the simple plunger arrangement as shown in Figure 6.

ROUTINE HANDLING PROCEDURE FOR NO. 10 TYPE CONTAINERS

The procedure for analyzing any No. 10 container, appropriately packaged and identified, is as follows

1. The containers are stored in the decontamination area of the laboratory shelter
2. Each container whose point of origin is outside the shelter must be wiped off and placed in position between the preliminary monitors. A 10-second count is initiated, and if a high counting rate is registered, the container is rejected and manually deposited outside the laboratory via the rejection chute.
3. If the container does not have an unacceptable gamma radioactivity level, it is placed in a storage rack. At this point it will have the lid removed and a numbered plastic container with an alpha and a beta detector imbedded in the bottom will be placed over it. With the plastic container in position, the No. 10 container is inverted and a water-tight heat seal effected at the open end of the plastic bag.

All biological specimens are repackaged in No 10 size plastic containers. This is done in the laboratory space adjacent to the decontamination area. Solid materials have to be compressed into a standard geometry using a simple piston arrangement as shown in Figure 5. The piston is left in the container after the standardization procedure has been carried out. The samples are counted and analyzed in the central area of the laboratory in the same manner as the plastic-encapsulated, recapped No 10 tin containers



COVER

PLUNGER AND PLASTIC
PACKAGING CONTAINER

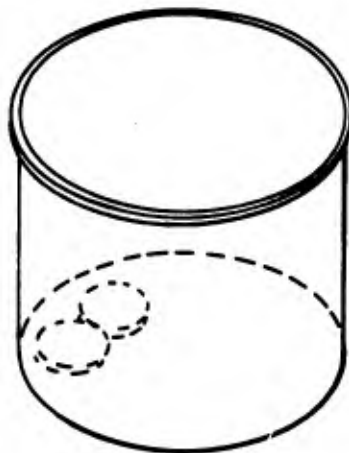


FIGURE 6. PLASTIC PACKAGING CONTAINER AND PLUNGER
FOR STANDARDIZING SAMPLE-COUNTING GEOMETRY

4. The packaged No. 10 container is now rack-stored in preparation for transfer into the counting room area of the laboratory. The actual transfer is accomplished through an airlock with the container sliding into position on an inclined storage rack.
5. Each container is placed, one at a time, in the detector canister and lowered into contact with the photomultiplier tubes and gamma radiation detectors (Figure 4). The container is keyed so that proper orientation is maintained for locating and aligning the alpha and beta detectors with the appropriate photomultipliers.
6. A pre-set counting and analysis time of less than one minute is necessary. When the count rate from all the detectors is below a specified level, the container is automatically rejected, a safe level of contamination is indicated on a visual display, and the electronic counting program is stopped and reset in preparation for the next container.
7. The batch number of each analyzed container is noted, and the container stored temporarily in the counting room area.
8. If a sample does contain contamination and the analysis is run to completion, then all the pertinent information - count rate for alpha, beta, and gamma, as well as all spectral information - is recorded and appropriately identified.

CHROMATOGRAPHIC ANALYSIS

Specimens, such as small quantities of blood and those requiring more detailed radioanalysis for type and quantity of isotopic constituents, require special handling and preparation. The most convenient schemes for separating the components and doing qualitative as well as quantitative analyses utilize differential migration techniques such as paper chromatography and electrophoresis, possibly combined with limited radiochemical procedures. Alpha, beta, and gamma analyses are then carried out using the same instruments described for analyzing No. 10 containers.

Alpha and beta counting of paper chromatograms are done in a specially constructed No. 10 size container which has anthracene and CsI(Tl) crystals for the beta and alpha detectors respectively. A motor-drive assembly in the container steps the chromatogram strip past the detectors at a predetermined rate. All counting procedures and analyses are carried out in the shielded environment of the canister.

In the situation where only gamma analysis is required, the chromatogram strip can be rolled into the form of a small cylinder and placed in a jig to locate it in the well of the canister's NaI(Tl) crystal. In the rolled-up configuration, the problem of self-absorption due to the paper should be negligible except for very low energy X-rays.

DETAILS OF THE CANISTER AND ALPHA BETA AND GAMMA DETECTORS

Simultaneous analyses for alpha, beta and gamma contamination of the contents of No. 10 containers are performed in the stainless steel detector canister shown in Figure 4. The canister, six feet long and eight inches in diameter, is supported by three extensions and is aligned vertically with leveling jacks. The detector package end of the canister resides in

the earth shield hole in the center of the laboratory, with the detectors about two feet below the ground level. The carriage holding the No. 10 containers travels at a rate of 0.8 feet per second and reaches positive stops in both the up and down positions. A water-tight access is provided opposite the detector section so that servicing and installation may be effected without removing the carriage lift mechanism. Loading and unloading of sample containers is done through a light-tight door near the upper end of the canister.

A $\frac{1}{2}$ -inch thick lead shield surrounds the detector package to reduce local background from natural radioactivity found in the soil. A 2-inch thick by 8-inch diameter lead plug resides permanently near the top of the canister to attenuate skyshine radiation.*

The detector package contains two photomultiplier tubes and their preamplifiers for detecting the light output of the alpha and beta detectors located in the No. 10 container package.** Light-tight shutters operated by rotary solenoids cover the alpha and beta detectors' photomultiplier tubes when there is no container in the counting position. A collimated, 2-inch diameter by 3-inch long, well-type NaI(Tl) crystal, integrated with a photomultiplier tube and preamplifier, constitutes the gamma detector for use with a pulse height analyzer to qualitatively identify gamma constituents and to estimate their concentrations. For this application a plastic scintillator anticoincidence shield is needed around the NaI(Tl) crystal to reduce background counts and improve the peak-to-total ratio of the spectra taken with the NaI(Tl) crystal (See Figure 7.) Two 1-inch diameter photomultiplier tubes are coupled to the anticoincidence shield.

* The entire canister including the detectors will weigh about 250 pounds.

** The alpha and beta detectors are aligned with their respective photomultipliers by proper keying of the No. 10 container.

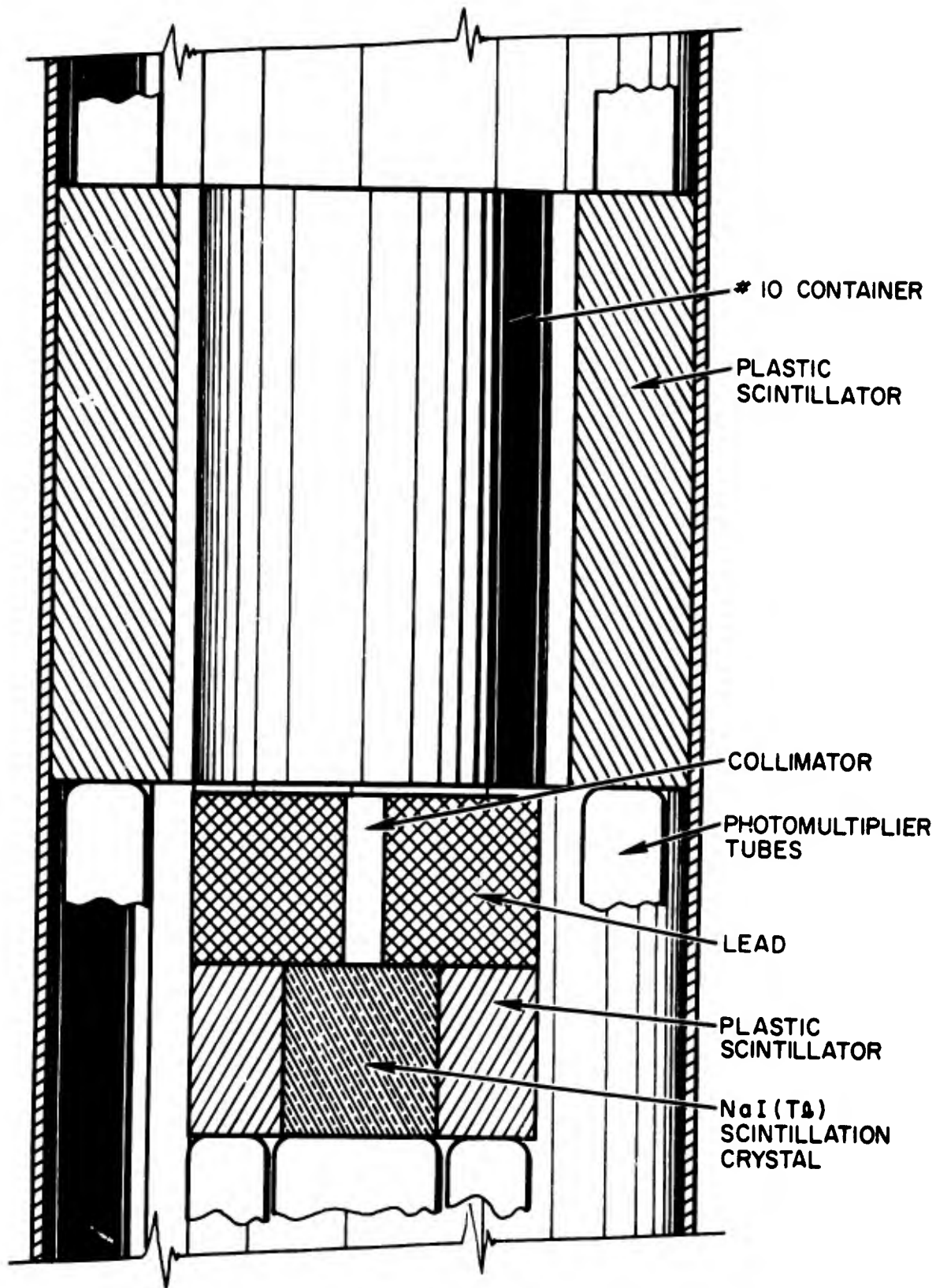


FIGURE 7. GAMMA DETECTION SYSTEM FOR GROSS COUNTING AND PULSE ANALYSIS

Gross counting of the gamma activity in the No 10 containers is done with a hollow cylindrical plastic scintillator. The container resides within the cylinder while in position for alpha and beta counting. The inner dimensions of the cylinder are the same as those of the No. 10 container (10.7 cm in diameter by 17.7 cm high). Eight photomultipliers are coupled to this plastic scintillator.

COUNT RATE ASSOCIATED WITH A 2-MR/HR GAMMA FIELD

The dose rate (R) in roentgens per hour is given by (3)

$$R = N_o E_o \mu_a / \rho$$

where N_o is the counting rate in gamma/cm²-sec

E_o is the photon energy

μ_a is the total absorption coefficient

ρ is the density

One roentgen equals $5.24 \times 10^7 \frac{\text{mev}}{\text{g}}$

$$\text{so } 1 \frac{\text{mr}}{\text{hr}} = \frac{5.24 \times 10^7 \text{ mev/g} \times 10^{-3} \text{ mr/r}}{3600 \text{ sec}} = \frac{N_o E_o \mu_a}{\rho} \gamma' \text{ s/sec-cm}^2$$

and μ_a/ρ for dry air at standard conditions of temperature and pressure equals $6.35 \times 10^{-2} \text{ cm}^{-1}$. Therefore, $N_o = 237.5 \gamma/\text{cm}^2\text{-sec}$ for a 1-mr/hr gamma field whose average energy is 1 Mev. The counting rate expected from a 100 per cent efficient, 1 cm² detector in a 2-mr/hr field is 475.0 counts/sec.

DETECTOR SHIELDING

In situations where the weight of the instruments associated with nuclear radiation detectors and detector shielding are not severely limited, sufficient lead, iron, mercury-filled

voids, concrete, etc., may be used to reduce the background radiation levels so that low level alpha, beta and gamma counting can be accomplished. The present requirements preclude the transportation of heavy shielding to the site. Therefore, either coincidence/anti-coincidence techniques must be employed to discriminate against background counts, or use must be made of materials available at the site for shielding purposes.

High counting rates are necessary to obtain reasonable counting times using coincidence/anti-coincidence systems. Since small concentrations of isotopes in foods and biologicals (low counting rates) must be detectable, this approach does not appear useful in rapid, gross counting applications.

For field operations, the earth itself is the most easily used material for radiation shielding. The earth shield will be, for the purposes of this study, a hole eight inches in diameter and three feet deep. Assume a detector package residing two feet below the surface of the ground and in the center of a radioactive contamination-free zone fifteen feet in diameter. The shielding provided by seven feet of earth between the detectors and the outside of the clean air plenum may be calculated by assuming the fallout source to be plane isotropic (see Figure 8).

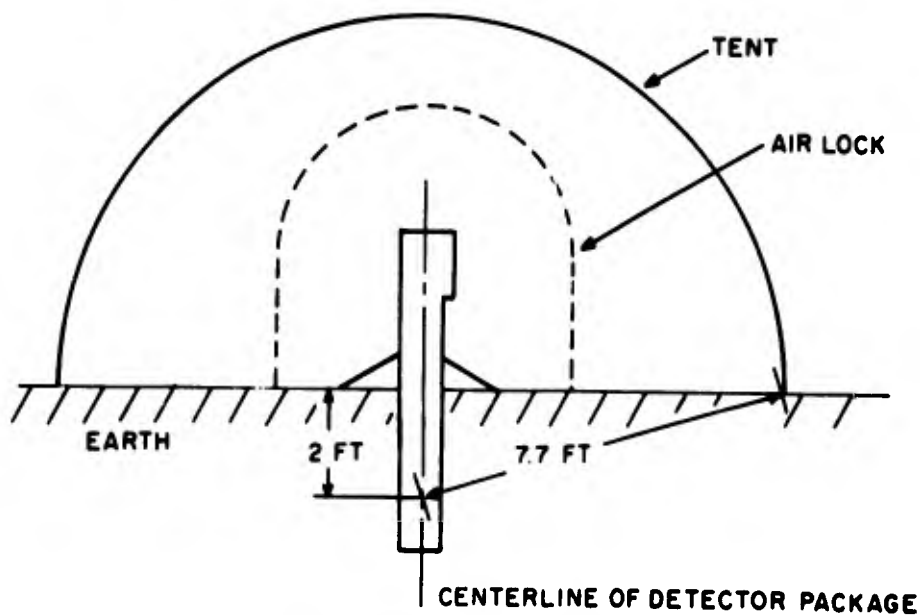


FIGURE 8. DETECTOR SHIELDING GEOMETRY

The flux reduction factor is then given by

$$\frac{N}{N_0} = \frac{e^{-\mu x}}{2\mu x}$$

where x = absorbing medium thickness (cm)

μ = attenuation coefficient (0.0706 cm²/g for water)*

N_0 = unshielded counting rate

N = shielded counting rate

ρ = density of the absorbing medium (g/cm³)

Thus, $N/N_0 = 1.01 \times 10^{-8}$ for an average photon energy of 1 Mev. (A lead shield would have to be 73-inches thick to achieve this same attenuation.) Thus a lead shield with a 6-inch diameter by 6-inch high detector cavity would weigh about 730 pounds, and this is too heavy for the laboratory to meet the specified weight requirements.

The skyshine contribution factor is 0.0013 as calculated for an 8-inch diameter hole 3 feet deep, from the equation⁽⁴⁾

$$D/D_0 = 1.2S(d + \tau)S_a(d + \tau, w)$$

This must also be reduced by the absorption due to any skyshine shielding. If a 2-inch thick lead skyshine shield is used, the transmission factor for 1-Mev gammas is 0.0196, making a total skyshine transmission contribution factor of 2.44×10^{-5} . Thus a flux at the top of the skyshine shield of 2 mr/hr = 475 cts/sec-cm² would be reduced to a flux of $475 \times 2.55 \times 10^{-5} = 0.012$ cts/sec-cm² at the position of the gamma detectors below the shield.

*The attenuation coefficient for water is used as an approximation for the attenuation coefficient for earth.

SENSITIVITY OF THE ALPHA DETECTOR

Assume a 1- μ curie alpha source uniformly distributed throughout the volume of water required to completely fill a No. 10 container. Also assume the alpha particle has an energy of 2 Mev. The mean range of an alpha particle R_A is given by⁽⁵⁾

$$R_A \text{ (mg/cm}^2\text{)} = 0.56R(\text{cm})A^{1/3} \quad \text{(Equation 1)}$$

where R is the particle range in dry air at standard conditions and A is the atomic weight of the material. Then a 2-Mev alpha particle has a range in water of

$R_A = 2.93 \times 10^{-3} \text{ g/cm}^2$, or dividing by the density of water,

$$R = 2.93 \times 10^{-3} \text{ cm}$$

The total activity recorded by a 4π detector surrounding a 1-cc volume of H_2O would arise from alpha particles originating no more than 0.00293 cm from the surface. The total number of alphas emerging from one face of the unit volume is given by

$$N = 3.7 \times 110^{10} \frac{\text{dis}}{\text{curie}} \times 1 \times 10^{-6} \text{ curies} \times \frac{V'}{V} \times \frac{1}{A \text{ cm}^2} \quad \text{(Equation 2)}$$

where V , the volume of the No. 10 container, is 1591 cm^3 , A is the surface area of a 1-cm^3 cube and V' the total volume from which alphas may emerge, is given by

$$\begin{aligned} V' &= 1 \text{ cm}^3 - (1.00 - 2R)^2 \times 1 \text{ cm}^2 \\ &= 1 \text{ cm}^3 - [1.000 - 2(0.00293)]^2 \times 1 \\ &= 0.0117 \text{ cm}^3 \end{aligned}$$

Then $N = 0.045 \alpha' \text{ s/sec-cm}^2$

The area of the detector photomultiplier is 5.06 cm^2 (1-inch diameter photocathode). Therefore, given a 50% counting efficiency for the detector, the total counting rate is

$$N_T = 0.14 \text{ counts/second}$$

For 6.3-Mev alphas, the range in water is $7.4 \times 10^{-3} \text{ cm}$. Therefore the counting volume V' is 0.029 cm^3 , and the counting rate for the alpha detector becomes

$$N_T = 0.28 \text{ counts/second}$$

On this basis, an alpha concentration necessary to give a reasonable counting time would be about $6 \times 10^{-3} \frac{\mu\text{c}}{\text{cm}^3}$.

Since the major portion of alpha energies lies between 4 and 6 Mev, a counting rate of about 100 counts per minute could be expected from the distributed source.* A background counting rate of 2 counts per minute is reasonable to assume, with the shielding provided, and so a one minute counting time would be adequate for gross alpha analysis.

SENSITIVITY OF THE BETA DETECTOR

The method used to calculate the sensitivity of the alpha detector can also be used to calculate the beta detector's sensitivity. Equation 2 will be used along with the assumption that a 1- μ curie beta source is uniformly distributed in a No. 10 container filled with water.

The extrapolated ranges of 0.1 Mev and 0.6 Mev betas in water are 0.143 cm and 0.227 cm respectively.⁶⁾ Assuming a detector efficiency of 30% and a detector cross sectional area of 5.06 cm^2 , the

*Assuming a 50 per cent counting efficiency

net counts per second N_T to be expected for the 0.1 and 0.6 Mev betas are 2.9 c/sec and 10.7 c/sec respectively. Background counting rates should be approximately 10 counts per minute or less. Hence in the two cases cited, a one-minute counting period is adequate for gross analysis of beta source activities equivalent to a source concentration of $6 \times 10^{-3} \frac{\mu\text{c}}{\text{cm}^3}$.

GROSS GAMMA COUNTING CAPABILITY

The plastic phosphor scintillator used for gross counting of low level gamma activity in No. 10 containers (See Figure 7) has a surface area given by

$$A = 2\pi (r_2^2 - r_1^2) + 2\pi r_2 l \quad (\text{Equation 3})$$

where r_1 = inner cylinder radius = 6.2 cm

r_2 = outer radius = 9.8 cm

l = cylinder height = 17.7 cm

$$A = 2\pi (r_2^2 - r_1^2 + r_2 l)$$

$$A = 535 \text{ cm}^2$$

The detector efficiency, assuming a plastic composition similar to methyl-methacrylate, is given by

$$\epsilon = 1 - e^{-\mu x} \quad (\text{Equation 4})$$

where $\mu_a = 0.0681 \frac{\text{cm}^2}{\text{g}}$ ($\mu = \mu_a \rho$)

$\rho = 0.936 \text{ g/cm}^3$

$x = 1.6 \text{ cm}$

Thus $\epsilon = 0.095$, and in a 2-mr/hr field, the unshielded detector would evidence a background counting rate of

$$R = (475 \frac{\text{cts}}{\text{s-cm}^2}) (535 \text{ cm}^2) (0.095)$$

$$R = 2.42 \times 10^4 \text{ counts/second}$$

Enclosing the detector in the earth shield reduces the counting rate by $1.01 \times 10^{-8} \frac{\text{cts}}{\text{sec}}$, so that the shielded counting rate (R_s) becomes

$$\begin{aligned} R_s &= 2.42 \times 10^4 \times 1.01 \times 10^{-8} \frac{\text{cts}}{\text{sec}} \\ &= 2.44 \times 10^{-4} \text{ cts/sec} \end{aligned}$$

As an example of the applicability of the preceding calculations to determining the sensitivity of the cylindrical plastic scintillator detector for gross counting applications, consider the case of a distributed gamma source whose activity is 1 μc and which is uniformly distributed throughout the volume of a No. 10 container filled with water. Assume a mean value distance equal to the container radius for each gamma photon. Then the reduction factor is

$$\begin{aligned} \frac{N}{N_0} &= e^{-\mu x} && \text{when } \mu = 0.0706 \frac{\text{cm}^2}{\text{g}} \\ &= 0.684 && \rho = 1.00 \text{ g/cm}^3 \\ &&& x = 5.35 \text{ cm} \end{aligned}$$

The area of the container from which emerging gamma rays can enter the plastic scintillator is $A_s = \pi Dh$. The percentage of gammas emerging from this portion of the container is

$$\frac{\pi Dh}{2\pi r^2 + \pi Dh} = 0.768 = 76.8\% \quad (\text{Equation 5})$$

The total number of gammas emerging (ignoring self-absorption effects) from the container is 3.7×10^4 and the number reaching the plastic scintillator is

$$0.768 \times 3.7 \times 10^4 \frac{\gamma}{s} = 2.84 \times 10^4 \frac{\gamma}{s}$$

Because of the absorption $e^{-\mu x}$ in the water and the low efficiency ϵ of the plastic for 1 Mev photons, the total number of events occurring in the scintillator is

$$2.84 \times 10^4 \times \epsilon \times e^{-\mu x} = N$$

$$1848 \frac{\text{cts}}{\text{sec}} = N$$

TYPICAL MEREL SYSTEMS AND COMPONENTS DESCRIPTION

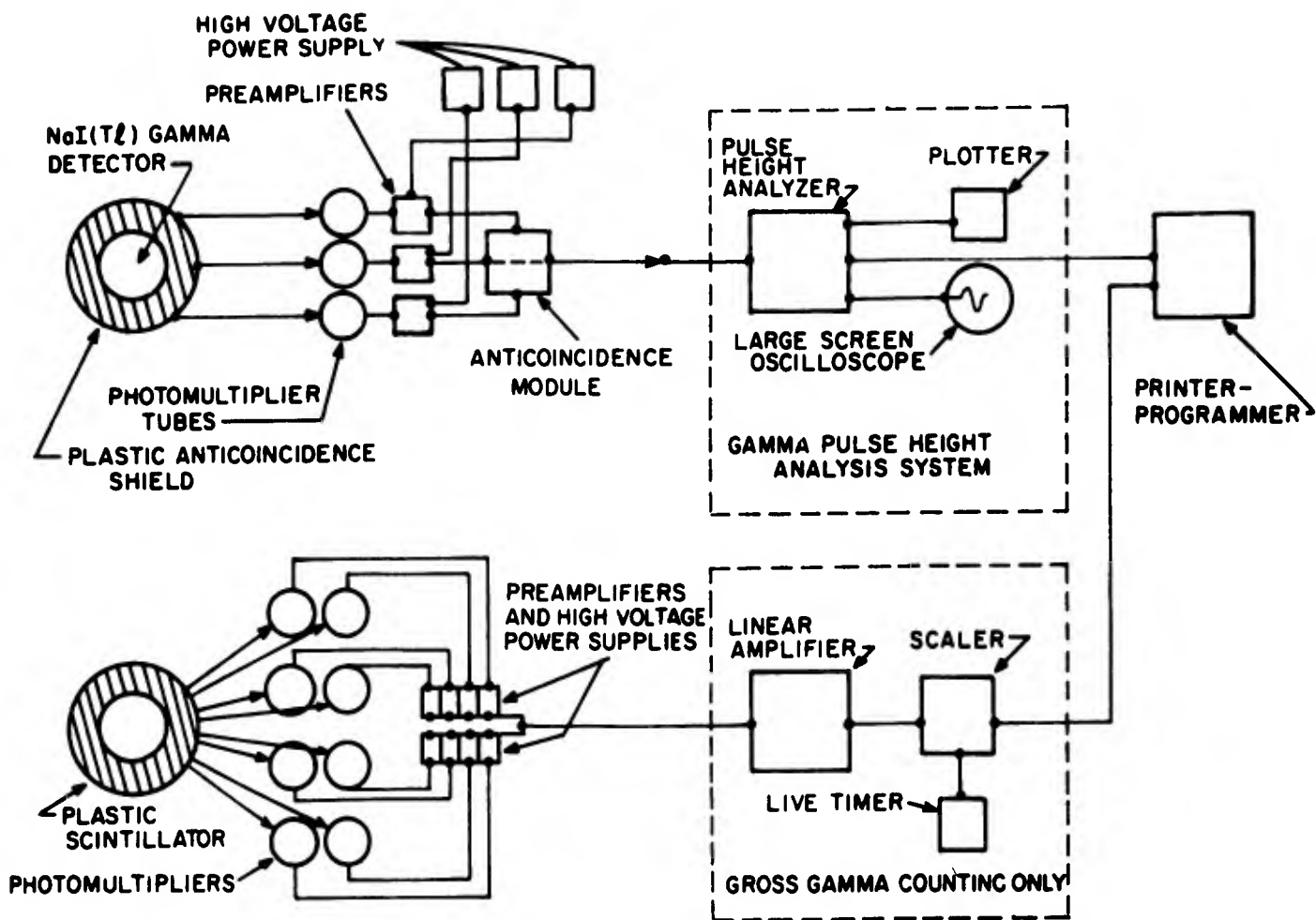
Block diagrams of the various nuclear radiation detection systems in the Mobile Environmental Radiation Evaluation Laboratory are shown in Figures 9 and 10. The dashed rectangular areas enclose the electronic components of each detector system which may be varied to increase or decrease the analytical capabilities of the system. All the systems illustrated can be assembled with commercially available components except the scintillator-photomultiplier combinations for gross gamma analysis (plastic well scintillator), alpha analysis, and beta analysis. Integrated sodium iodide crystals and photomultipliers are readily available from several companies.*

Some of the commercial units can perform more than one function; thus some consolidation is possible. For example, an RIDL Designer Series High Voltage Distribution Panel, Model 40-11, provides four individually variable output voltages between 500 and 3000 volts at 2 milliamperes; yet, the entire chassis is 8-1/16" wide by 8-9/16" high by 10" deep and weighs 5 lbs, 10 oz.** Appendix B lists a typical set of commercial components which will perform all the required functions. The RIDL Designer Series is used as the basis for the systems, but this should not be construed to mean that other manufacturers' components could not be used equally well. The Designer Series components will operate reliably over the ambient temperature range of 10°C to 40°C.***

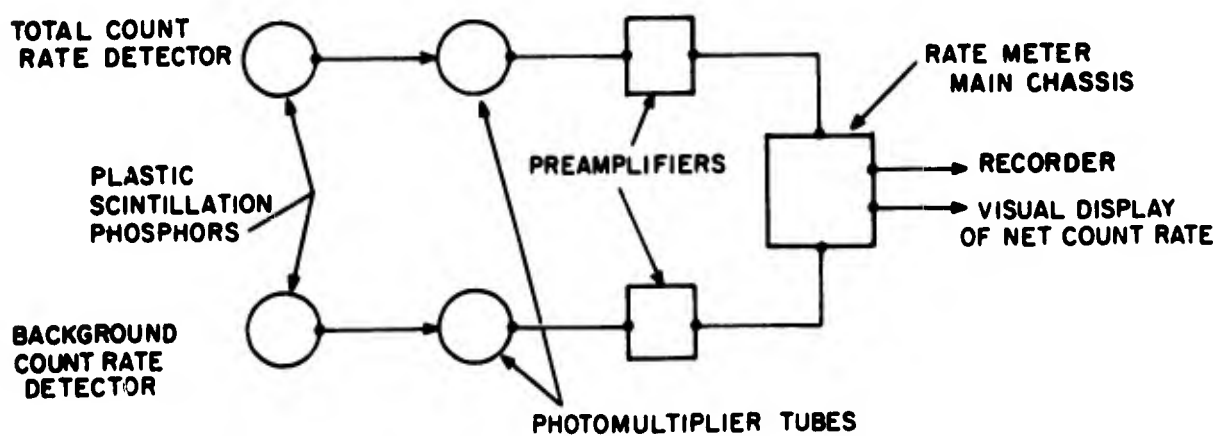
* Integral-line NaI(Tl) crystals and photomultipliers are available from the Harshaw Chemical Company, Cleveland, Ohio, and Isomet Corporation, Palisades Park, New Jersey.

** RIDL is the Radiation Instrument and Development Laboratory of Melrose Park, Illinois.

*** Private communication, RIDL Corporation.

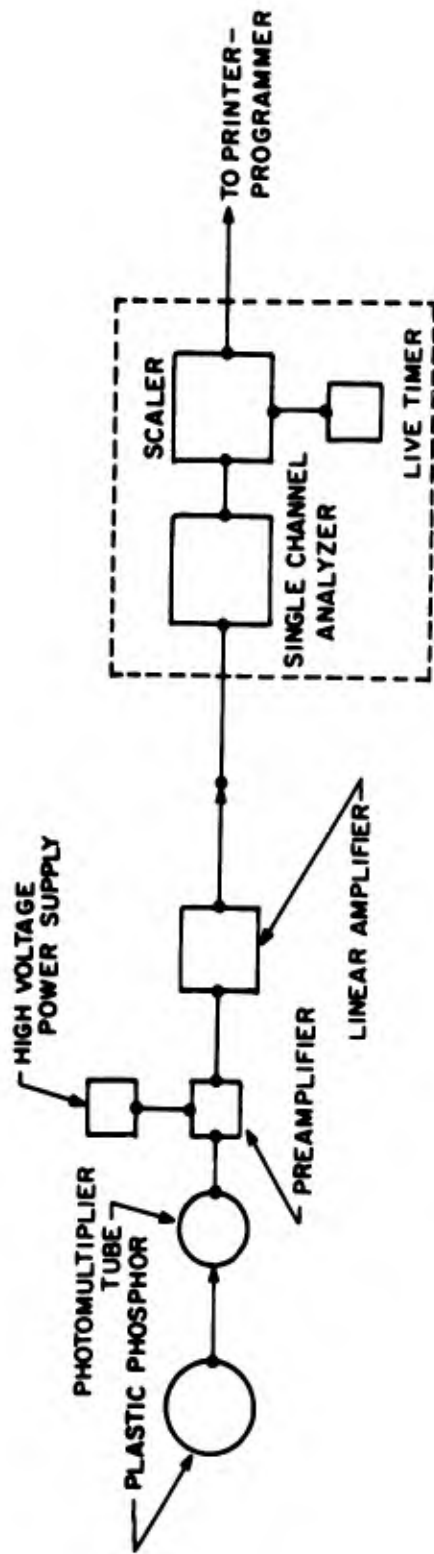


A. CANISTER GAMMA DETECTION AND ANALYSIS SYSTEM

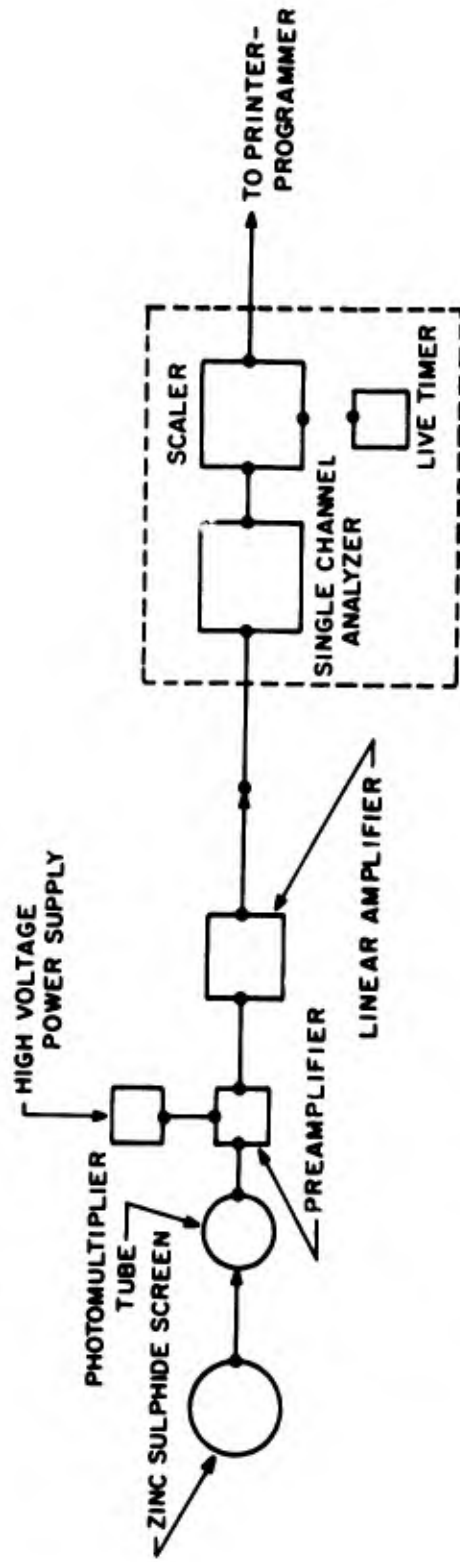


B. PRELIMINARY GAMMA MONITOR FOR NO. 10 CONTAINER

FIGURE 9. GAMMA RADIATION DETECTION SYSTEMS



A. GROSS BETA COUNTING WITH ENERGY DISCRIMINATION CAPABILITIES



B. GROSS ALPHA COUNTING WITH ENERGY DISCRIMINATION CAPABILITIES

FIGURE 10. CANISTER ALPHA AND BETA DETECTOR SYSTEMS

COST ESTIMATE FOR COMPLETE SYSTEM

The electronic components used in the mobile laboratory are available off-the-shelf. The laboratory structure and detector canister equipment are not available off-the-shelf, but can be developed using technology well within the present state of the art. This system would incorporate commercial quality items which are not developed to Military Specifications.

Approximately two man years of scientific and support labor would be required to design, develop, assemble, and check out the first complete mobile laboratory suitable for field use.

The electronic equipment required have been listed in Appendix B. The manufacturer's list price in effect at the time of publication has been shown for each piece of equipment. The total of the prices listed is \$37,437.

The developmental items have been listed in Appendix C. We have had some discussions with several firms capable of making these systems and have made a rough estimate of the cost to develop a "first unit" of each item. Individual estimates for each item are also shown. The total cost for the developmental items is estimated to be \$49,000.

Based on the above considerations, a rough estimate of the total cost for a complete mobile laboratory would be between \$150,000 and \$170,000. This estimate includes developmental costs which would not be incurred in the production of additional units; the cost of additional units is estimated to be of the order of \$80,000 to \$100,000 each.

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PART II

LITERATURE SURVEY

- Radioisotopes
- Radiation Detectors
- Radiochemical Techniques

RADIOACTIVE CONTAMINANTS

SOURCES OF FISSION PRODUCTS

A neutron-induced fission process, such as $U^{235} + n \rightarrow X + Y$, yields fission products X and Y, which are radioactive and must in turn decay, primarily by negatron emission, to other isotopes in the process of reaching a stable nuclear configuration. On the average a fission product undergoes about three successive beta transformations before becoming a stable middleweight nuclide. (7,8,op.cit.3) Consequently, there are over two-hundred such radioactive fission products. Table 1 gives the cumulative yields of various fission products from thermal neutron fissions in U^{233} , U^{235} , and Pu^{239} . (9) The half-lives of many of these isotopes are short and are of little consequence in considering the long-term effects of fallout or reactor product contamination. Table 2 shows the fission product chains with the respective half-lives and branching ratios. Of primary interest in this study is neutron activation due to a nuclear weapon detonation. The degree and type of induced radioactivity in food supplies, soils, and various other materials would be of immediate interest as would the human uptake of these radioactive materials. The qualitative identification and quantitative determination of the concentration of these isotopes is extremely difficult. In general, these tasks require radiochemical separation, particle identification, energy determination, and counting rate measurements.

Table 1. * CUMULATIVE YIELDS OF VARIOUS FISSION PRODUCTS FROM THERMAL NEUTRON FISSIONS IN U²³³, U²³⁵, AND Pu²³⁹

s = second m = minute h = hour d = day y = year * = metastable

Mass No.	Fission Product	U ²³³ % Yield	U ²³⁵ % Yield	Pu ²³⁹ % Yield ^{a, b, c}	Mass No.	Fission Product	U ²³³ % Yield	U ²³⁵ % Yield	U ²³⁵ % Yield	Pu ²³⁹ % Yield ^{a, b, c}
72	Zn ⁷² (49h)			0.00012	96	Zr ⁹⁶ (stable)	5.60			
77	Ge ⁷⁷ (12h)	0.010	0.0031 ^d		97	Zr ⁹⁷ (17h)		5.9 ^d		5.7
77	As ⁷⁷ (38.7h)	0.019			97	Mo ⁹⁷ (stable)	5.35			
78	Ge ⁷⁸ (86m)		0.01 ^d		98	Mo ⁹⁸ (stable)	5.18			
81	Se ⁸¹ * (57m)		0.0084		99	Mo ⁹⁹ (66h)	4.8			5.9
83	Se ⁸³ (25m)		0.22		100	Mo ¹⁰⁰ (stable)	4.40			
83	Br ⁸³ (2.4h)	0.75	0.51	0.085	101	Mo ¹⁰¹ (14.6m)		~5.6		
83	Kr ⁸³ (stable)	1.14			101	Ru ¹⁰¹ (stable)	3.00			
84	Br ⁸⁴ (31.8m)		0.90		102	Mo ¹⁰² (11.5m)		4.3		
84	Kr ⁸⁴ (stable)	1.50			102	Ru ¹⁰² (stable)	2.37			
85	Kr ⁸⁵ (10.3y)	0.56	0.293 ^d		103	Ru ¹⁰³ (39.7d)	1.6			5.8
86	Kr ⁸⁶ (stable)	3.18			104	Ru ¹⁰⁴ (stable)	0.96			
87	Br ⁸⁷ (55s)		3.1		105	Rh ¹⁰⁵ (35.3h)				3.9
89	Sr ⁸⁹ (51d)	6.5			106	Ru ¹⁰⁶ (1.01y)	0.28			5.0
90	Sr ⁹⁰ (28y)		5.8 ^d	1.5	109	Pd ¹⁰⁹ (13.4h)	0.040	0.030		1.5
91	Sr ⁹¹ (5.7h)		5.8 ^d	2.4	111	Ag ¹¹¹ (7.6d)	0.025			0.27
91	Sr ⁹¹ (2.7h)		5.3		112	Pd ¹¹² (21h)	0.016			0.10
91	Y ⁹¹ (58d)		5.35	3.0	115	Ag ¹¹⁵ (21m)		0.0077		
91	Zr ⁹¹ (stable)	6.53			115	Cd ¹¹⁵ * (43d)	0.001	0.0007		0.003
92	Zr ⁹² (stable)	6.5			115	Cd ¹¹⁵ (53h)	0.019	0.0097		0.038
93	Zr ⁹³ (1.1x10 ⁶ y)	7.10			121	Sn ¹²¹ (27.5h)	0.018			0.044
94	Y ⁹⁴ (16.5m)		5.4		123	Sn ¹²³ (136d)		0.0013		
94	Zr ⁹⁴ (stable)	6.82			125	Sn ¹²⁵ (9.6d)	0.050	0.013		0.072
95	Zr ⁹⁵ (65d)	5.9	6.2	5.9	127	Sb ¹²⁷ (91h)				0.39
95	Mo ⁹⁵ (stable)	6.10								

* Reference 4

a. The Cs, N, and yields are corrected to 5.6% yield for Ba¹⁴⁰.

b. Most values are corrected to a Ba¹⁴⁰ yield of 1.06 x 5.32% = 5.68%.

c. The xenon isotope yields are normalized to corrected Cs yields at mass 133.

d. Average value.

e. Measured value is 6.32%. The factor 1.06 is introduced to bring the total yield for light and heavy groups to 100% each.

Table 1. * (Continued) CUMULATIVE YIELDS OF VARIOUS FISSION PRODUCTS FROM THERMAL NEUTRON FISSIONS IN U²³³, U²³⁵, AND Pu²³⁹

s = second m = minute h = hour d = day y = year * = metastable

Mass No.	Fission Product	U ²³³ % Yield	U ²³⁵ % Yield	Pu ²³⁹ % Yield ^{a,b,c}	Mass No.	Fission Product	U ²³³ % Yield	U ²³⁵ % Yield	Pu ²³⁹ % Yield
127	Te ^{127*} (105d)		0.035		140	Ce ¹⁴⁰ (stable)	5.6		5.68
129	Te ^{129*} (37d)		0.35		141	Ce ¹⁴¹ (33d)			5.2
131	Sb ¹³¹ (23m)		2.6		142	Ce ¹⁴² (stable)	5.6		6.69
131	Te ^{131*} (30h)		0.44		143	Ce ¹⁴³ (33h)		5.7	5.4
131	I ¹³¹ (8.05d)	2.7	3.1 ^a	3.8	143	Nd ¹⁴³ (stable)	5.2		6.31
131	Xe ¹³¹ (stable)	3.74		2.87	144	Ce ¹⁴⁴ (285d)	4.1	6.0	5.28
132	Te ¹³² (77h)		4.7	5.2	144	Nd ¹⁴⁴ (stable)	4.0		5.29
132	Xe ¹³² (stable)	5.10		4.02	145	Nd ¹⁴⁵ (stable)	3.0		4.24
133	Sb ¹³³ (4.1m)		4.0		146	Nd ¹⁴⁶ (stable)	2.3		3.35
133	Te ^{133*} (63m)		4.5		147	Nd ¹⁴⁷ (11d)		2.7	
133	I ¹³³ (21h)		6.5	5.3	147	Sm ¹⁴⁷ (stable)	1.71		2.92
133	Xe ¹³³ (5.27d)		6.62	5.27	148	Nd ¹⁴⁸ (stable)	1.15		2.28
133	Cs ¹³³ (stable)	6.18		5.27	149	Pm ¹⁴⁹ (5.6h)		1.4	
134	Te ¹³⁴ (44m)		6.9 ^a		149	Sm ¹⁴⁹ (stable)	0.61		1.89
134	Xe ¹³⁴ (stable)	6.54		5.69	150	Nd ¹⁵⁰ (stable)	0.48		1.38
135	I ¹³⁵ (6.7h)	5.1	6.1 ^a	5.8	151	Sm ¹⁵¹ (80y)	0.27		1.17
135	Cs ¹³⁵ (2.6x10 ⁶ y)	4.9	3.1	5.53	152	Sm ¹⁵² (stable)	0.17		0.83
136	I ¹³⁶ (86s)	1.7		2.1	153	Sm ¹⁵³ (47h)	0.095		0.41
136	Xe ¹³⁶ (stable)	8.9	6.15	5.06	154	Sm ¹⁵⁴ (stable)	0.037		0.32
137	Cs ¹³⁷ (25y)	7.16	5.74	5.24	155	Sm ¹⁵⁵ (24m)		0.03	0.22
138	Cs ¹³⁸ (32m)				155	Eu ¹⁵⁵ (1.9y)			
139	Ba ¹³⁹ (84m)			5.7	156	Sm ¹⁵⁶ (10h)			
140	Ba ¹⁴⁰ (12.8d)	6.0	6.44 ^e	5.68	156	Eu ¹⁵⁶ (15.4h)			0.12

* Reference 4

a. The Cs, Nd and Sm yields are corrected to 5.6% yield for Ba¹⁴⁰.b. Most values are corrected to a Ba¹⁴⁰ yield of 1.06 x 5.32% = 5.68%.

c. The xenon isotope yields are normalized to corrected Cs yields at mass 133.

d. Average value.

e. Measured value is 6.32%. The factor 1.06 is introduced to bring the total yield for light and heavy groups to 100% each.

RADIOACTIVATION AND FALLOUT CONTAMINATION

Radioactive contamination of food and water can be the result of several processes. Direct activation of food materials and containers can take place when exposure to intense neutron and gamma fluxes has occurred. Such exposures are possible from nuclear or thermonuclear weapon detonations. Secondary forms of contamination can then occur from the fallout debris associated with nuclear weapon explosions or reactor venting.⁽¹⁰⁾ The fallout materials can be directly deposited on consumable materials, or indirect contamination can happen through animal and plant uptake of these contaminants and the subsequent human consumption of animal and plant products.

An activation analysis study has been carried out by the Quartermaster Food and Container Institute for the Armed Forces to determine the concentrations of various elements in beef, pork, ham, and chicken.⁽¹¹⁾ From the data obtained, the radiochemical characterization of neutron-induced activities has also been determined. In the period from 13 days to 125 days after irradiation, the major radionuclides found were the 14.2-day P^{32} and the 87-day S^{35} beta emitters, and 45-day Fe^{59} , 245-day Zn^{65} , and 5.2-year Co^{60} gamma emitters, in addition to the bremsstrahlen from the P^{32} . Table 3 shows the most important long-lived (half-life greater than 10 days) radioisotopes and the nuclear reactions producing them.

Table 3

Isotope	Reaction	Isotope	Reaction
P^{32}	$S^{33}(\gamma, p) P^{32}$	Fe^{55}	$Fe^{56}(\gamma, n) Fe^{55}$
P^{33}	$S^{34}(\gamma, p) P^{33}$	Zn^{65}	$Zn^{66}(\gamma, n) Zn^{65}$
P^{35}	$Cl^{37}(\gamma, pn) S^{35}$	Rb^{84}	$Rb^{85}(\gamma, n) Rb^{84}$
Mn^{54}	$Mn^{55}(\gamma, n) Mn^{54}$	I^{126}	$I^{127}(\gamma, n) I^{126}$

Table 4 shows additional activation products and additional reactions. In addition to neutron-activated products in food materials, gamma and electron-induced activities can also occur. (12,13)*

Radioactivities produced in food container materials by electron irradiation were also studied, since these activities could create a handling problem and could conceivably be leached from the containers into the food products themselves. The basic packaging materials were plastics, aluminum, and tinplate. For plastics in general the only activities observed were from C^{11} , Cl^{34m} , and F^{18} , and all of these decay to essentially background levels in a few hours. No activities from the aluminum containers were observed; however, the copper impurities showed up as Cu^{62} and Cu^{64} . Tinplate containers yielded small amounts of Mn^{56} , Fe^{53} , and Sn^{123} activities.**

While detailed description of the effects of ionizing radiation on foods is beyond the scope of this study, it must nevertheless be pointed out that other phenomena besides induced activities, which affect the wholesomeness of foods, occur because of intense neutron, gamma, and beta (electron) irradiation.⁽¹⁴⁾ These effects are mainly the radiation-induced chemical analogues of food spoilage resulting from such reactions as free radical (OH , HO_2 , and H_2O_2) production, rupture of chemical bonds, hemolysis of red blood cells in fresh beef, oxidation, color and odor changes, and enzyme inactivation.*** Since rupture of chemical bonds is possible, the resulting degradation products can have toxic properties, and these have been found in various studies of irradiated food wholesomeness. No toxicity is produced in foods which have been irradiated with up to 6×10^6 rads. No carcinogenic materials have been found

* Induced activities in some foods were studied as a function of incident electron energy.

** In all cases the activities induced were from μ curie sources and were well below the MPC values recommended by the NCRP.

*** Private communication, Dr. Rachael Makower and Dr. Lineweaver, U.S. Dept. of Agriculture Western Regional Laboratory, Albany, California.

Table 4
DATA FOR ACTIVATION PRODUCTS AND INTERFERING REACTIONS

<u>Element</u>	<u>Activation Product</u>	<u>Radiation Measured</u>	<u>Additional Reactions</u>
Phosphorus	14.2 -d P ³²	1.707 Mev β ⁻	S ³² (n,p), Cl ³⁵ (n,α)
Sulfur	87.0 -d S ³⁵	0.1617 Mev β ⁻	Cl ³⁵ (n,p)
Chlorine	3.0x10 ⁵ -y Cl ³⁶	0.714 Mev β ⁻	None
Scandium	83.9 -d Sc ⁴⁶	0.885 Mev γ	Ti ⁴⁶ (n,p)
Iron	45.1 -d Fe ⁵⁹	1.098 Mev γ	Co ⁵⁸ (n,p)
Cobalt	5.24 -y Co ⁶⁰	1.1728 Mev γ	Ni ⁶⁰ (n,p)
Zinc	245.0 -d Zn ⁶⁵	1.119 Mev γ	None
Selenium	127.0 -d Se ⁷⁵	0.402 Mev γ	None
Rubidium	18.66 -d Rb ⁸⁶	1.079 Mev γ	Sr ⁸⁶ (n,p)
Strontium	64.0 -d Sr ⁸⁵	0.513 Mev γ	None
Zirconium	65.0 -d Zr ⁹⁵	0.723 Mev γ & 0.756 Mev γ	U, Th fission
Ruthenium	39.8 -d Ru ¹⁰³	Mixed β ⁻	U, Th fission
Palladium	17.0 -d Pd ¹⁰³	Mixed γ	Cd ¹⁰⁶ (n,α)
Indium	49.0 -d In ^{114m}	0.190 Mev γ	None
Tin	119.0 -d Sn ¹¹³	0.393 Mev γ	Sb ¹²³ (n,p)
Tellurium	104.0 -d Te ^{123m}	0.158 Mev γ	None
Cesium	2.07 -y Cs ¹³⁴	0.796 Mev γ	Ba ¹³⁴ (n,p)
Barium	11.5 -d Ba ¹³¹	0.214 Mev γ	None
Cerium	32.5 -d Ce ¹⁴¹	Mixed γ	Pr ¹⁴¹ (n,p)
Hafnium	70.0 -d Hf ¹⁷⁵	0.089 Mev γ	U, Th fission None
Iridium	74.4 -d Ir ¹⁹²	Mixed β ⁻	Pt ¹⁹² (n,p)
Platinum	4.3 -d Pt ^{193m}	Mixed β ⁻	None
Uranium-238	2.346-d Np ²³⁹	Mixed β ⁻	None
Uranium-235	12.8 -d Ba ¹⁴⁰	Mixed β ⁻ (La ¹⁴⁰)	None

and the food is still considered wholesome even though color, odor, and taste changes may have occurred in varying degrees.*

Pollutants can be released by nuclear weapon detonation and by the high-temperature venting of nuclear reactors. During the Windscale event on 10 October 1957, in Great Britain, reactor pollutants, chiefly I^{131} followed by Ce^{137} , Sr^{89} , Sr^{90} , Ru^{103} , Ru^{106} , Zr^{93} , Nb^{95} , Ce^{144} and Pu^{210} were released. (14) The concentrations of major fission products were checked in food-stuffs and grassland foliage, with regular samples taken three miles from the reactor location. (15) Surveys were made and radiochemical analyses carried out for Sr^{89} , Sr^{90} , I^{131} , Cs^{137} , Ru^{103} , Ru^{106} , Zr^{95} , and total countable activity was recorded. Only the Sr^{89} , Sr^{90} , and I^{131} were found in amounts larger than the established permissible concentrations for food and water.

BIOLOGICAL SPECIMENS REQUIRING RADIOANALYSIS

The biological specimens requiring evaluation for radioisotope content include blood, urine, and fecal materials. Acceptable levels of activity for the various radionuclides in central body organs have been established by the International Commission on Radiation Protection and these standards are reprinted in the National Bureau of Standards Handbook 69. (op. cit. 2) Levels of acceptable activities in humans lie in the milli-microcurie range.

* Private communication, Major Oscar Snyder, Army Natick Laboratory, Natick, Massachusetts.

INSTRUMENTS FOR MEASURING NUCLEAR RADIATIONS

INTRODUCTION

Many systems have been devised for measuring nuclear radiations. Some of them are specific for a particular type of radiation (i.e., alpha, beta, or gamma), while others will detect all these radiations with varying degrees of efficiency. Among the more generally useful measuring instruments are ionization chambers; Geiger counters; proportional counters; gas flow counters; solid state (semiconductor) detectors; solid state scintillators such as NaI(Tl), CsI(Tl), KI, anthracene and stilbene; plastic and liquid scintillators; zinc sulfide screens; and thermoluminescent CaF_2 and LiF_2 . A short discussion of several types of detectors and their applications will be presented to facilitate the choosing of adequate types of radiation detection systems (α , β , γ) for the Mobile Environmental Radiation Evaluation Laboratory.

ALPHA PARTICLE DETECTION

The range of an alpha particle in air at standard conditions of temperature and pressure is given by

$$R = 0.309E^{3/2}$$

where E is the particle energy. Typically, under these conditions, a 6-Mev alpha particle has a range of about 0.6 cm in air (op.cit.5). This means any detection system which is separated from the source, except in vacuum, by more than a few centimeters will not be able to detect any but the most energetic alpha particles. Thus any efficient alpha detection system must either have the source virtually in contact with the detector, or the intervening space between the detector and source must be evacuated. This is especially true in the

determination of alpha particle energy. Another ramification of the short range associated with alphas is the requirement that detection must occur with virtually no dead space at the surface of the detector. Detector windows must either be non-existent or exceedingly thin, as in the case of surface barrier semi-conductor detectors or windowless gas flow counters. Determination of particle energy by pulse height analysis can be accomplished with surface-barrier silicon junction and lithium-drifted silicon semiconductor detectors, with ionization chambers, and/or with anthracene, CsI, or stilbene scintillation crystal detectors. An example of the necessary electronic circuitry for semiconductor pulse height analysis of charged particles is shown in block diagram form in Figure 11.

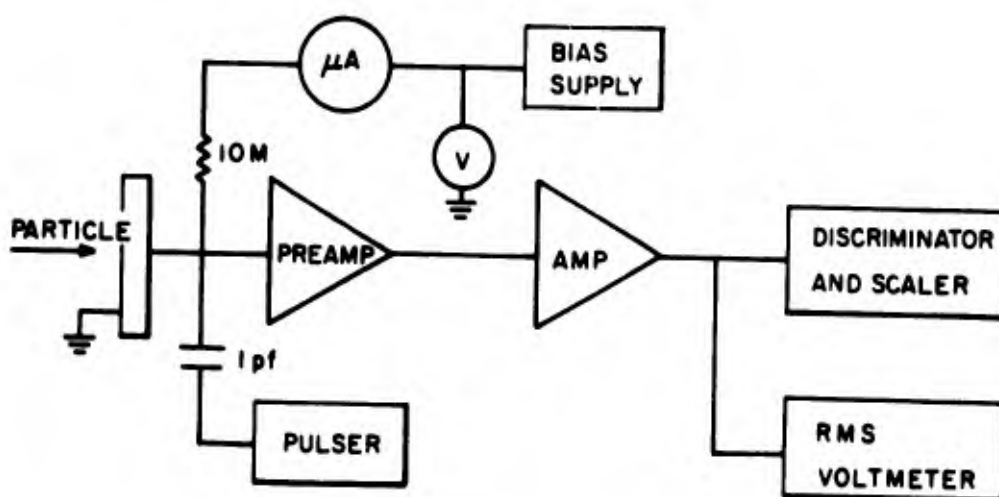
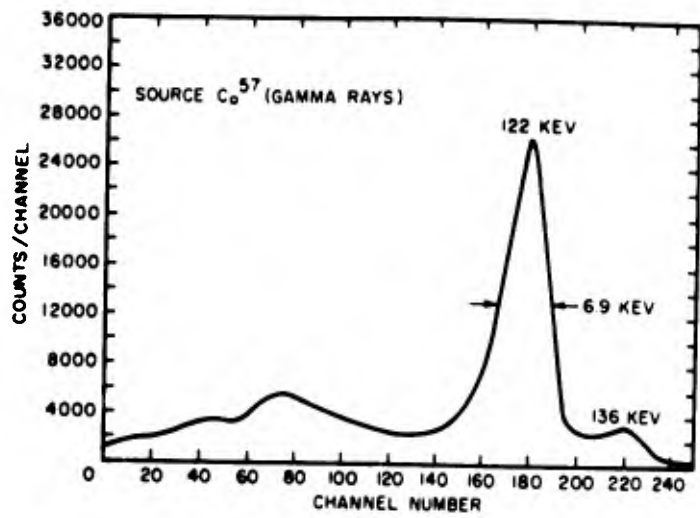
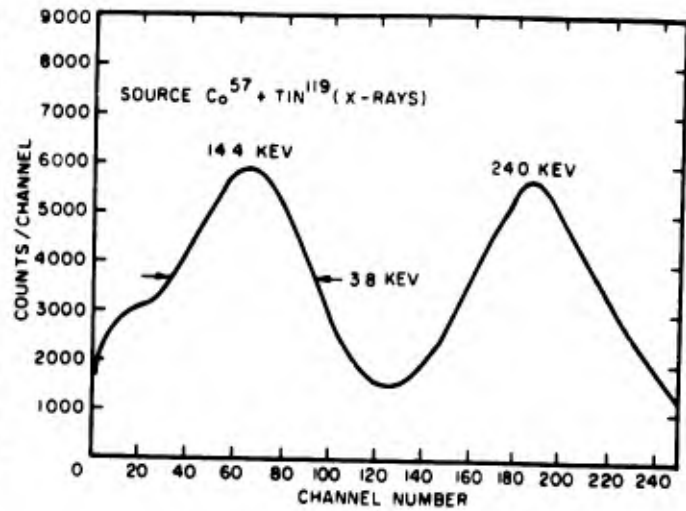


FIGURE 11. BASIC CIRCUIT FOR CHARGED PARTICLE COUNTING

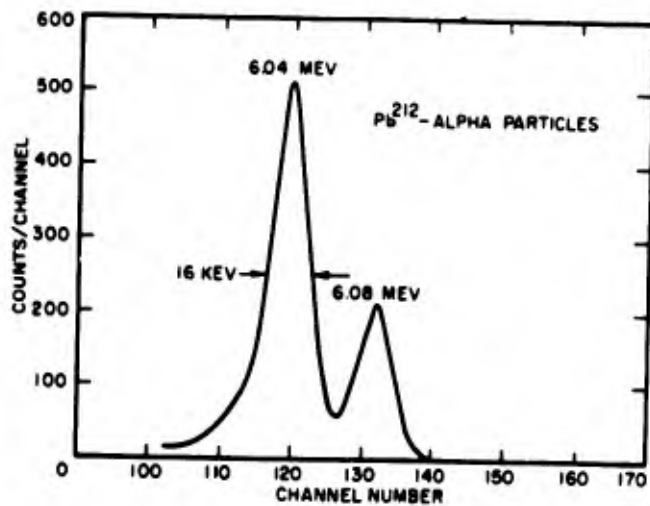
Typical alpha, beta, and gamma energy spectra obtained with lithium-drifted germanium semiconductor detectors are shown in Figures 12a, b, c, d, e, f. Figure 13 is a similar diagram for gross counting of alphas without any pulse height analysis capability. The detection efficiency for all alpha particles reaching the detector is essentially 100 per cent.



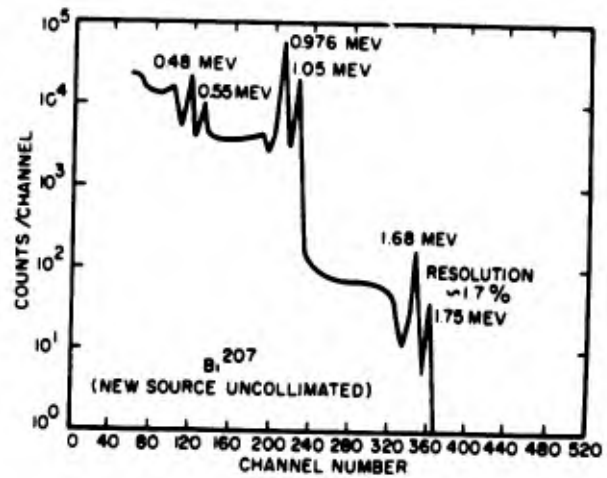
A.



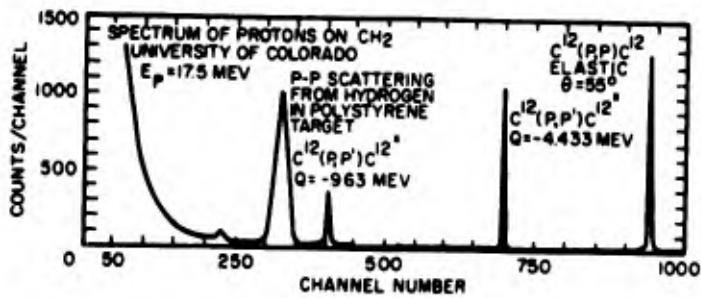
B.



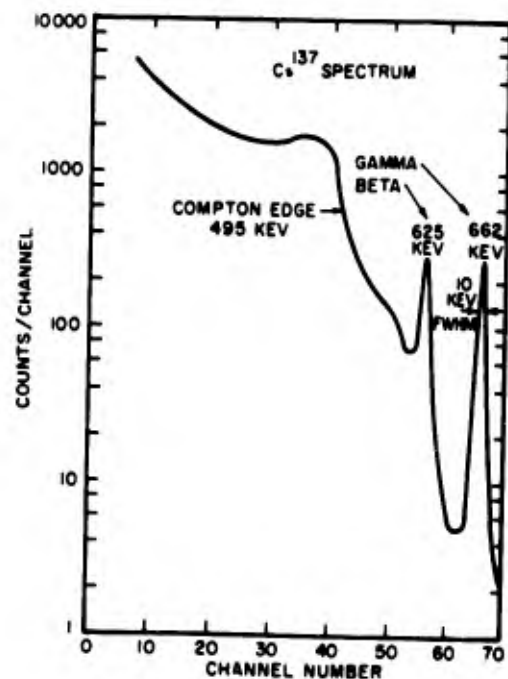
C.



D.



E.



F.

FIGURE 12. ENERGY SPECTRA OBTAINED WITH LID-GERMANIUM DETECTORS

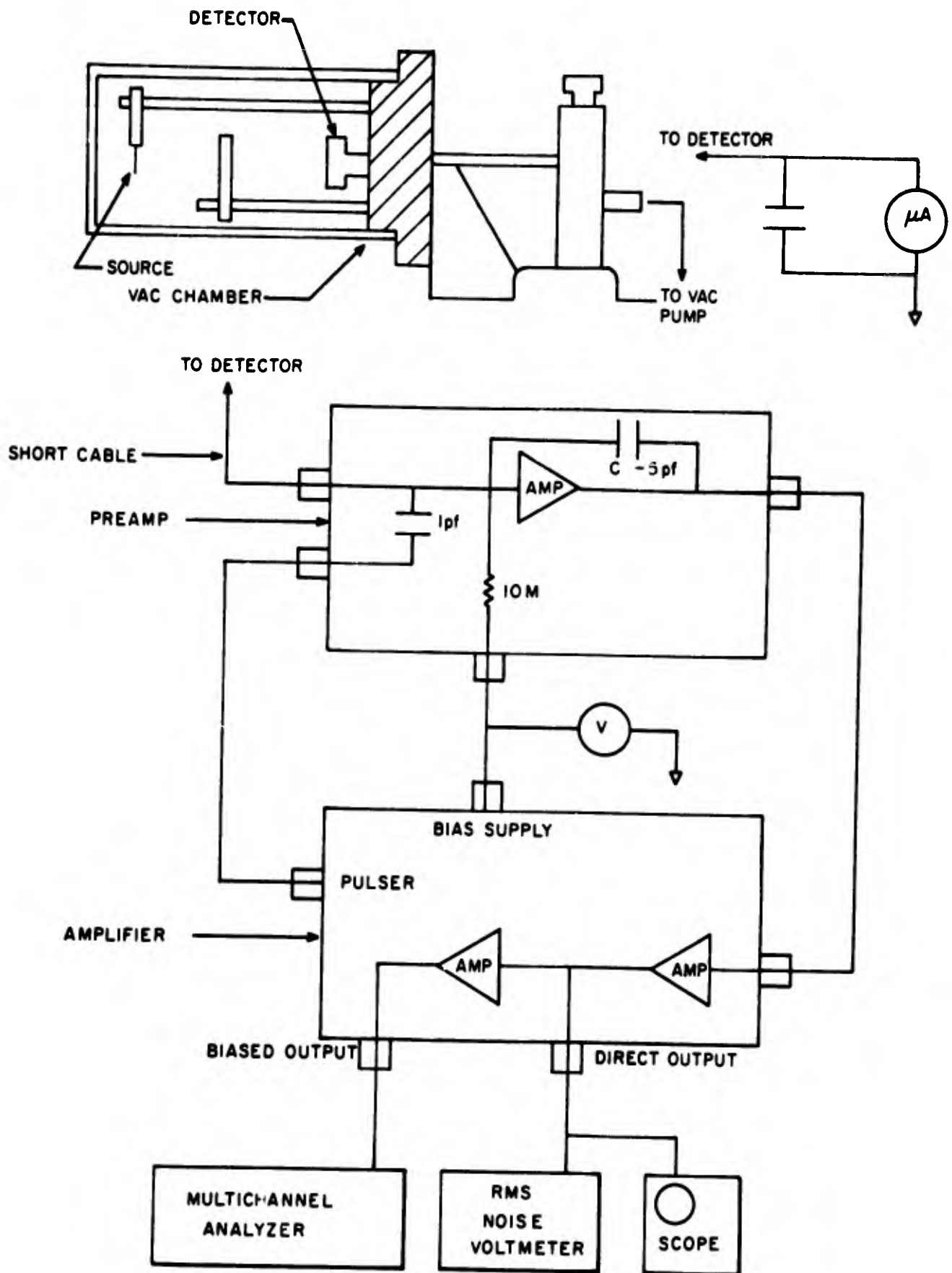


FIGURE 13. COMPLETE CIRCUIT FOR CHARGED PARTICLE SPECTOMETRY

Surface barrier gold-germanium and surface barrier silicon detectors have been used to measure the energy spectrum of fission fragments from the neutron-induced fission of U^{235} and the energy spectrum of alpha particles from uranium. (16) An evacuated system was provided for the source-detector geometry to minimize the energy loss of the particles in traveling from the source to the detector. The detectors were operated at liquid nitrogen temperature to eliminate the noise generated in the detector. Alpha measurements were made of a source containing 93 per cent U^{235} , 6 per cent U^{238} , and 1 per cent U^{234} with some trace of U^{232} and its daughter products. A 24-mm x 24-mm Au-Ge detector and a 17-mm diameter Au-Si detector were used to obtain the spectra shown in Figures 14 and 15 respectively.

An energy spectrum obtained with a gridded ionization chamber is shown in Figure 16. Ionization chamber measurements rely on the collection efficiency of the chamber for ion pairs created by the passage of an ionizing particle (α , β , γ , n, etc.) through the gas-filled volume between the collecting electrodes. For alpha particles with ranges less than the dimensions of the collecting electrodes and the electrode separation, the voltage pulse due to the ionization is proportional to the total energy of the ionizing particle; hence pulse height analysis is possible. The short range of alphas requires that the nuclide sample be thin and situated in the ionization chamber. (If the sample is located on the negative collecting electrode in a parallel plate electrode chamber, this becomes virtually a 2π counting geometry.) Self-absorption corrections must be made for source thickness for a uniformly distributed sample. The fraction of alphas lost by self-absorption has been calculated to be

$$F = \frac{1}{2} t/R.$$

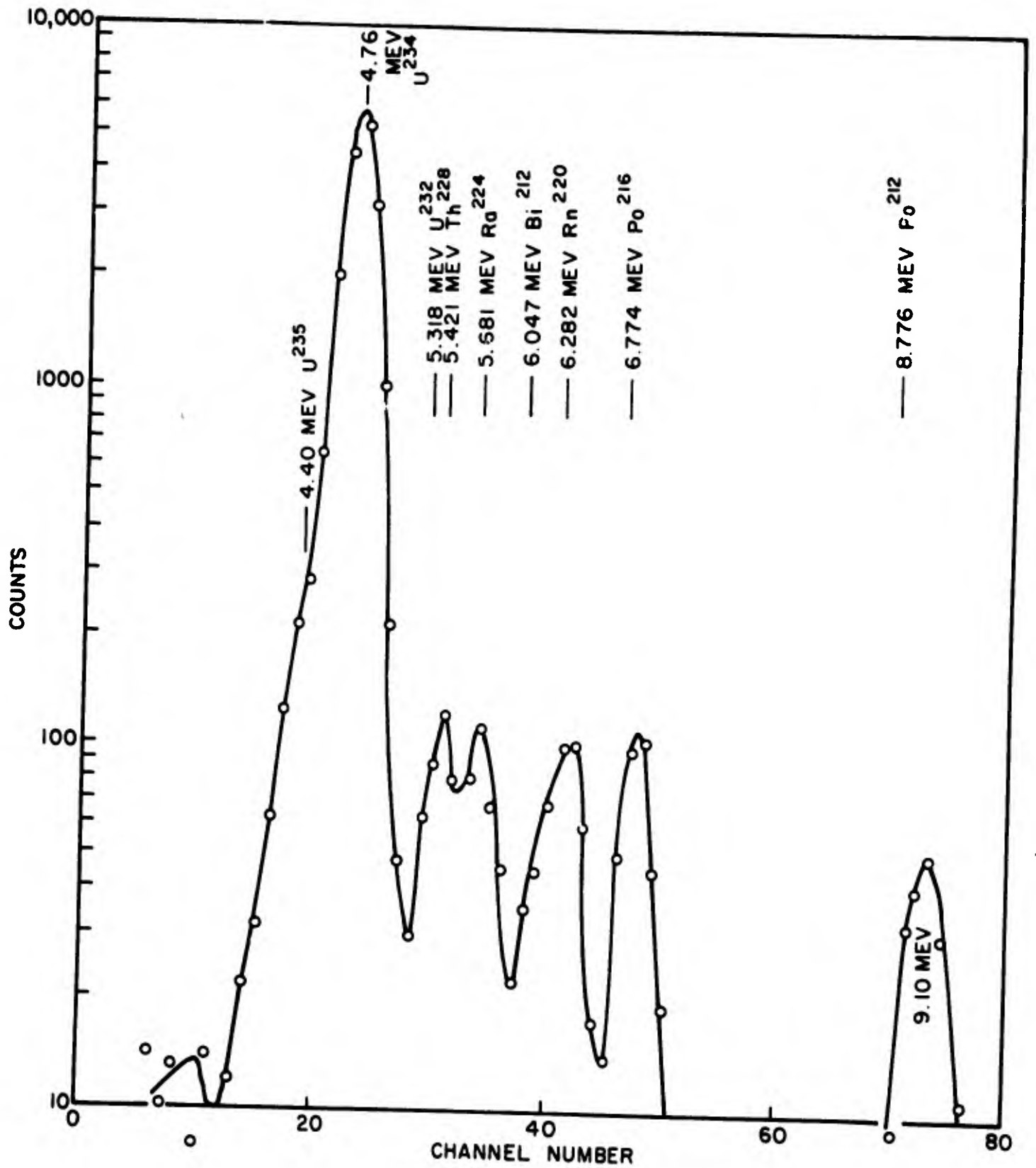


FIGURE 14. ALPHA SPECTRA OBTAINED WITH Au-Ge DETECTOR

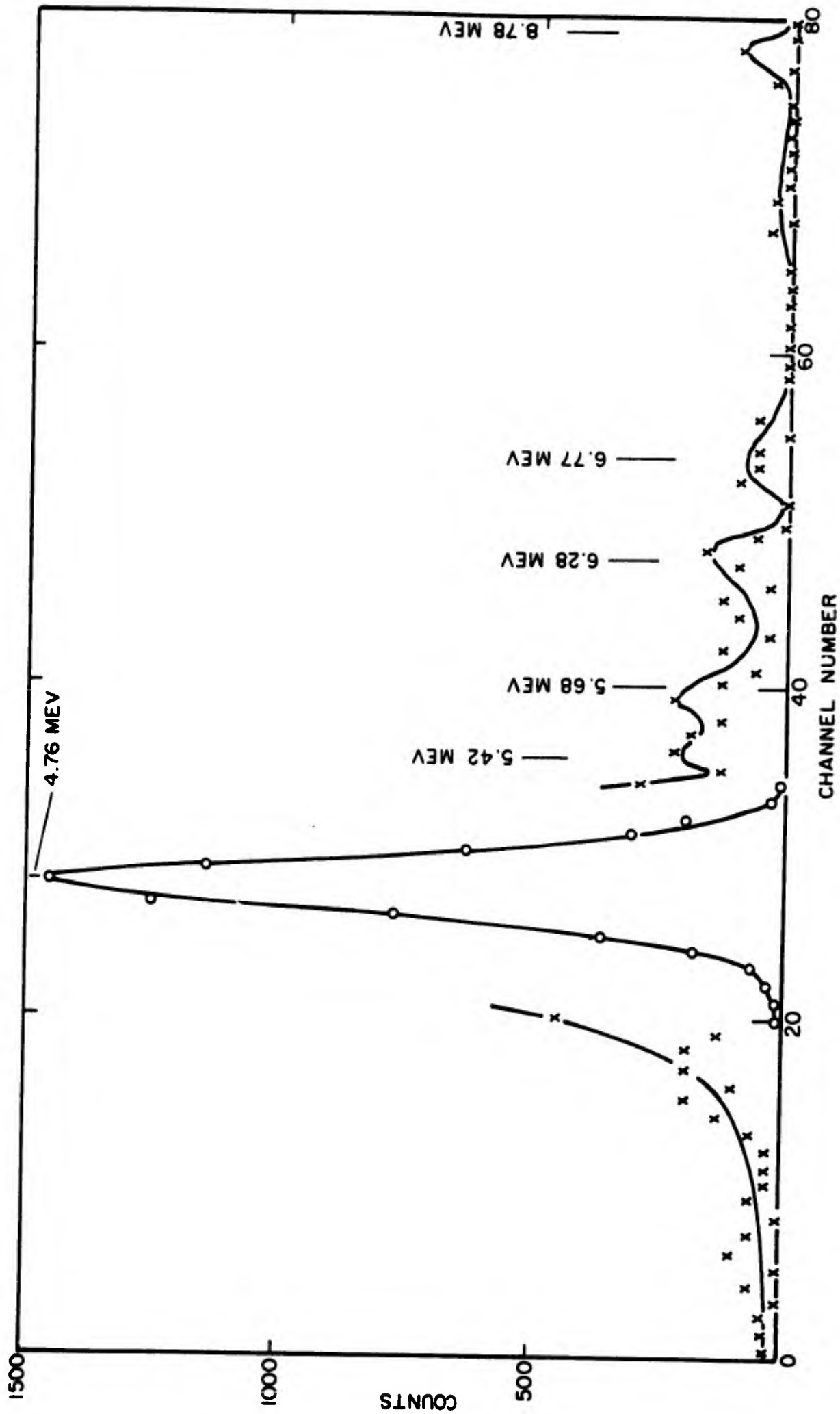


FIGURE 15. ALPHA SPECTRA OBTAINED WITH Au-Si DETECTOR

where t is the sample thickness and R is the effective range of the alpha in the sample material. (17)*

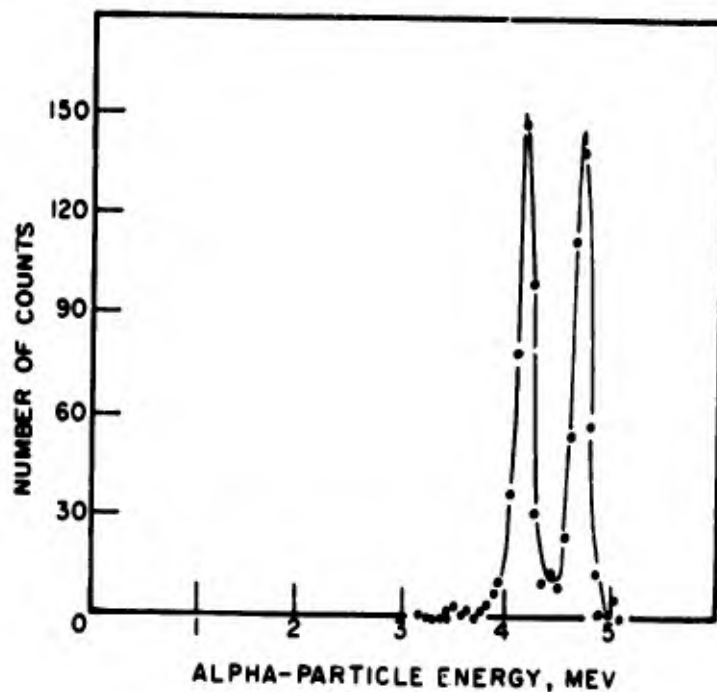


FIGURE 16. ENERGY SPECTRUM OBTAINED WITH GRIDDED IONIZATION CHAMBER

Alpha particle scattering is generally not a problem, and the scattering factor is usually about 1.0. Backscattering increases with increasing atomic number and with decreasing alpha energy. The backscattering factor for a platinum sample plate holder in a 2π geometry is about 1.04. (In low geometry detectors, i.e., less than $\pi/25$, backscattering is a negligible effect.)

Alpha spectroscopy has been accomplished with a modified Frisch Grid Ionization Chamber and a 256-channel pulse height analyzer. (18) Over-all resolution is better than one per cent. **

* Effective range refers to the distance an alpha can travel in a medium and still have sufficient energy to register a count in the detector.

** The energy measurement resolution is defined as the ratio of the full width of the measured energy distribution to energy at the peak of the distribution. The width is measured at a position which is one-half of the peak value of the distribution.

The calculated resolution of the chamber which delivers pulses having a Poisson distribution is

$$R = (8 \ln 2)^{\frac{1}{2}} \frac{I}{E}$$

where E is the energy of the alpha particle in electron volts and I is energy expended in producing an ion pair. The distribution is a statistical function of the chamber and is determined by the ionization potential of the gas. Thus for a 5.3-Mev alpha particle and argon gas, the resolution is 0.53%. A Tracerlab RLD-1 Frisch Grid chamber (having an electron collection time of 1 μ second) and preamp coupled to an RCL A-61 linear amplifier and an Argonne type 256-channel pulse height analyzer constitutes such a measuring system. Typical spectra are shown in Figures 17 and 18. Resolutions of 0.81%, and 1.05% were obtained for (Am^{241} , Pu^{234} , Th^{239}) and (Pu^{234} , U^{234} , U^{235} , U^{238}) respectively as groups.

The properties of several scintillators are shown in Table 5. For alpha particle counting, the most useful scintillator is silver-activated zinc sulfide, since it has a very high energy-to-light conversion efficiency. Because it is a fine powder, however, it becomes difficult to obtain good energy resolutions and its low transparency means that only very thin layers may be used. Frequently the powder is dusted on a Mylar film backing for support, or it may be dusted directly on the photomultiplier tube's photocathode surface.* Large- and small-area alpha probes for survey instruments have been developed using these techniques. These instruments are notably gamma insensitive in fields up to 4.5 roentgens per hour. The probes, which are

*Mylar-backed silver-activated zinc sulfide is available from William B. Johnson and Associates, Mountain Lakes, New Jersey, and Simon Adhesive Products Corp., Division of Litton Industries, 35-02 48th Avenue, Long Island City, New York.

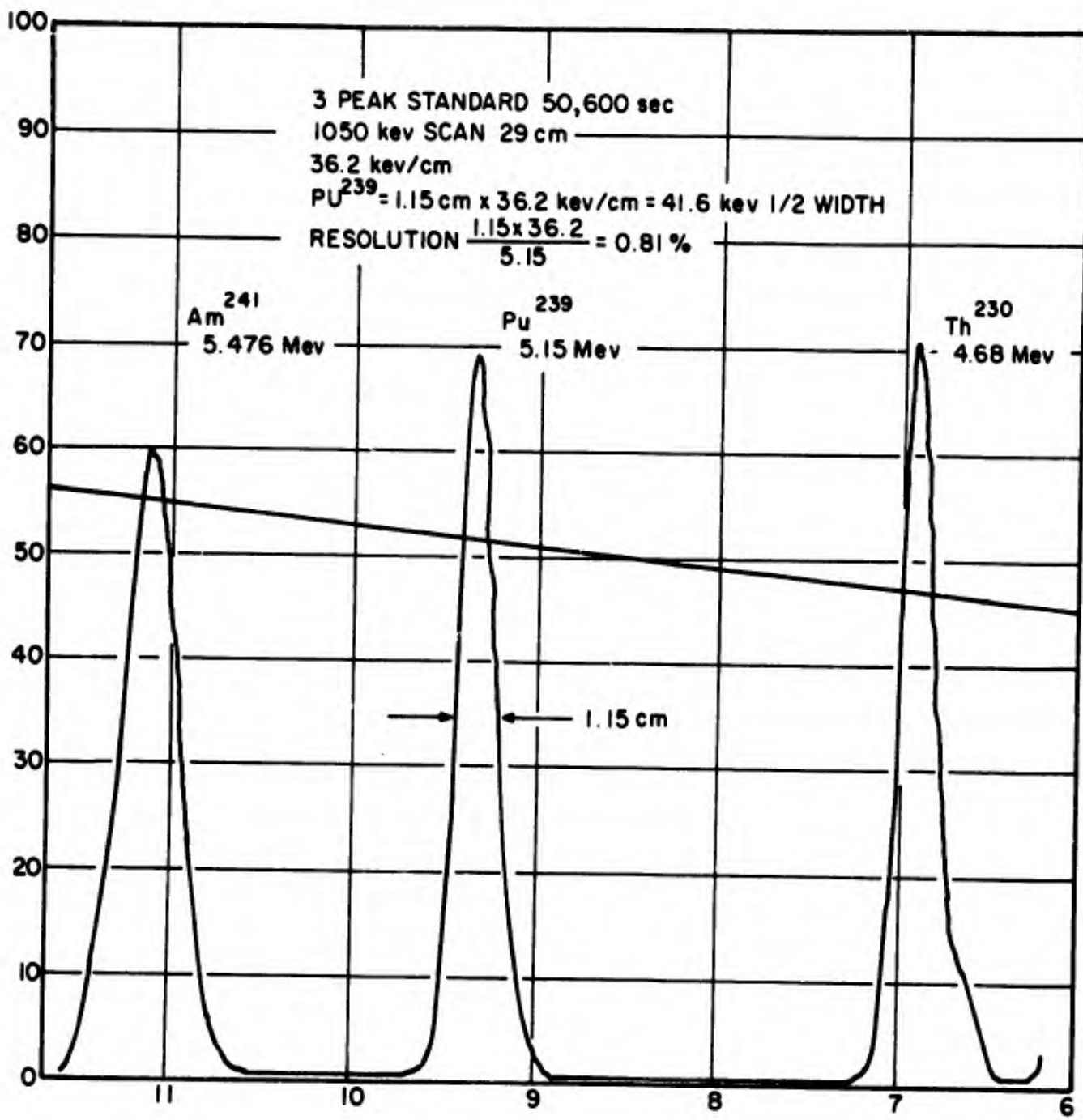


FIGURE 17. PLOT OF 50,600-SECOND COUNT OF Th²³⁰, Pu²³⁹, AND Am²⁴¹, SHOWING RESOLUTION OF 0.81%

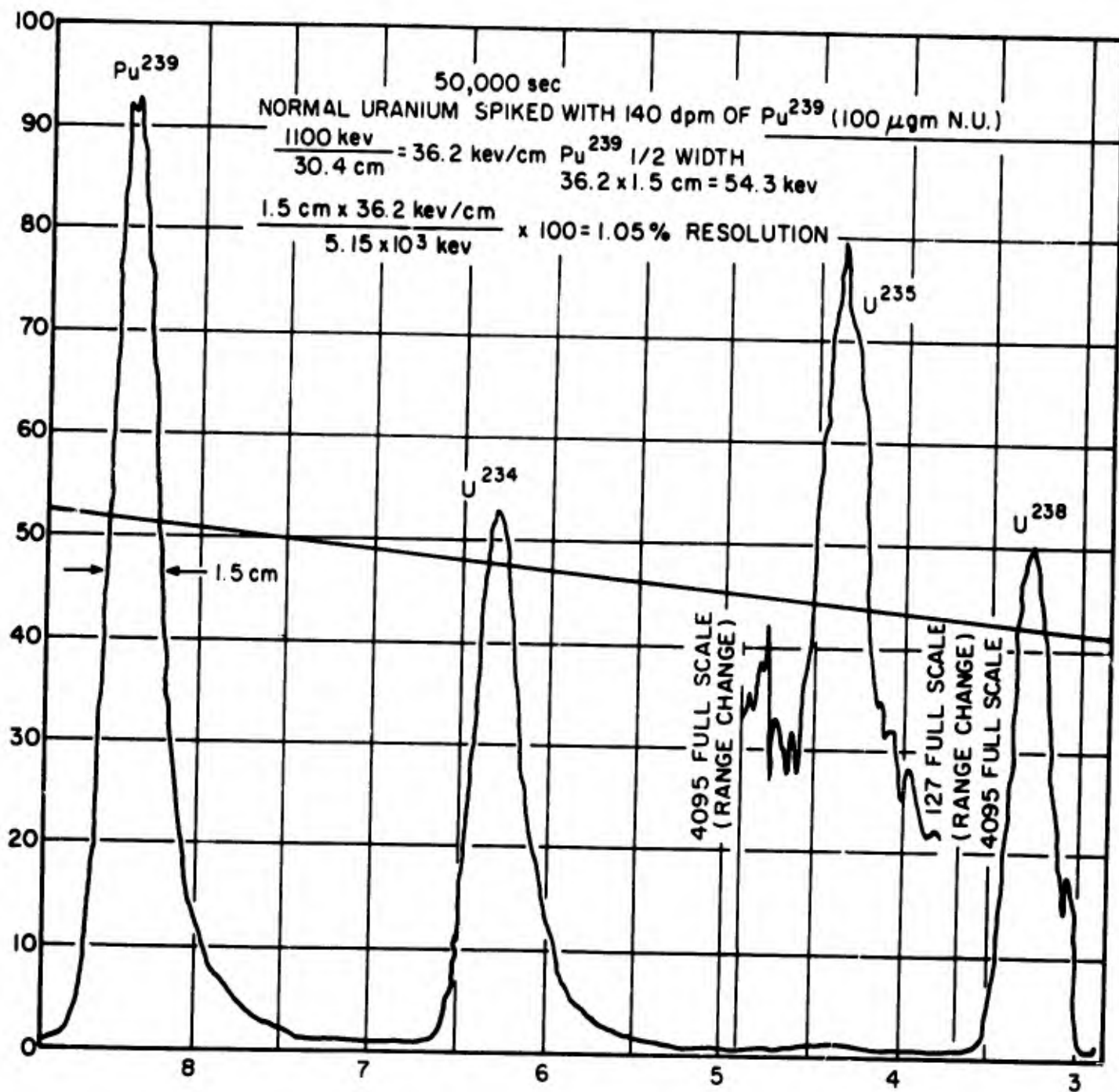


FIGURE 18. ALPHA SPECTRUM OF 100 μgm OF URANIUM AND dpm OF Pu^{239} SHOWING RESOLUTION OF 1.05%

TABLE 5. Properties of Several Scintillators*

Scintillator	Density, g/cm ³	Wavelength of maximum, emission, Å	Relative beta-ray pulse-height	Alpha- beta ratio, %	Decay time sec
1. Anthracene crystal	1.25	4,400	100	9	$2.7 \times 10^{-8} \dagger$
2. Trans-stilbene crystal	1.15	4,100	60	9	3 to $7 \times 10^{-9} \dagger$
3. Xylene + 5 g/liter of terphenyl + 0.01 g/liter of diphenylhexatriene	~ 4,500	48	9	
4. 100 g polyvinyltoluene + 4 g terphenyl + 0.1 g diphenyl stilbene	...	~ 3,800	48	9	
5. Terphenyl in polystyrene	1.1	3,900-4,300	15	...	5×10^{-9}
6. NaI(Tl)	3.67	4,100	210	44	$2.5 \times 10^{-7} \dagger$
7. LiI(Sn)	5,300	12	~93	
8. LiI(Eu)	~ 4,400	75	~95	
9. ZnS(Ag)	4.10	4,500	200	100	$10^{-5} \dagger$

* Primarily from R.K. Swank, Nucleonics, 12:14 (March, 1954).

† From J. B. Birks, "Scintillation Counters," Chap. 6, McGraw-Hill Book Company, Inc., New York, 1953.

as large as 2 x 7 inches, include a single photomultiplier such as the RCA 6655A, a zinc sulfide screen, and a light piper. In this case, the silver-activated zinc sulfide screen (10 to 15 mg/cm²) is bonded to a 1/16-inch thick lucite plaque. Instruments of this type have been used at Hanford for routine Pu²³⁹ contamination monitoring. (19,20,21,22)

Another alpha energy analysis system utilizes a thin CsI crystal as the detector. (23) The system was developed primarily to facilitate the detection of low levels of airborne plutonium in the presence of radon (Rn²²²) and thoron (Rn²²⁰). The instrument described is capable of resolving the presence of 1.2 disintegrations of plutonium per minute per cubic foot of air in a background of 12 disintegrations per minute per cubic foot of radon or thoron in less than four hours.* The system is based on the concept that since Pu²³⁹ emits alphas of 5.15, 5.13, and 5.10 Mev, and the daughter products of radon and thoron emit alphas ranging in energy between 5.3 and 8.95 Mev, an adequate alpha energy analyzer would be able to count only plutonium alphas. However, in a practical system there is an air gap between the source (filter paper collector with air drawn through it) and the detector; hence almost a continuum of energies is exhibited in the energy spectrum. Figure 19 shows one such typical spectrum for plutonium, and Figure 20 the energy spectrum for the radon-thoron background. The pulse heights from the plutonium show little contribution due to background; hence, a single channel analyzer with proper input discrimination could be adjusted to determine the plutonium concentration. The relatively few counts contributed by the

*This is an equivalent sensitivity of 40 MPC-hour where
1 MPC = 2×10^{-12} c/cm³.

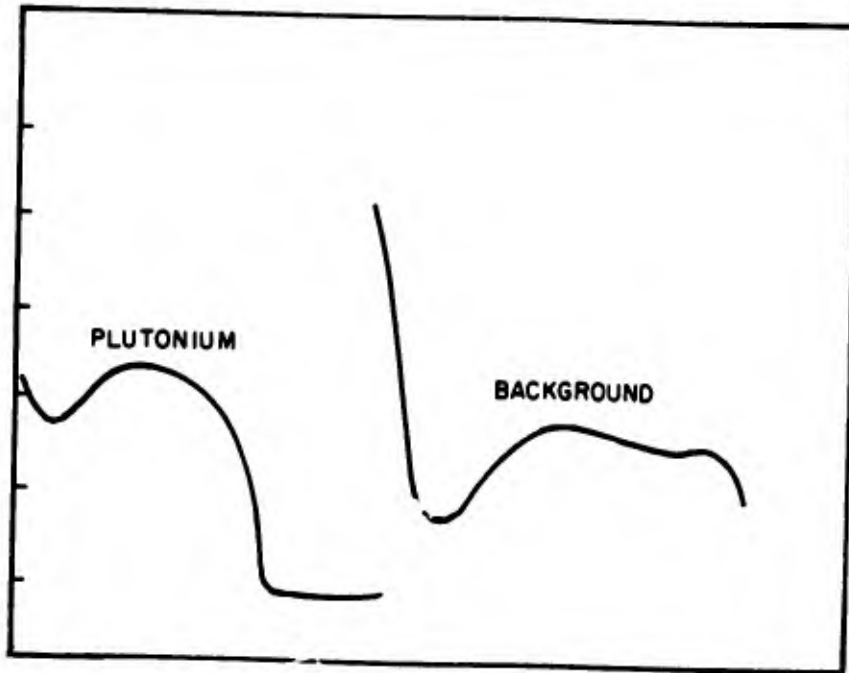


FIGURE 19. PLUTONIUM AND BACKGROUND SPECTRA

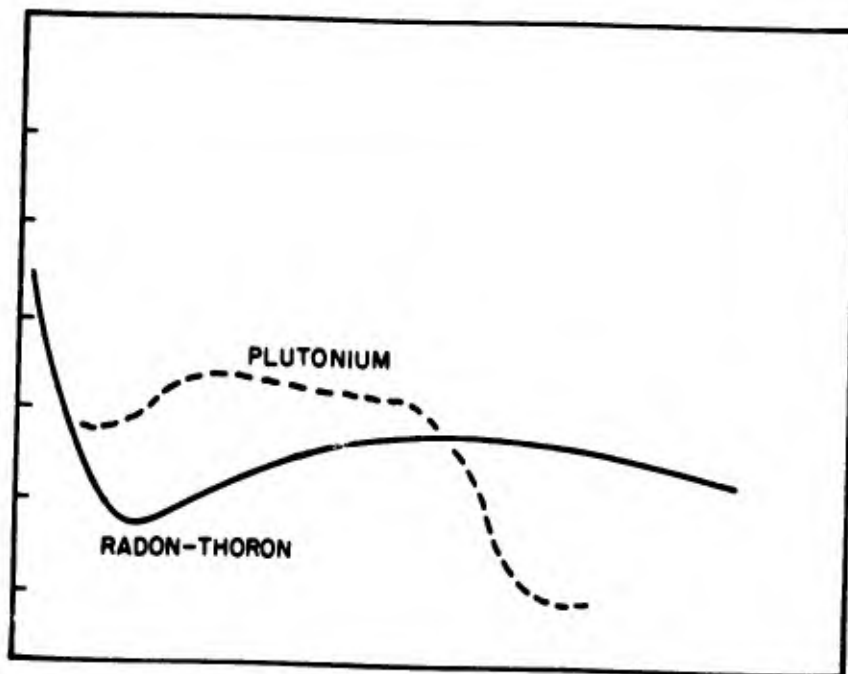


FIGURE 20. PLUTONIUM AND RADON-THORON SPECTRA

background can be compensated for by counting those pulses with heights above the plutonium cutoff and using them to balance out the contribution in the plutonium channel.*

Figure 21 is a schematic representation of the detection and counting circuitry. The detector is a 0.003-inch thick CsI(Tl) crystal coupled to a photomultiplier tube. Standard commercial pulse amplifiers, single channel analyzers, and count rate meters are used.

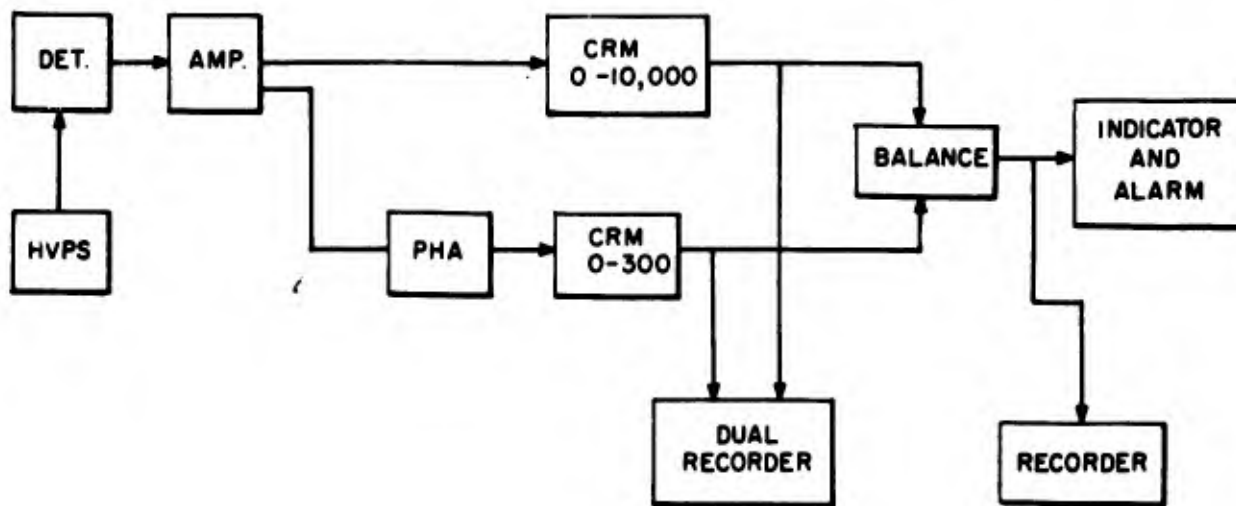


FIGURE 21. DETECTION AND COUNTING CIRCUITRY

Windowless gas flow counters use the principle of the proportional counter and are widely used for alpha counting. The counter is operated in the voltage region where gas multiplication is present and yet the proportionality to particle energy is still maintained. The gas (such as argon, methane, or helium) flows continuously through the chamber

*The authors of Reference 31 also indicate that semiconductor detectors could also be used in this application. They would give better pulse height resolution, however, this would require the use of large surface area detectors or arrays of smaller surface area detectors to provide a large area.

where the sample to be counted is located. Hence, the term "windowless". Plateaus in the graph of counting-rate versus tube-voltage are located in the same manner as for the Geiger tube. (24) The starting voltage for the alpha plateau represents the condition for which the most energetic alpha particles dissipate their entire energy in the counter, and the gas multiplication is just sufficient to allow the pulses to pass the discriminator. A typical counting-rate versus counter-voltage curve for a flow-type proportional counter and a Ra-D-E-F alpha-beta source is shown in Figure 22. With this type of counter it is possible to do alpha counting in the presence of large fluxes of betas and gammas because their specific ionization is low compared to that of the alpha particles.* This also means that very low levels of alpha activity can be counted, and this is very important for health physics applications.

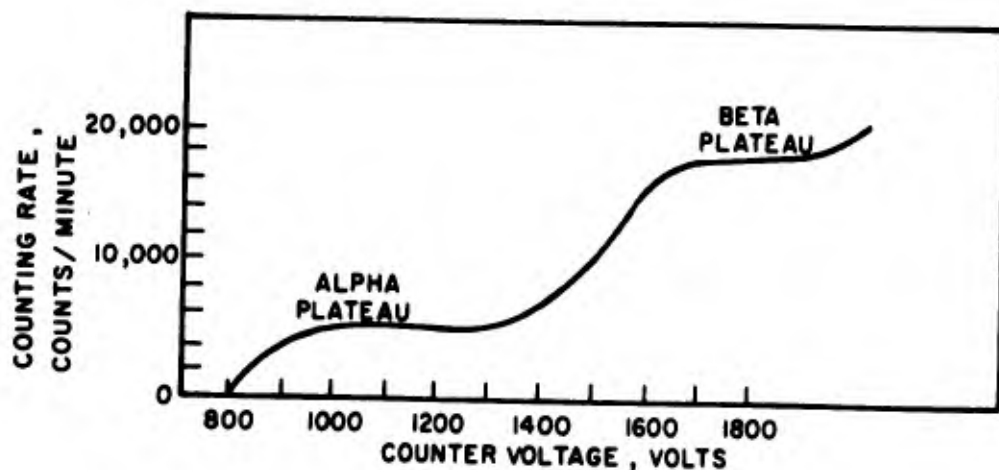


FIGURE 22. COUNTING RATE vs COUNTER VOLTAGE FOR A FLOW-TYPE PROPORTIONAL COUNTER WITH A NATIONAL BUREAU OF STANDARDS Ra-D-E-F SOURCE

*The limitation is reached, however, when the background of betas and gammas becomes so high that the superposition of their pulses results in signals of sufficient amplitude and frequency to interfere with the counting of alpha particles.

A "piggy back" system of two thin radiation detectors superimposed on each other and both counting a common sample was developed at Argonne National Laboratory. (25) The detector nearest the sample has a thin Mylar window which passes both alpha- and beta-gamma-radiations, and is operated as an alpha proportional counter with the output biased to count only alphas. This detector is mechanically separated from the second detector by an aluminum plate which is sufficiently thick to stop alpha particles, but thin enough to pass energetic betas and essentially all gammas. The second detector is operated on the upper end of the limited proportional region. The signal from each detector is amplified and recorded separately.

A zinc sulfide (silver-activated) alpha detector has been used in the field to measure low levels of alpha contamination on filter paper swipes. (26) An over-all counting efficiency of 70% of 2π with a U_3O_8 standard was obtained. The unit is not sensitive to gamma radiation up to 10 r/hr and since the swipe is enclosed in the counting chamber, it is not sensitive to external beta sources. The counter is portable and the batteries have an operating life of about 80 hours. The sample chamber consists of a circular recess in a slide located beneath the scintillation probe and as close to it as possible.

A portable counter weighing 20 pounds and utilizing a hemispherical 2π proportional flow counter using propane gas was developed to count smear samples for health physics applications. (27) The sample tray slides into position at the bottom of the hemisphere and is sealed to the counter. A plateau for alphas extends from 1400 to 1630 volts, while the beta plateau is in the range beyond 2300 to 3800 volts. The alpha background count is about 0.15 cts/min, and the beta background is about 60 cts/min. A propane gas flow of 3 ml/min is the operational level. Power requirements are 45 watts at 110 volts ac.

A portable instrument for gross alpha, beta, and gamma detection, and/or for single channel gamma energy analysis with either count-rate-meter or scaler presentation of data has been developed for radioecology studies at the Hanford Atomic Products Operation. (28) All solid state circuitry is used, and the instrument is line-operated, but is also capable of 40 hours of continuous operation from a 20V, 6A-hr-nickel cadmium storage battery.* The alpha probes are zinc sulfide screens coupled by an air light pipe to RCA phototubes. The typical alpha counting efficiency is 25 per cent. In general, a 5-Mev alpha particle will produce about 1/10 as much light as a 5-Mev electron in all organic single crystals, plastic phosphors, and solution phosphors. In the inorganic phosphors, an alpha particle will give from less than half to perhaps 70% as much light as the electron, depending on the phosphor. (29)

An alpha detection system was developed by E. C. Wingfield to monitor waste streams. (30) The detector was a silver-activated ZnS screen, mounted on a plastic disk. The fluids to be monitored - mainly water - were pumped through a stainless steel cell into direct contact with the zinc sulfide screen. The lower level of detectability was 3.3×10^{-10} curies/ml with a cell flow rate of about one gallon per minute and an inlet pressure of five pounds per square inch.

BETA MEASUREMENTS

The resolution in beta-ray scintillation spectroscopy is defined as the full width of the pulse distribution at half maximum counting rate for monoenergetic electrons (op.cit.29).

*The gamma scintillation detectors are Harsnaw integral assembly 2 x 2 inch NaI(Tl) crystals. The beta probes are Ne102 terphenyl in polyvinyltoluene scintillators.

The resolution is inversely proportional to the square root of the number of photoelectrons to a first approximation. Hence, the choice of a phosphor which has the highest light output per Mev of energy lost is important. Sodium iodide produces the largest pulse height per Mev of the inorganic scintillators, and anthracene produces the largest pulse height for the organic scintillators. The problem of high energy electron scattering is quite serious for NaI(Tl). As a rule the scattering increases with atomic number; thus some 80% to 90% of the entering electrons will scatter out before stopping. It is therefore difficult to obtain a good beta spectrum in NaI(Tl) with an external source. In anthracene, however, only 8% of the entering betas will scatter out of the crystal, and some of this scattering may be reduced by collimation so that the betas enter the detector at normal incidence. (The beta sources for spectrometry purposes must be thin to prevent backscattering effects and self-absorption.)

Organic materials are generally used for detecting beta particles because their fast response and availability in both liquid and solid solutions contributes to their utility (op.cit. 5). In addition they have lower backscattering cross sections than the inorganic scintillators. Organic liquid scintillators are useful for counting weak betas, since many of the beta emitters can be mixed directly into the scintillator solution. Organic scintillator response to monoenergetic electrons from Ba^{137m} is shown in Figure 23.

Anthracene crystals are commonly used as scintillation detectors in beta ray spectrometry. However, below 100 Kev anthracene is non-linear in its light output versus energy deposited in the crystal. Typically, the scintillator has a thickness equal to the range of the most energetic beta particle expected. Distortion of the energy spectrum occurs if the beta is scattered out of the crystal having deposited

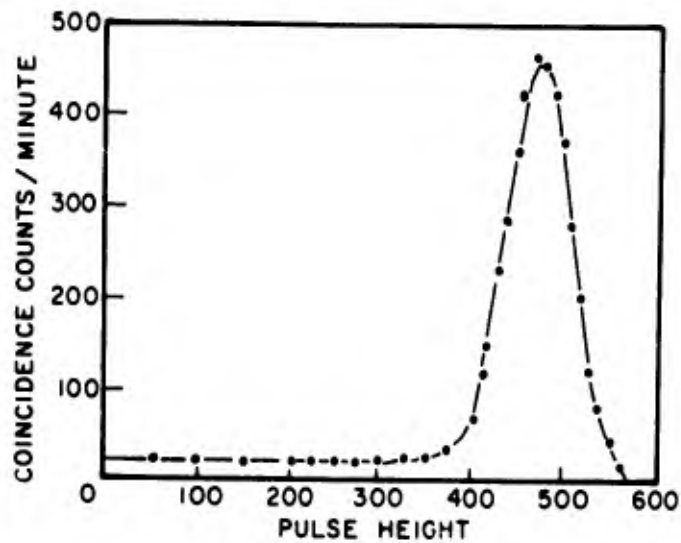


FIGURE 23. SCINTILLATOR RESPONSE TO MONOENERGETIC ELECTRONS (Bo^{137m})

only a fraction of its energy in the crystal. The general configuration of a beta spectrometer system is shown in Figure 24.

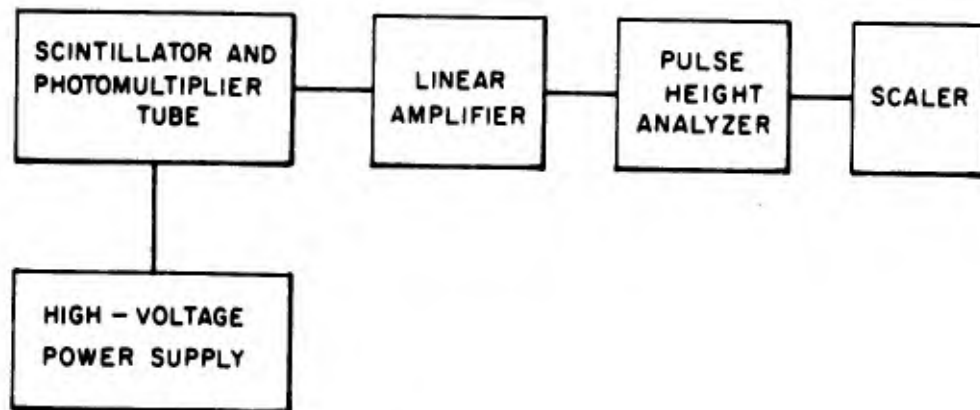


FIGURE 24. SCINTILLATION SPECTROMETER DIAGRAM

Thallium-activated cesium iodide, $CsI(Tl)$, is not deliquescent, in contrast to $NaI(Tl)$, and is thus easier to use in thin wafer form in beta spectrometers. The light output versus energy deposited in the crystal is more nearly linear at low energies than in the case of $NaI(Tl)$.

Beta scintillation counting can be very effectively done with plastic phosphors which are thick enough to stop the beta particle, but too thin to stop all but the lowest energy gamma and X-rays. Typically these detectors are of the order of 0.005 to 0.125 inches thick. Figure 25 shows one such counting system using a mercury-filled shield to reduce the background. (31)

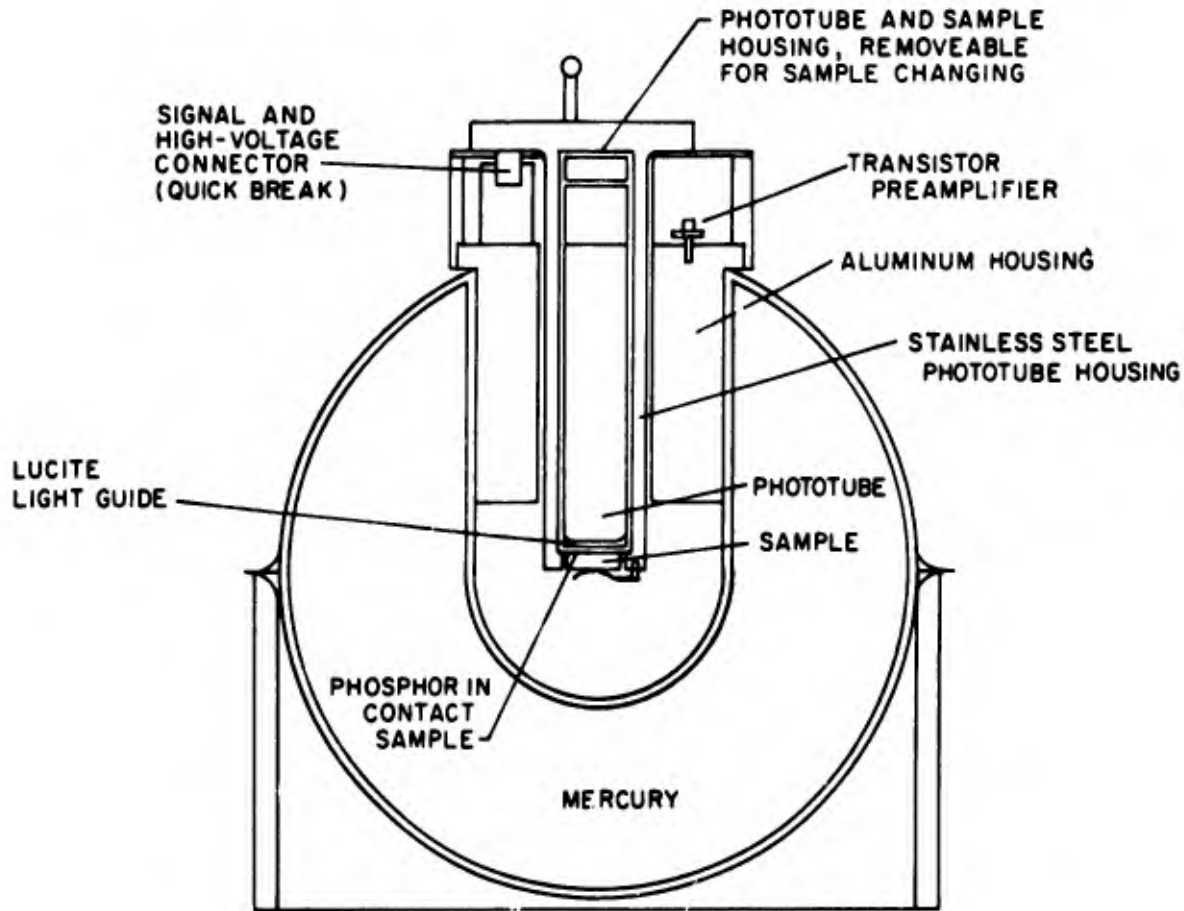


FIGURE 25. LOW BACKGROUND BETA-COUNTER

Table 6 gives the maximum energy for total absorption of betas incident normally to various thicknesses of phosphor.

TABLE 6

THICKNESS (inches)	AREA DENSITY (mg/cm ²)	MAXIMUM ENERGY FOR TOTAL ABSORPTION (Mev)
0.005	13.5	0.10
0.01	28	0.16
0.02	49	0.22
0.04	100	0.34
0.06	150	0.46
0.10	252	0.68
0.125	325	0.82

The use of a plastic light pipe is also an effective method of reducing the background due to the potassium and radium isotopes in the photomultiplier tube's glass envelope. For a 1-inch diameter by 0.010-inch thick phosphor, the background counting rate had a mean value of 0.46 counts per minute. The efficiency reported for the counter is shown in Table 7.

TABLE 7

ISOTOPE	PERCENT OF 2 π EFFICIENCY	BETA ENERGY (MAXIMUM) (Mev)
Ca ⁴⁵	12.3%	0.25
W ¹⁸⁵	13.5%	0.43
Tl ²⁰⁴	19.5%	0.76
Y ⁹¹	37.7%	1.54

Another method of reducing background counting rate and the weight of the shielding materials in both beta and gamma counting is to use anticoincidence shields around the detectors. See Figure 26.

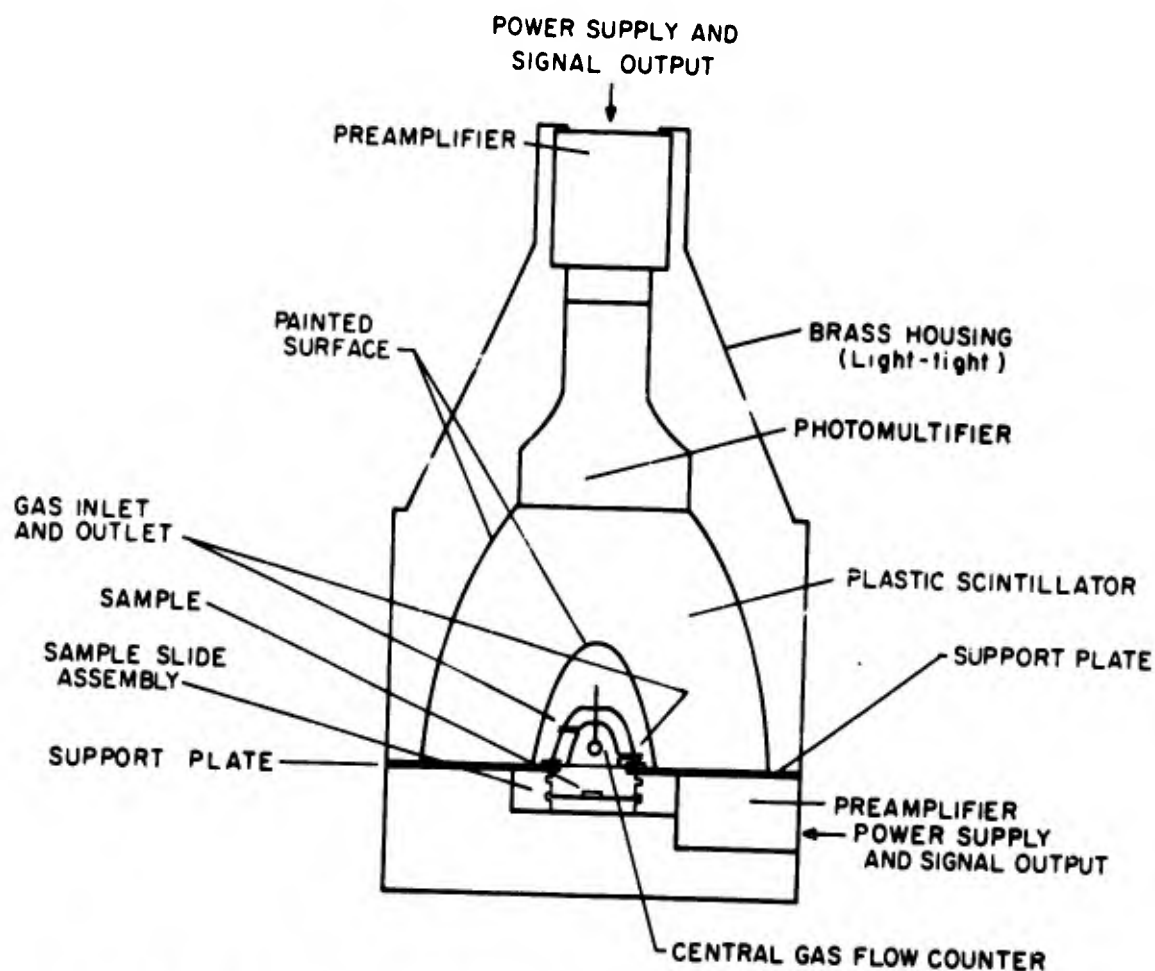


FIGURE 26. A SCINTILLATOR-SHIELDED LOW BACKGROUND BETA-COUNTER

Here the beta detector is an end window gas flow counter* (operated as a proportional counter) and surrounded by a plastic anticoincidence shield.** (32) The anticoincidence phosphor is optically coupled to a 5-inch Dumont 6364 photomultiplier. Events registering in time coincidence by the proportional counter and the plastic anticoincidence shield are

* Tracerlab Model 6E14P.

** NE102 produced by Nuclear Enterprises.

normally not counted and recorded. The output of each detector, however, is individually monitored. The system's performance can be checked by going from the anticoincidence to the coincidence mode and checking that the sum of the events registering in coincidence and anticoincidence modes equals the number of events registered by the proportional counter alone. The overall efficiency of this system is 40% for a flat 2-cm diameter Y^{90} source at a distance of 2 mm from the window. The background (measured when the entire system is enclosed by a 10-cm thick iron brick shield) is 1800 ± 100 counts/min and 9.5 ± 0.3 counts/min for the scintillation and proportional counters respectively. The coincidence background (chance or accidental coincidence) is 8.7 ± 0.3 counts/min and the anticoincidence background is 0.80 ± 0.05 counts/min. A block diagram of the measuring system is shown in Figure 27.

The pulse height response due to various isotopes placed in a well (of equal wall and floor thickness) in a plastic scintillator has been measured.⁽³³⁾ One-cubic-centimeter samples were placed in thin-walled (0.010 inches) plastic test tubes in the well. The light output was monitored with an RCA 6199 photomultiplier. The high energy betas yielded a line-shaped energy spectrum which approached the true beta spectrum as the thickness of the detector was increased. This can be inferred from the Bethe-Block formula which shows that the energy loss of a beta traversing an absorber goes as $1/V^2$ where V is the particle velocity. For very thin scintillators, it is the rate of energy loss dE/dt rather than total energy loss that is measured. The effect of increasing the sample volume was also investigated, and it was noted that a lowered detection efficiency resulted. At small volumes the sensitivity rapidly decreased due to self-absorption. A slower decrease was noted at larger volumes because of the inefficient light

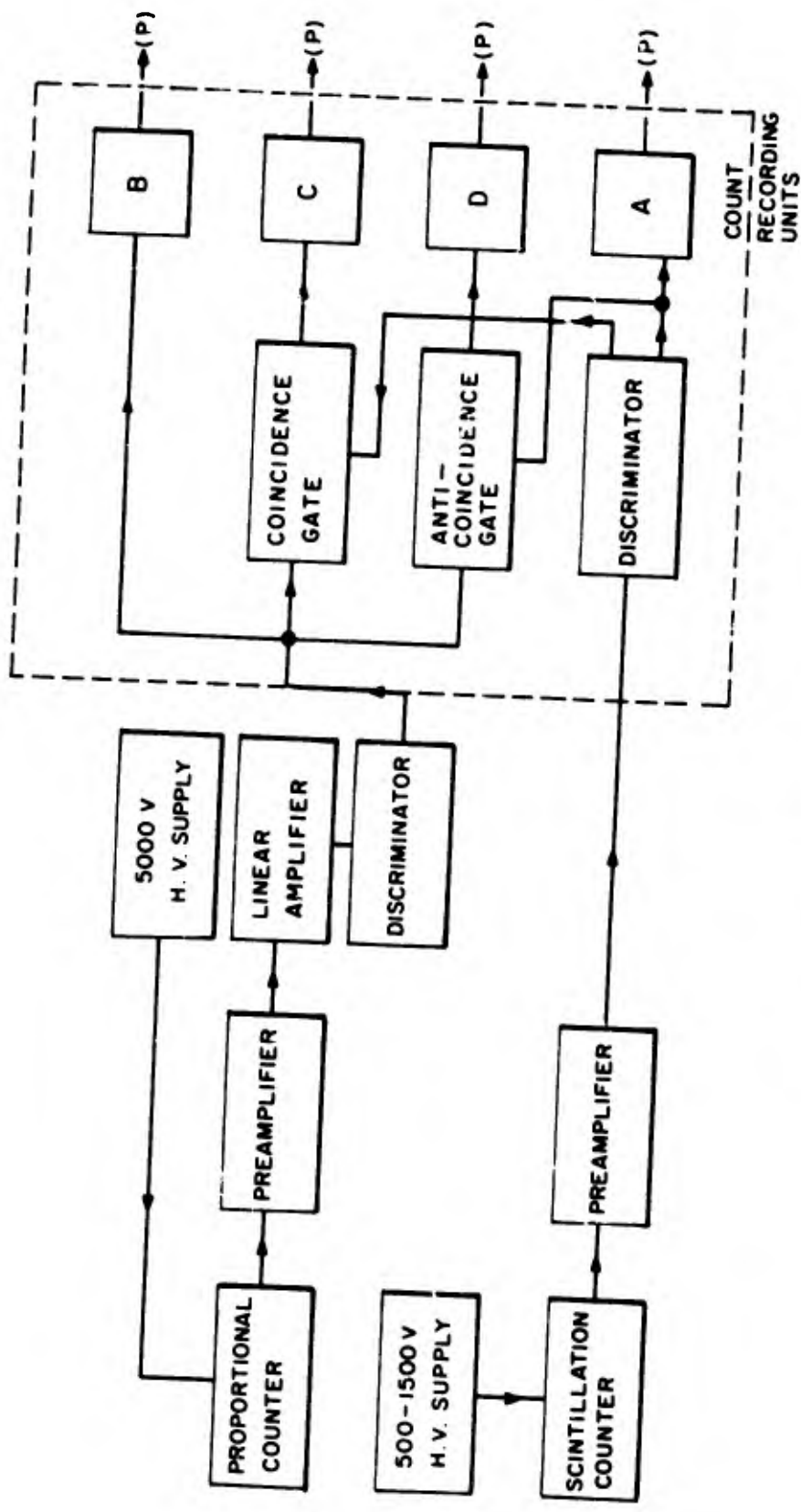


FIGURE 27. DIAGRAM OF COUNTER AND ASSOCIATED ELECTRONIC CIRCUITS

collection from the top of the scintillator (Figure 28). In general, the specific gravity of sample solutions must be the same if a uniform beta detection efficiency is to be maintained. The plastic well counter is quite suitable for directly measuring beta activity in aqueous and body fluid samples.

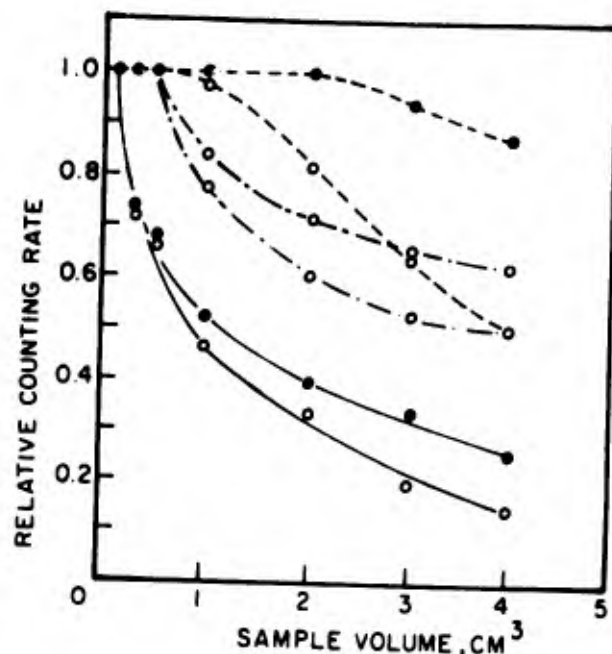


FIGURE 28. INCREASED SAMPLE VOLUME LEADS TO LOWER DETECTION EFFICIENCY. WATER WAS ADDED TO SAMPLE IN WELL OF 1-INCH DIAMETER, 1.5-INCH DEPTH, AND 1/8-INCH WALL.

The Health and Safety Laboratory of the USAEC has developed a low level beta scintillation counter having a counting efficiency of 30% for the 1.32 Mev betas of K^{40} . Each sample to be counted was located within a 15/16-inch diameter on a flat plaque of plastic scintillator 1-1/32-inch in diameter. (34) The sample is held in place by a thin covering of Mylar film and a retaining ring. The scintillation phosphor light output (0.010 inches of Pilot B phosphor) is viewed by a photomultiplier.* With the phosphor in direct

*The plastic phosphor can be replaced by a silver-activated zinc sulfide screen (Ag-ZnS dusted on Mylar film) for alpha particle detection. A counting efficiency for this source-sample geometry of 50% of 2 can be realized.

contact with the sample to be counted, no optical coupling is necessary between the phosphor and the photomultiplier. No anticoincidence shielding is used; however, approximately 1 inch of lead surrounds the photomultiplier to a height of 5 inches. The counter-sample geometry is shown in Figure 29.

For beta emitters of moderate energy, thin organic scintillators provide a lower background for a given efficiency than do gas-filled counters ⁽³⁵⁾. A typical background relationship might be 7 counts/hr and 30 counts/hr for a 1-inch diameter scintillation counter and a standard gas-filled counter respectively. For a given sensitive cross sectional area a thin circular scintillator has a minimum volume. Half the wall area of such a counter can be made an anticoincidence counter, thus about half the gamma ray background will cancel itself out. One such counter, shown in Figure 30, uses a 0.35-mm thick anthracene scintillation crystal. This is inserted into a well-shaped plastic anticoincidence shield which has an access slot for Mylar film-mounted samples. For betas having 0.6-Mev and 0.3-Mev end point energies, this counter has efficiencies of about 30% and 20% respectively.

Accurate measurement of the surface dose due to extended beta sources requires the use of scintillators which are thin compared to the lowest energy betas to be measured. A thick layer of scintillator will give a relatively low response at low beta energies. ⁽³⁶⁾

A device for scanning unidimensional radiochromatograms of compounds containing (or labeled with) beta emitters has been developed using end window and windowless gas flow counters. ⁽³⁷⁾ Two inches of lead surrounds the detector and the resulting background is 17.7 counts/minute.

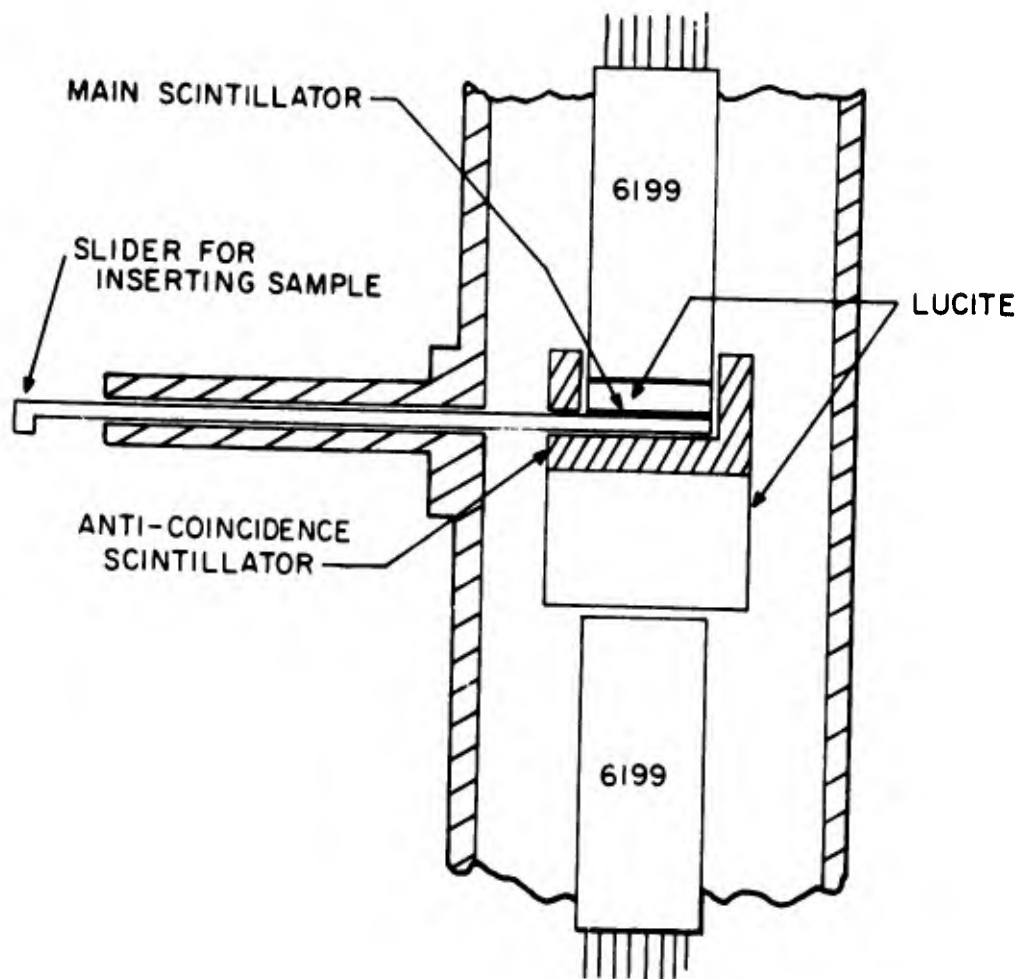


FIGURE 29. SCHEMATIC ARRANGEMENT OF THIN-SCINTILLATOR COUNTER
 IN WELL-SHAPED ANTI-COINCIDENCE COUNTER

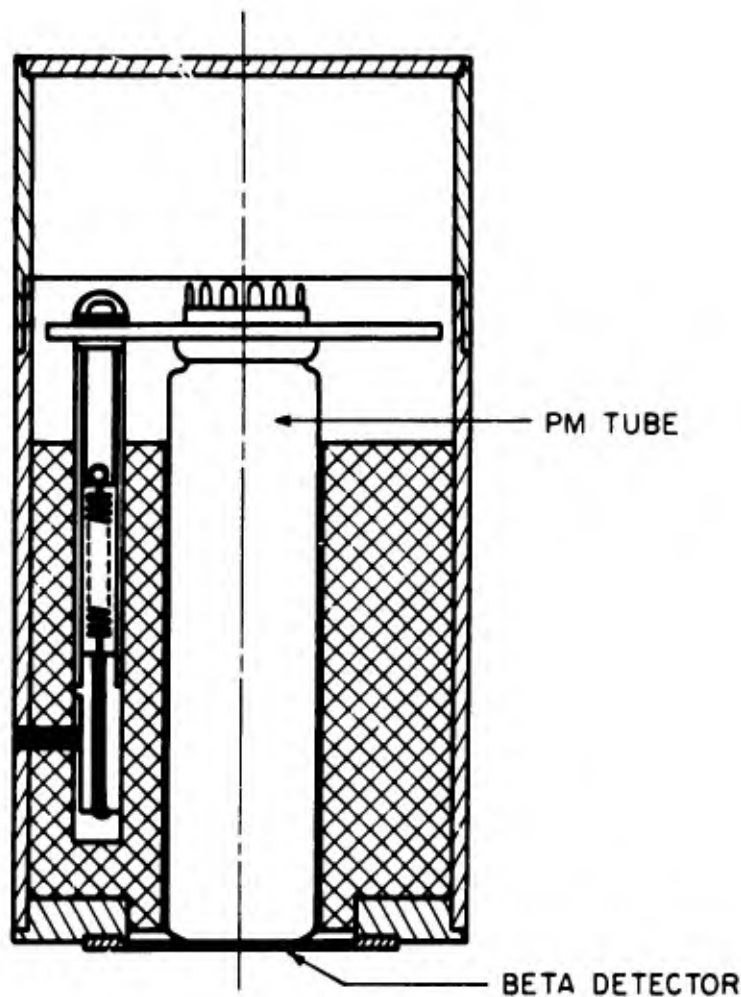


FIGURE 30. LOW LEVEL BETA COUNTING HEAD

Instruments have been developed for measuring the concentration of Sr^{90} in drinking water. The maximum permissible concentration, MPC, of Sr^{90} in drinking water is $40 \mu\mu\text{curies}$ per liter, which is about 84 counts per minute per liter. ⁽³⁸⁾ Very stable counting equipment circuitry is therefore necessary. Rings of 10 and 36 Geiger tubes respectively have been used for anticoincidence shielding for two different instrument configurations.* Anticoincidence guarding of the scintillation

*The ring of Geiger tubes is mainly a cosmic ray anticoincidence shield.

detectors was also provided. Samples to be counted were placed on a 1-inch diameter Millipore filter and covered with a 0.001-inch thick Mylar foil. Backgrounds of 0.7 c/m and 0.2 c/m with counting efficiencies of 15% to 18% and 80% respectively were obtained with the Geiger and scintillation counters for Sr⁹⁰.

GAMMA RADIATION MEASUREMENTS

It has been noted in the discussion of alpha and beta radiation measurements that the range of these particles is generally short in an absorbing medium, and hence the amount of absorber between the source and the sensitive volume of the detector must be minimized. This is not necessarily the case for gamma and x-radiation measuring instruments. While the ionization phenomenon is a secondary process by which a photon loses energy, other more basic interactions with matter also decrease the photon energy. The three most important of these interactions are the photoelectric effect, Compton scattering, and pair production. (39)*

In the photoelectric effect the incident photon is completely absorbed by the atom, and a photoelectron is ejected from the highly bound orbital atomic electrons. The ejected electron is from the K shell more than 80% of the time. (op.cit.3) Momentum is conserved by the recoil of the entire residual atom. The energy of the ejected photoelectron is $T = h\nu - Be$ where $h\nu$ is the photon energy and Be is the binding energy of the orbital electron. The cross section (τ_a) for photoelectric interactions with materials of differing atomic number (Z) can be approximated by

$$\tau_a \sim \text{const.} \frac{Z^4}{(h\nu)^3} \text{ cm}^2/\text{atom.}$$

* Other interactions occur, but they are small effects and are not generally basic to a form of radiation detector. Such effects are coherent (Raleigh) electron scattering, annihilation radiation, fluorescence radiation, and bremsstrahlen.

The linear attenuation coefficient for the photoelectric effect is then given by $\tau = \tau_a N$ where N is the number of atoms per cubic centimeter. Tables of photoelectric absorption coefficients have been developed from theoretical calculations. (40)

Compton scattering is the scattering of a photon by free (or very lightly bound) electrons (Figure 31).

For low energy photons, this case can be treated classically by conservation of momentum considerations. The energy of the struck electron is given by

$$T = h\nu_0 - h\nu$$

$$T = h\nu_0 \frac{\alpha(1 - \cos \theta)}{1 + \alpha(1 - \cos \theta)} \quad \text{where } \alpha = \frac{h\nu_0}{M_0 c^2}$$

and the energy of the scattered photon is

$$h\nu' = \frac{M_0 c^2}{1 - \cos \theta + (\frac{1}{\alpha})}$$

The linear attenuation coefficient for Compton scattered photons is given by $\sigma = NZ_e \sigma_e$, where σ_e is the electronic collision cross section. Tables of mass attenuation coefficients, $(\frac{\sigma}{\rho}$ where ρ is the density in g/cm^3) are given in Reference 40.

Pair production occurs with increasing frequency when the incident photon energy is greater than 1.02 Mev. The photon is completely absorbed, and in its place an electron-positron pair appears. The kinetics of this interaction are given by

$$h\nu = (T_- + M_0 c^2) + (T_+ + M_0 c^2)$$

where $h\nu$ is the incident photon energy, $M_0 c^2$ is the rest mass energy of the electron and positron, and T_- and T_+ are the kinetic energies associated with the electron and positron

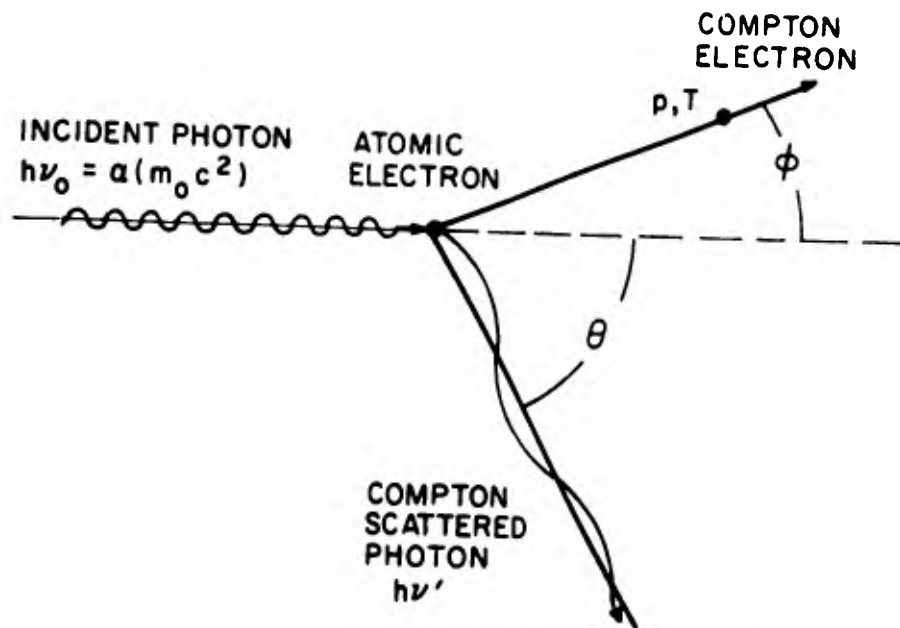


FIGURE 31. TRAJECTORIES IN THE SCATTERING PLANE FOR THE INCIDENT PHOTON $h\nu_0$, THE SCATTERED PHOTON $h\nu'$, AND THE SCATTERING ELECTRON WHICH ACQUIRES MOMENTUM p AND KINETIC ENERGY T

respectively. The pair production linear attenuation coefficient, K , is given by

$$K = {}_a K N \text{ cm}^{-1}$$

where N is the number of atoms per cubic centimeter and ${}_a K$ is a nuclear cross section. While pair production generally occurs in the field of a nucleus, it can also take place in the field of an electron. Consequently, calculations of cross sections and attenuation coefficients have also been made for this case.*

From the preceding discussion the total linear attenuation coefficient associated with the probability of a photon traversing a given amount of absorber is $\mu_0 = \alpha + \tau + k$ where μ_0 originates in the expressions

$$\begin{aligned} I &= I_0 e^{-\sigma x} e^{-\tau x} e^{-kx} \\ &= I_0 e^{-(\sigma + \tau + k)x} \\ &= I_0 e^{-\mu_0 x} \end{aligned}$$

Each exponential is the cross section for survival by a photon, for the Compton, photoelectric, and pair production interactions. The total mass absorption coefficient (μ_0/ρ) for several materials and photon energies are given in Reference 40.

A typical curve of mass attenuation coefficients for sodium iodide is given in Figure 32.

Detection and energy measurements are based on these interactions as follows:

1. Ionization caused by the secondary electrons created through photoelectric, Compton, and pair production effects in the various materials used in ionization chambers can be

*For computational procedures and tabulated values of the cross sections see References 3 and 39.

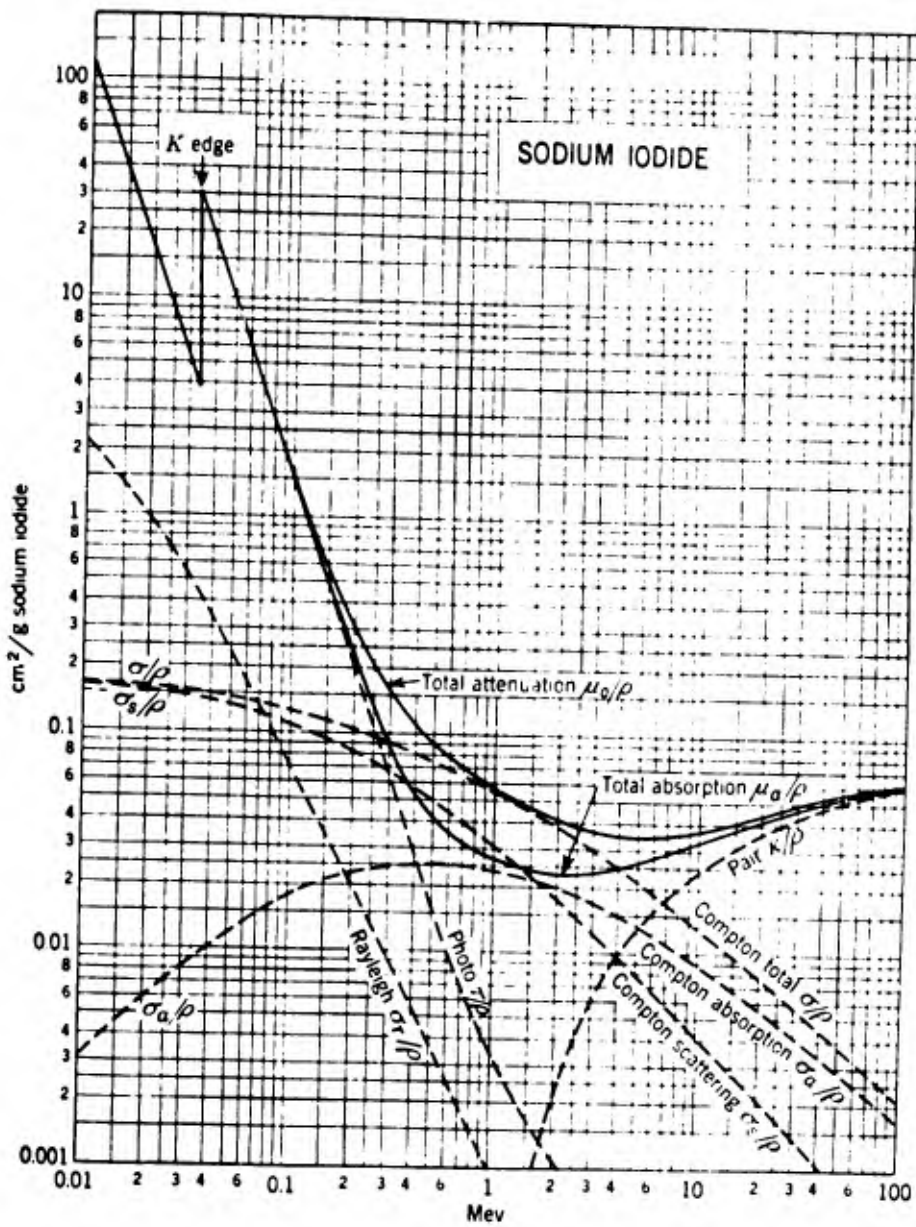


FIGURE 32. MASS ATTENUATION COEFFICIENTS FOR SODIUM IODIDE

measured by collecting the ion pairs formed and either counting the voltage pulses or averaging the pulses and measuring an ion current instead. A similar situation exists for solid state ionization chambers, except that electron-hole pairs are created instead of ion pairs. These ionization chambers include Geiger and proportional-counter types.

2. The scintillation counter is perhaps the most efficient and sensitive instrument for counting photon fluxes and measuring the photon energies. All three types of interactions, i.e., photoelectric, Compton, and pair production, occur in NaI(Tl), CsI(Tl), anthracene, Ne102 liquid scintillators, etc. Where only gross counting is required, these effects occur in such a short time interval that only the summation pulse is observed and counted. Corrections for geometry and scintillator efficiency must be made to obtain an absolute figure in terms of number of photons incident on the detector per square centimeter per second.* Many instruments of both the laboratory and portable type employ scintillators for their detectors.

3. Scintillation spectrometers have been developed for performing energy analysis of photons from gamma- and x-ray sources. The most efficient scintillator and the one having the highest light output per Mev of energy deposited in it is

*The intrinsic efficiency of a solid state detector such as NaI(Tl); semiconductor detectors, such as silicon junctions; and liquid scintillators, such as a xylene-terphenyl-diphenyl-hexatirene, is given by

$$\epsilon = (1 - e^{-\mu_0 x})$$

where μ_0 is the total absorption coefficient and

x is the thickness of the scintillator.

NaI(Tl). Instrumental effects, counting statistics (source strength), energy, and the effects due to interaction of radiation with matter in general, challenges the interpretation of energy spectra. A typical pulse height distribution (i.e., an energy spectrum which has not been corrected for the previously listed effects) is shown in Figure 33 for the 0.835 Mev gamma from Mn^{54} . * (41) The total absorption peak (photoelectric effect) is the shaded area. Most of the events which occurred in the unshaded area are due to Compton scattering, in which the scattered photon escaped from the NaI(Tl) crystal and hence only deposited a fraction of its energy in the detector. The indicated backscatter peak is due to photons which traversed the NaI(Tl) crystal, were subsequently scattered 180° back into the crystal, and were then totally absorbed. A second example of a pulse height spectrum is shown in Figure 34. The 3.13 Mev gamma of S^{37} is apparent along with two escape peaks and an annihilation radiation peak. Since this is a high energy gamma (>1.02 Mev), pair production effects are seen in the form of three peaks. All the energy in excess of 1.02 Mev is carried away by the positron-electron pair. Annihilation of the positron yields two 0.511 Mev photons. If both of these quanta completely escape detection in the crystal, the resulting energy loss in the crystal will be 1.02 Mev below the energy of the total absorption peak, and a peak will appear which is indicative only of the kinetic energy of the pair. This is the "double escape peak". When only one of the annihilation quanta is totally absorbed, then a peak which is 0.511 Mev less in energy than the incident photon energy will appear. This is the "single escape peak". The low intensity peak at 0.511 Mev is due to the interaction of high energy photons (>1.02 Mev) in the material around the detector, and the subsequent detection of the annihilation radiation of the pair-produced quanta from these high energy photons.

*The pulse height measurement is a determination of the amplitude of the pulse from the photomultiplier. Pulses of differing amplitude are sorted and stored according to height in a multi-channel analyzer with a magnetic core memory.

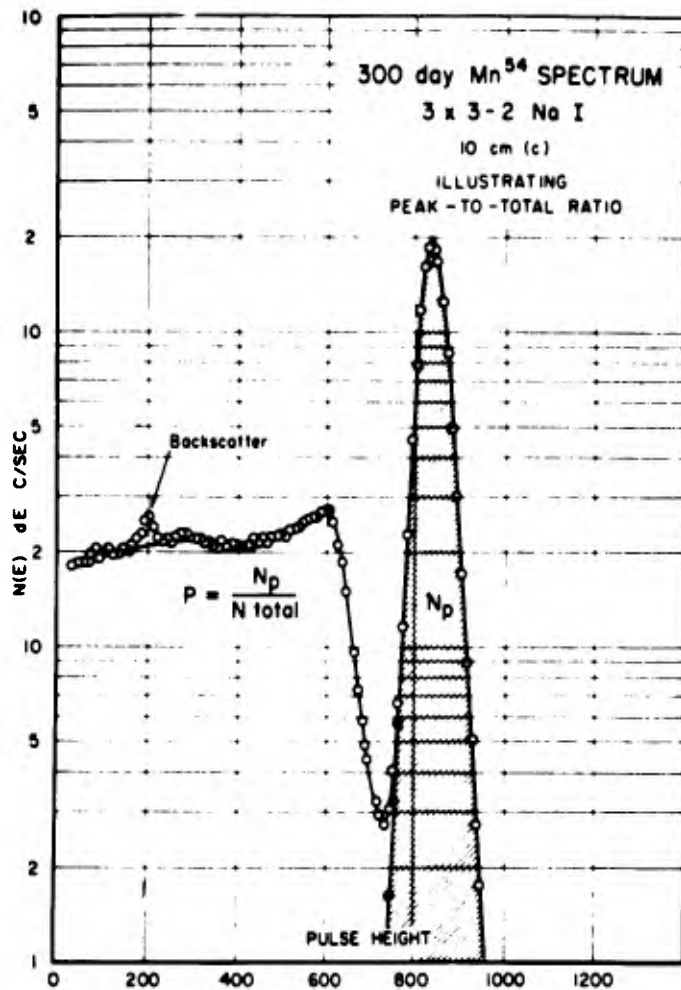


FIGURE 33. ILLUSTRATION OF PEAK-TO-TOTAL RATIO

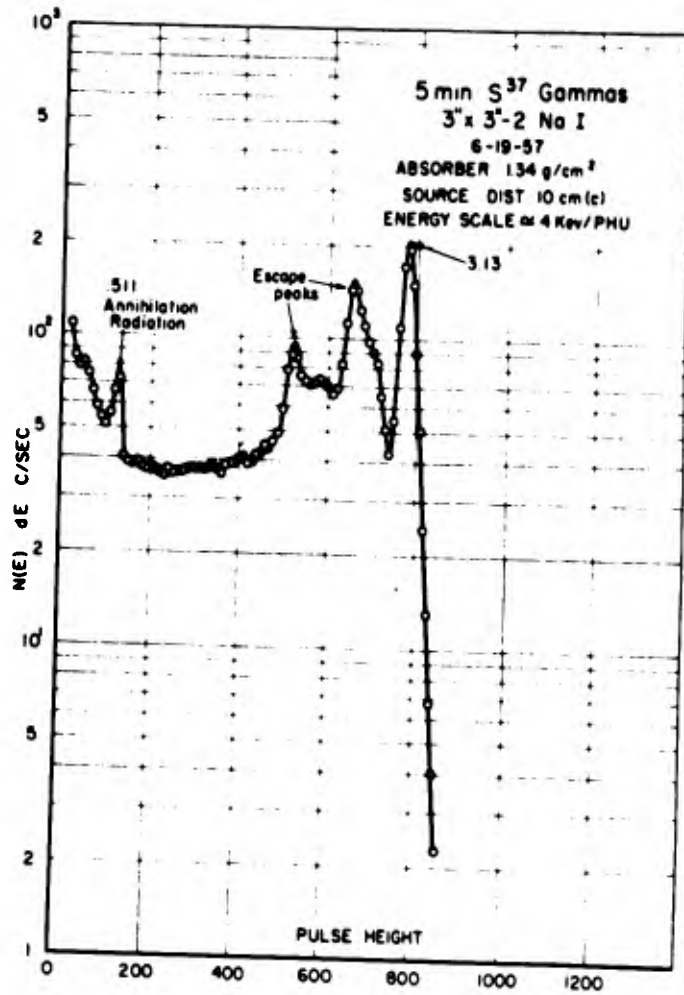


FIGURE 34. GAMMA-RAY SPECTRUM OF 5 MIN. S^{37}

Another peak-produced effect is caused by the escape of the iodine K-X-ray created by a photoelectric interaction in the sodium iodide crystal. The peak is reduced in energy from that of the total absorption peak by the energy of the iodine K-X-ray.

The determination of the absolute counting rate in terms of photons per second per cm^2 of a source can be obtained from the pulse height distribution. The area under the photopeak is defined as the symmetrical Gaussian, fitted to the high energy side of the experimental photopeak. The ratio of the area under the photopeak to the total area under the pulse height distribution (peak to total ratio) can be determined with standard calibration sources and source-detector geometries. The absolute emission rate of any single gamma ray will then be given by

$$N_o = \frac{N_p}{\epsilon PA}$$

where N_p is the area under the photopeak

ϵ is the absolute electron efficiency for the incident photon energy

P is the peak to total ratio

A is the correction for a source absorption and any beta absorber used in the measurement.

The corrected emission rate N_o can be further corrected to absolute source strength and absolute dose rate. Source strength is obtained from the fact that

$$1 \text{ curie} = 3.7 \times 10^{10} \text{ disintegrations per second.}$$

Hence,

$$S = \frac{N_o \frac{\text{dis}}{\text{sec-cm}^2}}{3.7 \times 10^{10} \frac{\text{dis}}{\text{sec-curie}}} = \frac{N_o}{3.7 \times 10^{10}} \frac{\text{curies}}{\text{cm}^2} \times \frac{a}{r^2} \times 4\pi$$

where a is the cross sectional area of the detector and
 r is the source to detector distance.

An approximation to the dose rate is given by

$$R \cong 0.55 \Sigma(h\nu)mr/(hr)(mc) \text{ at one meter from the source.}^*$$

The qualitative identification of radioisotopes is made by first determining the energy of any prominent peaks in the spectrum and then comparing the energies with the emission energies of known sources and spectral distribution tabulations, such as those of Reference 41. Calculation of escape peaks, annihilation radiation, and other features can then be carried out according to the formulæ in References 3, 29, and 41.

Of the many instruments developed for measuring gamma radiation, one such device (an ionization chamber) may be described as follows: The ionization chamber consists of two electrodes to define the collecting region of the gas volume.⁽⁴²⁾ A potential of 360 volts is maintained between the electrodes. The chamber is cylindrical, with a volume of 12.250 liters. A vibrating reed electrometer is used to measure the small (10^{-14} amperes) ion currents. To record total background radiation for a period of time, the electrometer integrates the current output of the ionization chamber. This is done by monitoring the charging of a calibrated capacitor for a known length of time. Using this technique, backgrounds of $0.0528 \frac{mr}{hr}$ have been measured.

Another sensitive ratemeter method for measuring gamma radiation couples vibrating reed electrometers, capable of measuring currents as low as 2×10^{-17} amperes, to thimble ionization chambers.^{** (43)} Dose rates as low as $0.5 \frac{mr}{hr}$ have been measured.

* A detailed calculation of dose rate and intensity appears on Page 723 of Reference 8.

** The vibrating reed electrometer is an Applied Physics Corporation Carey Model 31 with high megohm resistor turret.

The variation of counting rate with sample volume is shown in Figure 35. The reduction in counting rate is greatest for the lower energy gamma rays. Knowledge of this curve then allows a routine type of correction for sample volume. This has been especially useful in the measurement of radioactivity in fecal specimens. (These specimens were compressed into a standard cylindrical geometry before counting was done.)

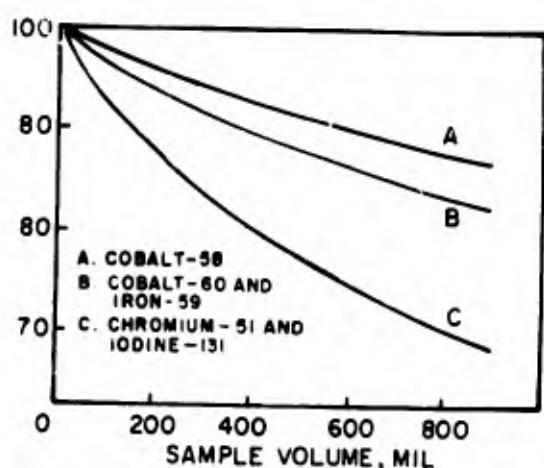


FIGURE 35. EFFECT OF INCREASING SAMPLE VOLUME OF CONSTANT ACTIVITY ON COUNTING RATE

Gamma detectors using plastic scintillators have been developed for counting large volume (up to 1000 ml) samples. (44) The plastic phosphor is mounted on a 16-inch photomultiplier and is operated with conventional electronic counting circuitry and pulse height analyzers. Figure 36 is a cross-sectional view of the well detector.

Two scintillation-type exposure ratemeters have been developed which utilize negative feedback circuitry to help eliminate the inherent dc amplifier drift problems associated with portable, electrometer-type, beta-gamma ion chamber instruments. (45) Both instruments utilize NaI(Tl) crystals one millimeter thick to produce a maximum sensitivity of 0.05 mr/hr full scale with a drift rate of 3% of full scale per hour. One instrument has logarithmic scales while the second has linear scales. Both instruments have ranges extending from 0.5 mr/hr to 5.0 R/hr. Each instrument weighs 5 pounds.

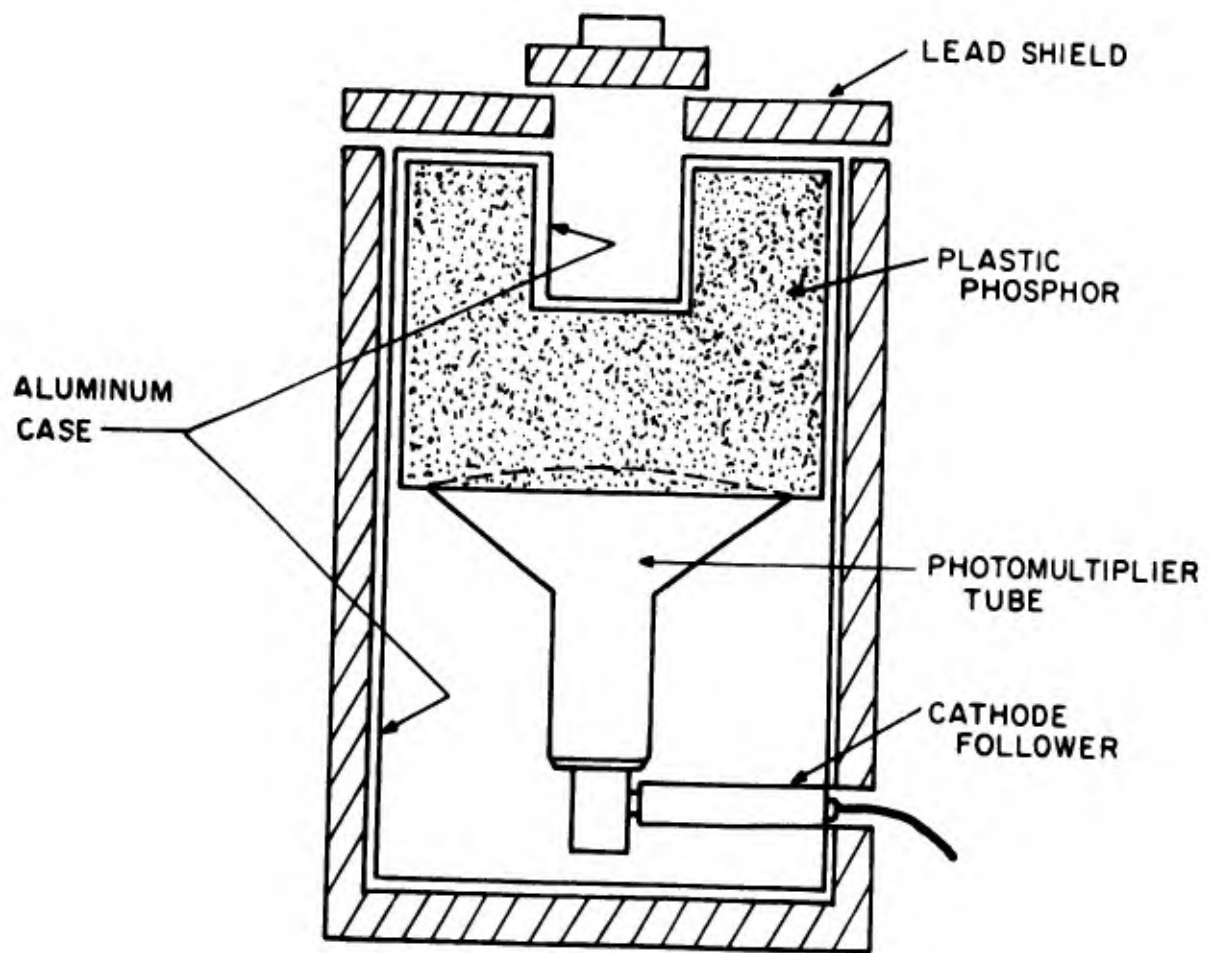


FIGURE 36. CROSS-SECTIONAL VIEW OF THE WELL DETECTOR

An in-line gamma monitor has been developed to measure the gross gamma activity of a solution contained in a closed sample cell. ⁽⁴⁶⁾ (The solution can be continuously circulated through the cell.) See Figure 37. The detector is a NaI(Tl) scintillator coupled to an RCA 6655 photomultiplier. The current from the photomultiplier tube is measured by an electrometer circuit. Solutions of known activity are used to calibrate the system.

A stack effluent radioisotope monitor has been developed to simultaneously analyze three gamma components of I^{131} and Ru^{103} and the beta emission from Ru^{106} in the effluent material. NaI(Tl) crystals and photomultiplier tubes make up the gamma detectors, and an anthracene crystal and a photomultiplier tube make up the beta detector.

Scintillation counters have also been used in satellite and high altitude rocket experiments. ⁽⁴⁷⁾ Cosmic rays, auroral particles, Van Allen radiation, and solar plasmas have all been subjects of measurements. Light weight, compact, counters, and ruggedized components (such as photomultiplier tubes) were necessary. These requirements have helped advance the state-of-the-art of circuitry sophistication, component ruggedness, and microminiaturization. Miniature and/or ruggedized components are now commercially available products.

Anticoincidence mantles surrounding sodium iodide crystals have been frequently used to reduce background effects in counting low levels of sample activity and to reduce the Compton distribution in the pulse height spectra by detecting escape radiation. Liquid and plastic scintillators have been used as the mantle material. One such device consists of a 5-inch diameter by 5-inch high NaI(Tl) crystal with a 3/4-inch diameter by 3-3/8-inch long well and a plastic scintillator anticoincidence mantle 18 inches in diameter by 18 inches high. ⁽⁴⁸⁾ The well in the plastic mantle is 5-5/8 inches in diameter and 12 inches

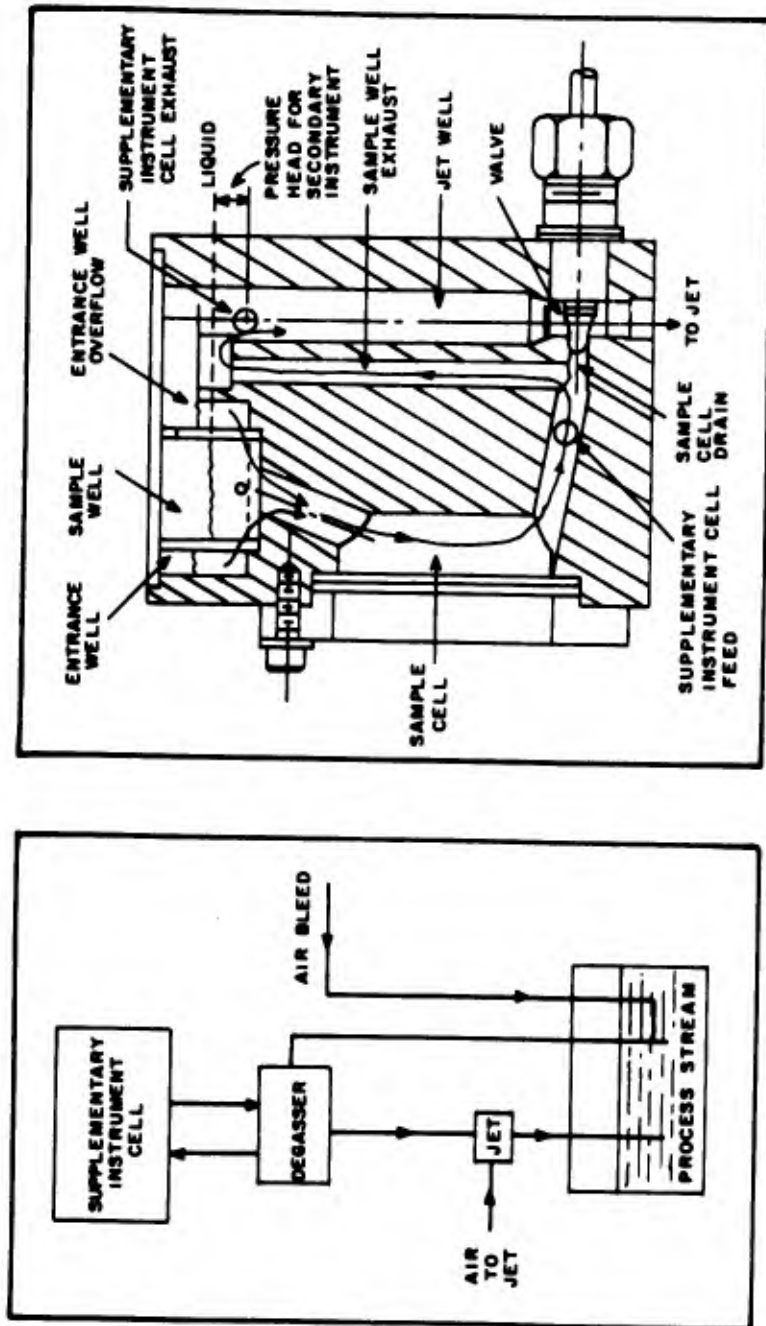


FIGURE 37. JET AIRLIFT SAMPLING SYSTEM AND CROSS SECTION OF DEGASSER

deep with a 1-inch diameter sample access hole to the NaI(Tl) crystal. (Figures 38, 39.)

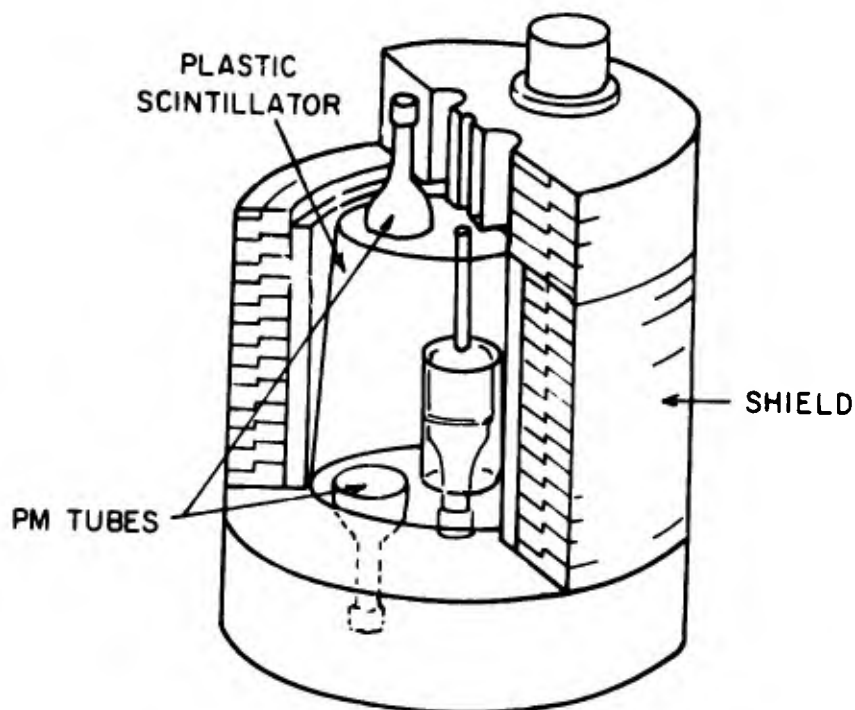


FIGURE 38. THE GAMMA-RAY SPECTROMETER

The entire package is surrounded by three inches of lead and one inch of mercury. The top and bottom of the shield are eight inches of iron. The output of the 5-inch photomultiplier tube is fed to a pulse height analyzer. The pulses from the four phototubes mounted on the anticoincidence (AC) shield are summed in one of two modes depending on the sample activity. For sample activities greater than 5 μ curies most of the events in the AC shield are due to escape radiation from the NaI(Tl) well crystal. In this case, the AC signals are summed and fed to the anticoincidence gate of the pulse height analyzer. If the sample activity is low, most of the AC pulses are due to background radiation, and in this case the signals are summed in pairs and placed in coincidence. The coincidence requirement eliminates all counts due to tube noise and results in more stable operation. However, some low energy gamma rays escaping from the NaI(Tl) are not counted in this mode. The efficiency of the AC mantle depends

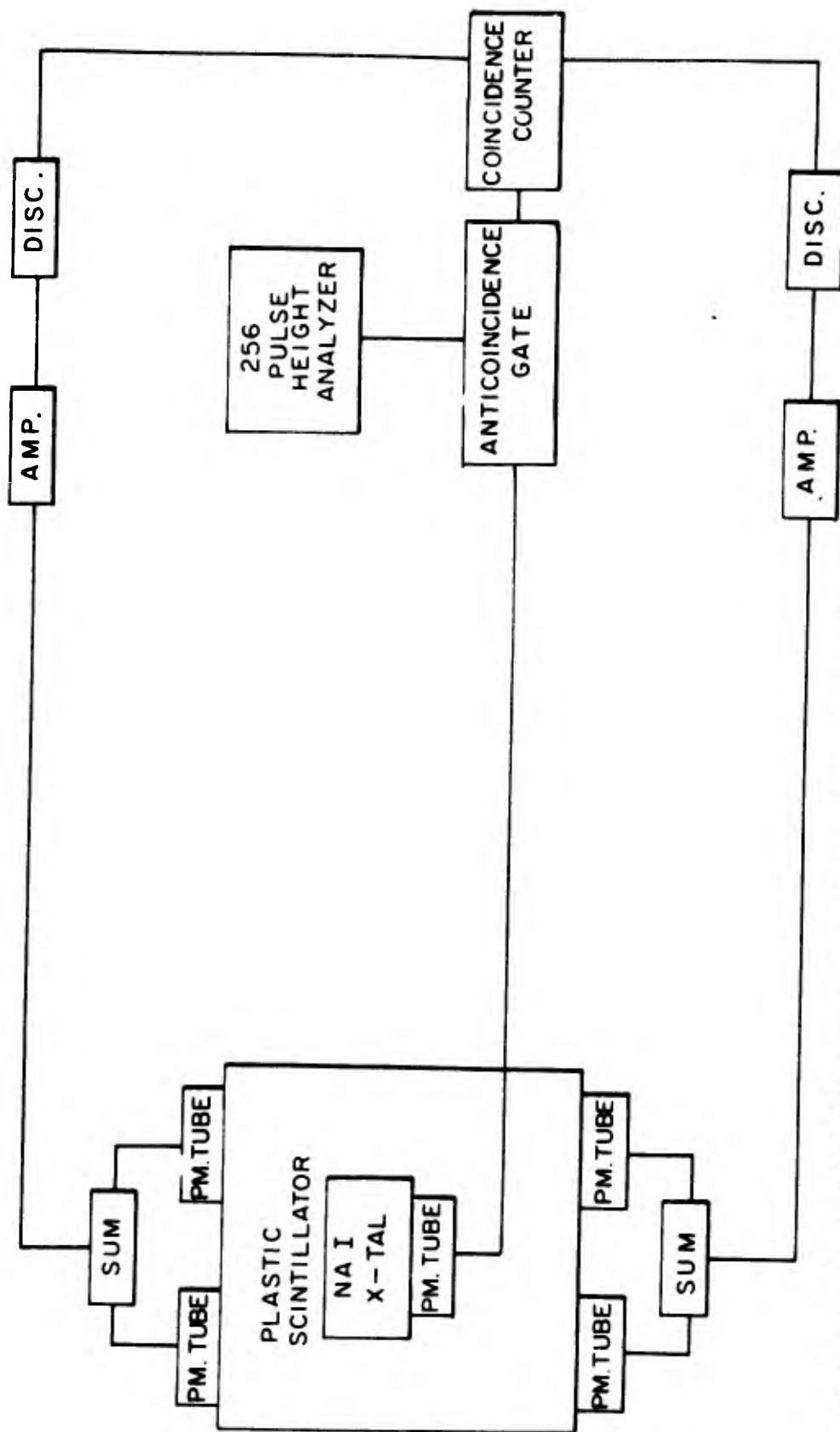


FIGURE 39. INFORMATION FLOW USING THE ANTICOINCIDENCE MANTLE WITH PAIRS OF PHOTOMULTIPLIER TUBES IN COINCIDENCE. (A light-tight shield encloses the NaI crystal and its photomultiplier tube.)

on the cross section of the plastic to escape quanta from the sodium iodide crystal, and on the electronic capability for detecting small energy losses in the plastic. The number of escape gamma-rays detected in the mantle is given by

$$\Delta N_o = N_o \eta_c (E_{\min})$$

where $\eta_c(E_{\min})$ is the detection efficiency of the plastic for an escape Compton quantum and is a function of E_{\min} , the minimum energy transfer required to trigger the AC gate. The theoretical efficiency of plastic for Cs^{137} varies from 81%, if all interactions are detected, to 70%, if only energy transfers greater than 50 Kev trigger an anticoincidence pulse. The peak-to-total ratio for single energy gamma emitters with and without the anticoincidence shield is shown in Figure 40, along with the total absorption peak efficiency for the NaI(Tl) crystal, plotted as a function of initial gamma-ray energy. The effect of the anticoincidence shield on the background integral counting rate of the NaI(Tl) crystal is generally a reduction in the counting rate by a factor of two.

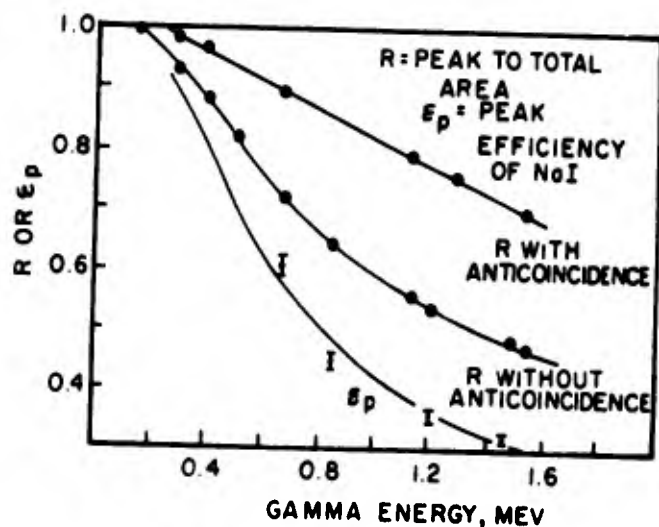


FIGURE 40. R AS A FUNCTION OF INITIAL GAMMA-RAY ENERGY.

Figure 41 is a differential spectrum with and without the AC mantle. Most of the background radiation above 3 Mev is eliminated by the AC mantle. The lower background counting

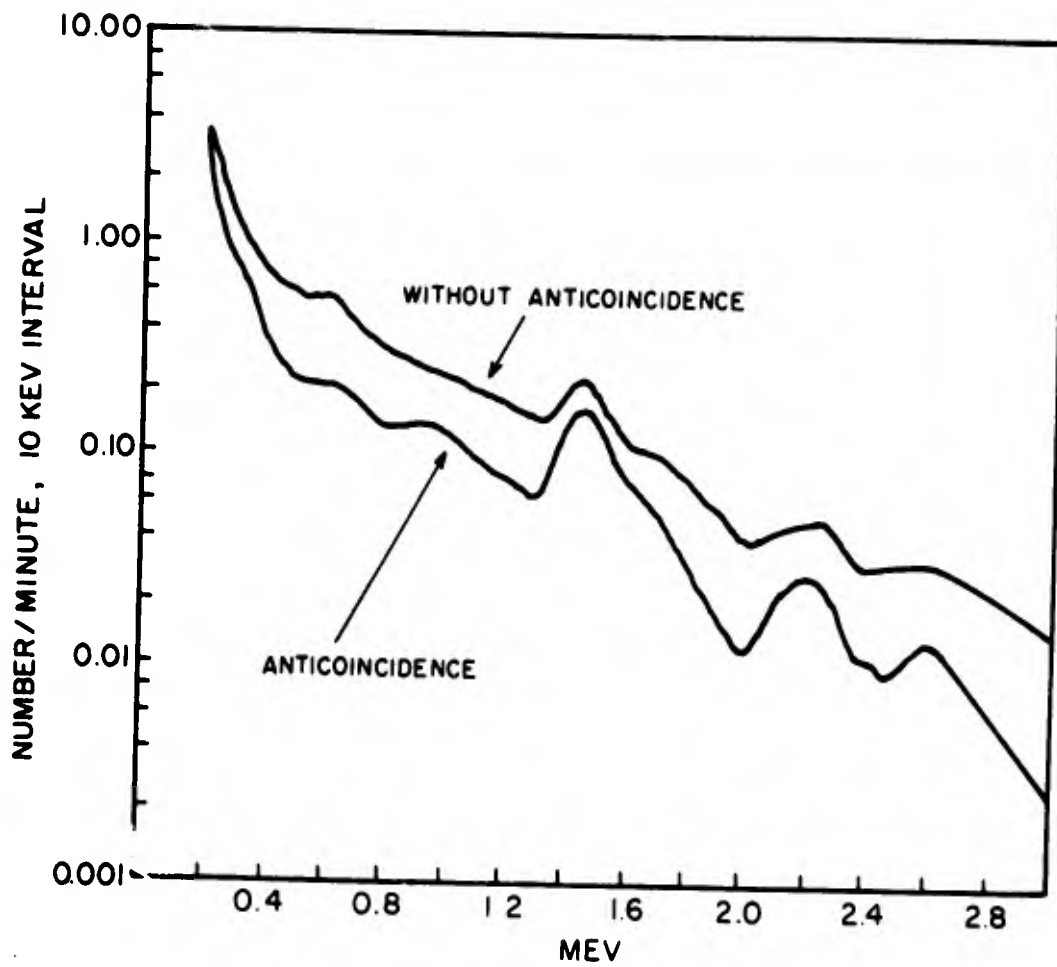


FIGURE 41. DIFFERENTIAL BACKGROUND SPECTRA OF THE NaI WELL CRYSTAL WITH AND WITHOUT ANTICOINCIDENCE (SUM SPECTROMETER)

rate in the AC mode is due to reduced Compton spectra in the large crystal and the detection by both scintillators of radioactivity and build-up radiation originating in the shield. Both background spectra and the spectra of low activity samples consist of small peaks superimposed on a continuous distribution of gamma ray energies. Since the number of primary gamma rays recorded in a total absorption peak is not affected by the AC mantle, the ratio of spectra taken with and without antineutrino incidence is an aid in locating small peaks hidden in the continuous distribution.

Experiments with various thicknesses of lead shield surrounding a 3-inch diameter by 3-inch high NaI(Tl) crystal have indicated that a 4-inch thick shield is optimum for reducing background for low level radioactivity measurements. Added shielding does not significantly reduce the background sufficiently to justify the added weight.⁽⁴⁹⁾ Graded shields consisting of 1/16 inch of cadmium and 1/32 inch of copper are used within the shield castles to suppress the 75-Kev lead K-X-ray.

An automatic analyzing system was established at Hanford for monitoring reactor effluent cooling water.⁽⁵⁰⁾ A combination of mechanical coupling, automatic radiochemical separation of interfering isotopes, and quantitative beta and gamma counting has been utilized to measure the total sample activity as well as the concentrations of Na²⁴, As⁷⁶, Np²³⁹, Mn⁵⁶, Cu⁶⁴, Si³¹, and P³². Gamma scintillation counting techniques are used to measure Na²⁴, Np²³⁹, Cu⁶⁴, As⁷⁶, and Mn⁵⁶ sample concentrations. Pulse height analysis is necessary to measure the Na²⁴ and As⁷⁶. A gamma-gamma coincidence detection system is used to measure positron annihilation radiation from Cu⁶⁴*. The gamma detectors are 3-inch diameter by 3-inch

*Beta counting with a gas flow proportional counter is used to obtain the gross beta emission rates of Si³¹ and P³².

high NaI(Tl) crystals mounted on Dumont 6363 photomultiplier tubes. Five pulse height analyzers (single channel type) and a dual channel count rate meter (for making Compton corrections to the Mn⁵⁶ and Np²³⁹ measurements) were used. Each pulse height analyzer is set to the energy range of a particular isotope to be analyzed. The counting efficiencies of the monitor were determined by counting standardized sources of each isotope. Table 8 lists the monitor sensitivity and the minimum detectable quantities.

TABLE 8

ISOTOPE	BACKGROUND C/M	MINIMUM DETECTABLE LIMIT $\mu\text{c/cc}$
As ⁷⁶	600	100×10^{-6}
Np ²³⁹	1650	50×10^{-6}
Cu ⁶⁴	3.3	30×10^{-6}
Na ²⁴	5	400×10^{-6}
Mn ⁵⁶	350	70×10^{-6}
P ³²	250	10×10^{-6}
Si ³¹	250	10×10^{-6}
Gross Beta	250	20×10^{-6}

The responses of various sizes of NaI(Tl) crystals (including well type) to point sources, extended sources, and large volume sources have been determined experimentally.⁽⁵¹⁾ In some cases anticoincidence shielding was used. The relative counting efficiencies and the shapes of the observed spectra of point sources of 0.32-, 0.662-, and 1.11-Mev gamma rays were compared for NaI(Tl) crystals of several dimensions.*

*The NaI(Tl) crystals had the following dimensions:
 a. 3 inches in diameter by 3 inches high.
 b. 9 inches in diameter by 4 inches high.
 c. 9-3/8 inches in diameter by 8½ inches high with a well.

A typical spectra is shown in Figure 42 for the 1.11-Mev gamma of Zn^{65} in a point source geometry. A spectrum from a large volume sample containing Zn^{65} is shown in Figure 43 and a comparison of the response of a large crystal to various sizes of Cs^{137} sources is shown in Figure 44. It is obvious from these illustrations that the more nearly a source-detector configuration approaches that of a point source on the axis of the detector, the better the peak resolution and the photofraction (peak to total ratio) become. It follows then that a collimated extended source will give better photofraction values than the uncollimated cases shown here. (52)

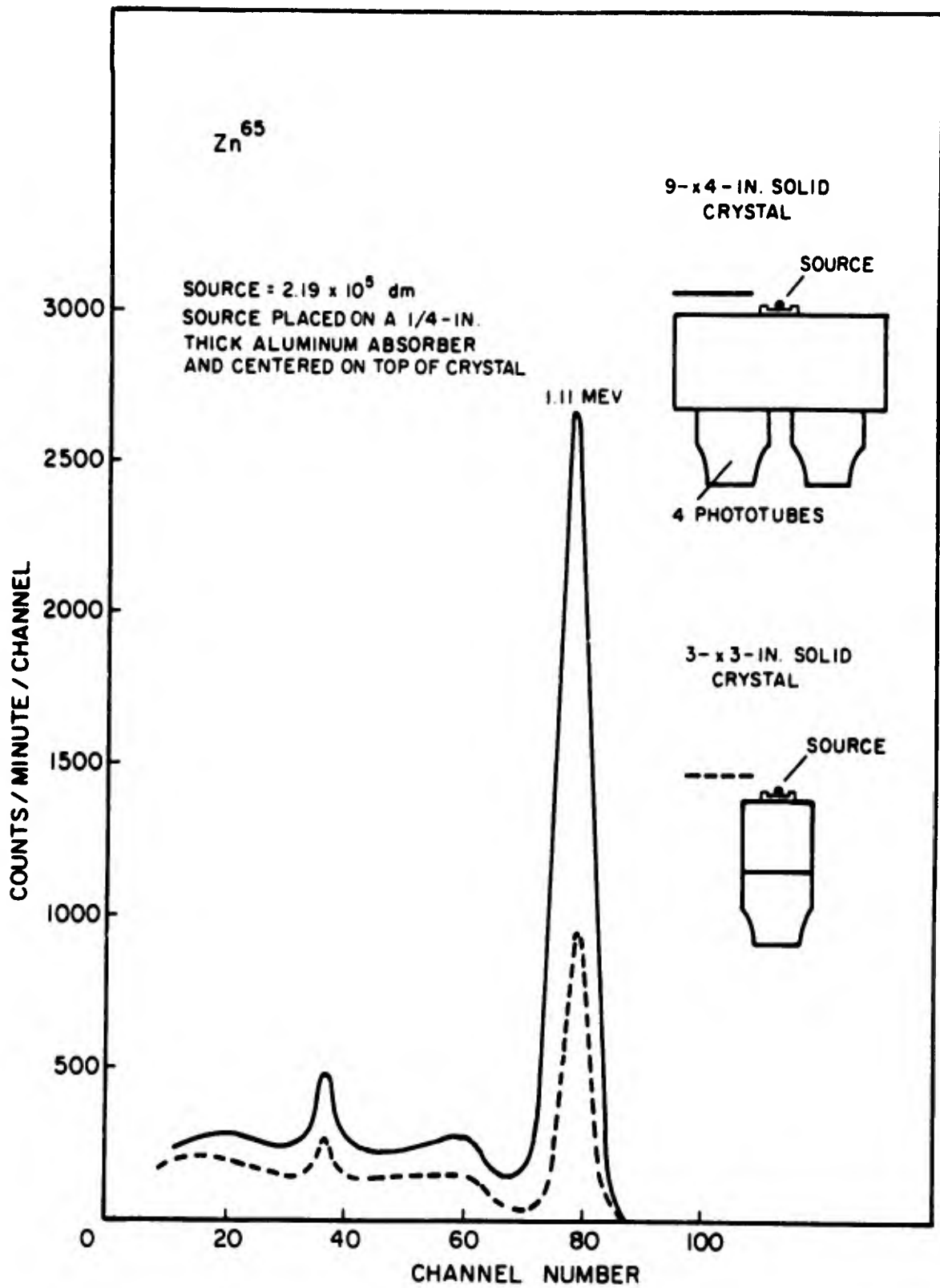


FIGURE 42. COMPARISON OF RELATIVE COUNTING EFFICIENCIES OF A SMALL AND LARGE CRYSTAL WITH A POINT SOURCE OF ZINC-65 GAMMA-RAYS

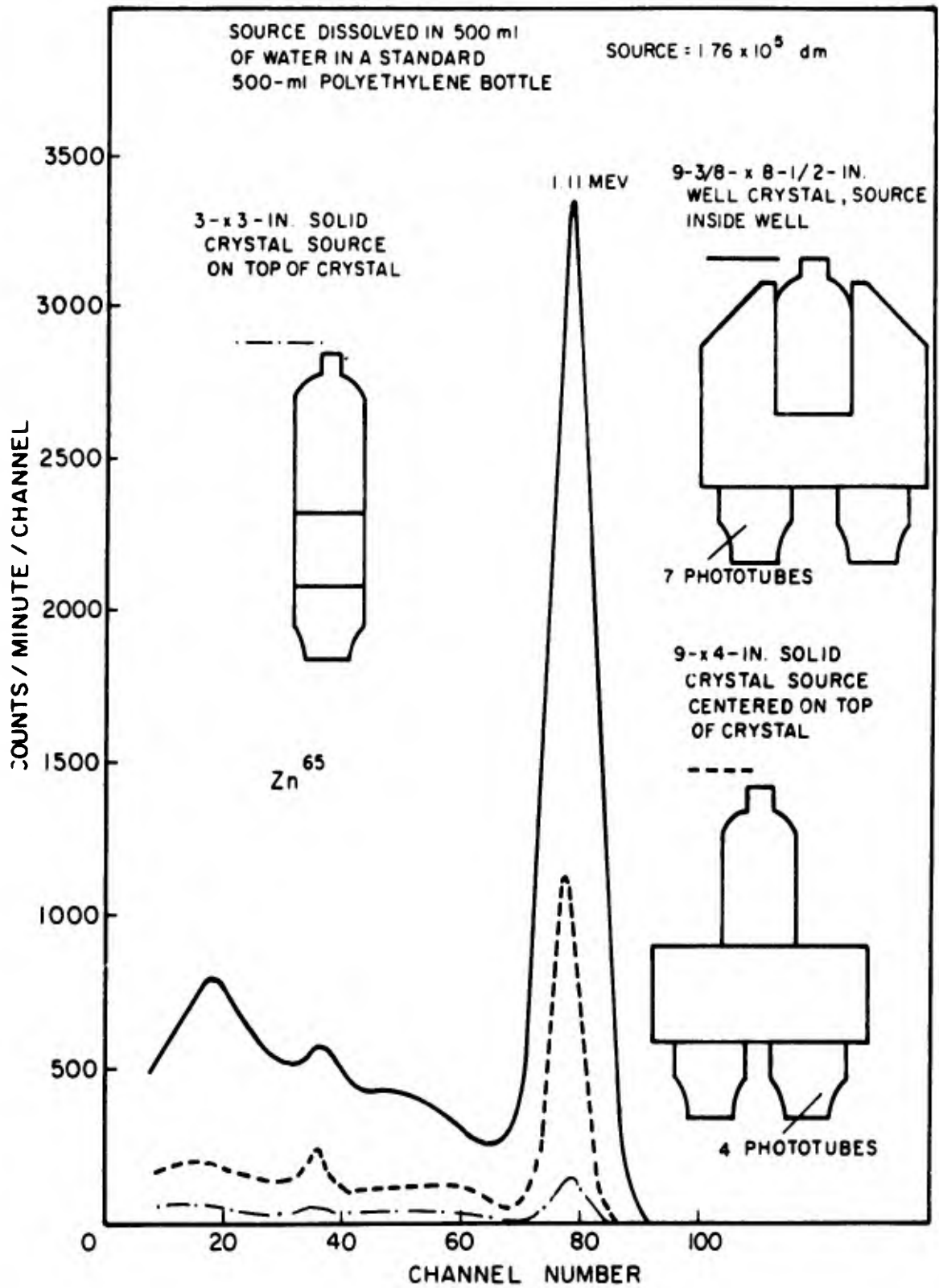


FIGURE 43. COMPARISON OF RELATIVE COUNTING EFFICIENCIES OF THREE CRYSTALS FOR ZINC-65 GAMMA-RAYS IN A LARGE VOLUME SAMPLE

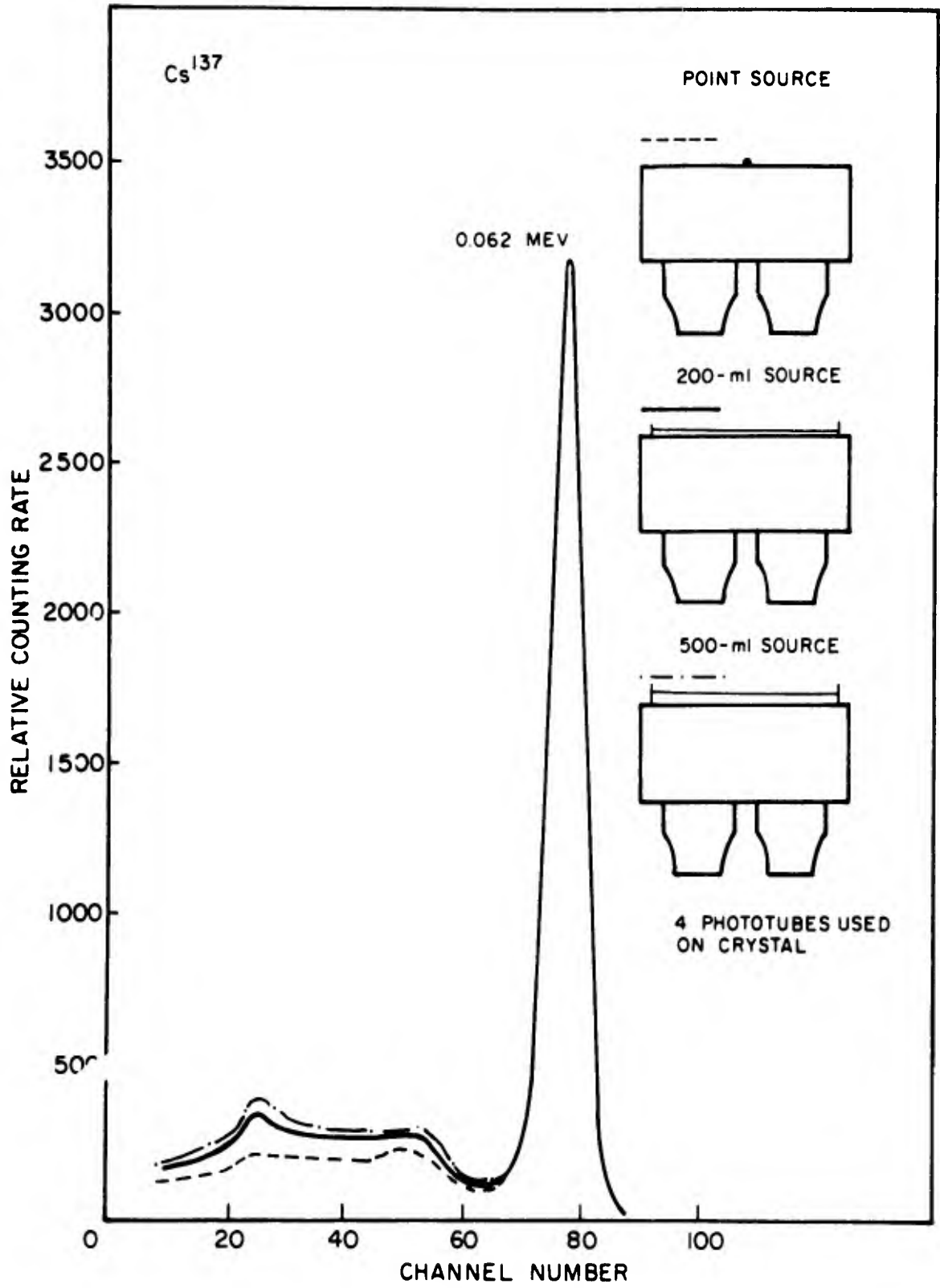


FIGURE 44. COMPARISON OF RESPONSES OF A 9- BY 4-INCH CRYSTAL TO CESIUM-137 SOURCES OF DIFFERENT SIZES (Counting rates are normalized at the photopeak)

RADIOISOTOPE ANALYSIS USING CHEMICAL AND PHYSICAL CHEMICAL TECHNIQUES

INTRODUCTION

Radiochemical procedures have been developed for performing isotope separation. Among the manifold uses of such separations, one of the most significant is the determination of radionuclide contamination in human subjects. The contamination can be combinations of alpha-beta- and gamma-emitting isotopes, and these must be separated and identified to provide data on half-life, particle emission, and energy. This information combined with biological uptake data facilitates the estimation of the potential health hazards.⁽⁵³⁾ Radiochemistry can also be used to help analyze nuclear accidents, to determine if a nuclear excursion has occurred, to estimate fluxes at various locations, and to identify contamination.⁽⁵⁴⁾ From this information some estimate can be made of the severity of the nuclear reaction and the potential and real hazards associated with it.

RADIOCHEMICAL SEPARATIONS

A scheme capable of separating U, Be, Ba, Sr, Mo, Ru, Cd, Cs, Y, and Np in good yield from a single sample has been developed for use with fission product analysis of dust samples from the Nevada Test Site.⁽⁵⁵⁾ The procedure may be summarized as follows: barium and strontium are precipitates as sulfates. Ruthenium is distilled as the tetroxide from perchloric acid. The residue is evaporated to wet dryness and the carriers dissolved in 12 N HCl. The carriers are then loaded into an anion exchange column, and the cesium and yttrium are eluted with 12 N HCl. The neptunium is eluted with 1 N HCl - 0.1 N HF and the cadmium is eluted with 3 N NH₄OH. Finally, to elute the molybdenum, NH₄OH - NH₄C₂H₃O₂ is used. There is also a decontamination procedure for each of the separated elements to remove traces of contaminating elements. Repeated centrifuging,

decanting, precipitation, and ion exchange procedures are required at various points throughout the analysis prior to counting. The separation requires a significant amount and variety of laboratory reagents and hardware. It is necessary to have a radiochemist or a well trained technician perform the analysis, which requires several hours.

Radiochemical procedures which are specific for separating particular radionuclides or fission products as a group from certain biologicals, such as blood and urine, have been established. For example, a procedure for separating fission products from urine is abstracted as follows: (56) Add 25 ml of CaCl_2 solution to a 750 ml aliquot of a 24-hour urine specimen. Add 75 ml of Sulkowitch reagent, dropwise. Allow to stand at least 12 hours, then remove the supernatant urine. Filter precipitate, transfer to a planchet for counting, and allow to dry for $1\frac{1}{2}$ hours at $80^\circ - 90^\circ\text{C}$ before counting. The fission product recovery is about 85%.

A co-precipitation type of determination of Sr^{90} in urine has been described in a 30-step procedure set forth by L. B. Farbee, and summarized along with radiochemical separation procedures for tritium, actinium, americium, protactinium, uranium, radium, plutonium, thorium, polonium and ruthenium. (57, 58)

In general, quantitative radiochemical determination of radioactivity follows an outline similar to that shown in Table 9. (The necessary reagents are also shown.) Precipitation, centrifuging, and filtration are the major operations. (59) The nuclides ultimately alpha-counted in the separation procedure outlined in the table are Ra^{226} , Ra^{223} , Ra^{224} , Th^{230} , Th^{227} , Bi^{214} , Bi^{212} and Po^{210} . Other radiochemical separation procedures have been devised so that separations of the components of interest can be accomplished in the presence of competing chemical reactions.

TABLE 9

Sample	
	Fuse with sodium peroxide, leach with water, neutralize, make 1.0M hydrochloric acid, add bismuth, saturate with hydrogen sulfide, filter
Precipitate Bismuth sulfide carrier COUNT Bismuth-214 Bismuth-212 Polonium-210	Filtrate Boil out hydrogen sulfide, add zirconium, add phosphate, centrifuge
	Precipitate Zirconium phosphate carrier
	Thorium-232 determination. Accumulate short-lived radium isotopes for known interval of time centrifuge
	Thorium-230 Thorium-227 determination. Dissolve in oxalic acid, add praseodymium carrier, add zinc sulfide
	Filtrate Discard
	Precipitate Zinc sulfide and praseodymium oxalate
	Liquid Adjust to pH-3, add zinc sulfide, saturate with hydrogen sulfide, filter
	Precipitate Zirconium carrier Discard
	COUNT Thorium-230 Thorium-227
	Filtrate Add barium, add sulfuric acid, centrifuge
	Liquid Discard
	Precipitate Barium sulfate carrier Liquid Discard
	Add water, add zinc sulfide, filter
	Filtrate Discard
	Liquid Discard
	Precipitate Zinc sulfide and barium sulfate COUNT. Radium-226 Radium-223
	Precipitate Zinc sulfide carrier lead, bismuth and polonium isotopes, discard
	Filtrate Add barium, add sulfuric acid, add zinc sulfide, filter
	Precipitate Zinc sulfide, barium sulfate COUNT. Radium-224

CHROMATOGRAPHY

The properties of differential migration of solid adsorbants and solvents can be used as a basis for separating many different substances. The migration rate for a particular adsorbant and solvent is characterized by a constant, R , which is defined as⁽⁶⁰⁾

$$R = \frac{\text{migration of adsorbed substance}}{\text{migration of the solvent}}.$$

In columnar chromatography the adsorbant can consist of such substances as activated charcoal, activated alumina, calcium carbonate, silica gel, etc. The solvents may be hydrocarbons, alcohols, etc., depending on the substances to be separated. Application has generally been limited to separations of organic substances.⁽⁶¹⁾ Filter paper chromatography requires between 10λ and 50λ of solution containing about $50 \mu\text{g}$ of substance to be separated.* The solution is deposited in a drop on the paper at one end and dried. The end of the filter paper near the sample strip is then dipped into a solvent and left while the migration of the components takes place. This type of chromatographic migration is characterized by an R_f value which is the ratio of the distance from the starting point traversed by the sample's constituents to the distance traversed by the solvent. Numerical values of R_f vary between zero and one, depending upon the solvent systems and the chemical reactions which can occur. Thus for all chromatographic separations, particular combinations of the substances to be separated, the solvent, and the porous sorption medium are necessary. Very extensive tables of R_f values are available.^(62,63)

Quantitative separation of the heavy metals An, Cd, Cu, Ni, Bi, Hg, thallous Tl and Ag from lead can be done by a downward elution procedure. The eluting solvent is 50 ml of diethyl ether,

* one $\lambda = 10^{-3}$ milliliters of solution

30 ml of methanol, 22 ml of water, and 4 ml of Anala-R-nitric acid. Figure 45 shows the R_f value of 14 cations in a descending elution with the solvent $\text{Et}_2\text{O}-\text{MeOH}-\text{H}_2\text{O}-\text{HNO}_3$ in ratios of 50:30:20:2.

Paper chromatographic procedures have been developed for separating microgram amounts of barium, strontium, calcium and magnesium.⁽⁶⁴⁾ A downward elution method is used with a 40-to-50-cm long strip of Whatman No. 1 filter-paper. One-half centimeter spots of the solutions to be analyzed are introduced via capillary tubes at 2-cm intervals along the paper. The eluting solvent consists of 50 ml of methanol, 30 ml of isopropanol, 2 ml of formic acid, 15 ml of water and 2.5 g of ammonium formate. The R_f values in this solvent are shown in Table 10.

TABLE 10

<u>Element</u>	<u>R_f</u>
Barium	0.30
Strontium	0.45
Magnesium	0.75
Calcium	0.60

To perform highly accurate quantitative work in this separation procedure normally requires purification of the filter paper to remove trace impurities of silica, alumina, ferric oxide, calcium oxide, magnesium oxide, and some metals. However, if the substance of interest is a radionuclide, this automatically eliminates the interfering influence of the trace impurities except for such effects as radiation absorption, and should therefore negate the prior treatment requirement. It also implies that highly purified water and reagents are not prerequisites for performing a chromatographic separation of radioactive elements.

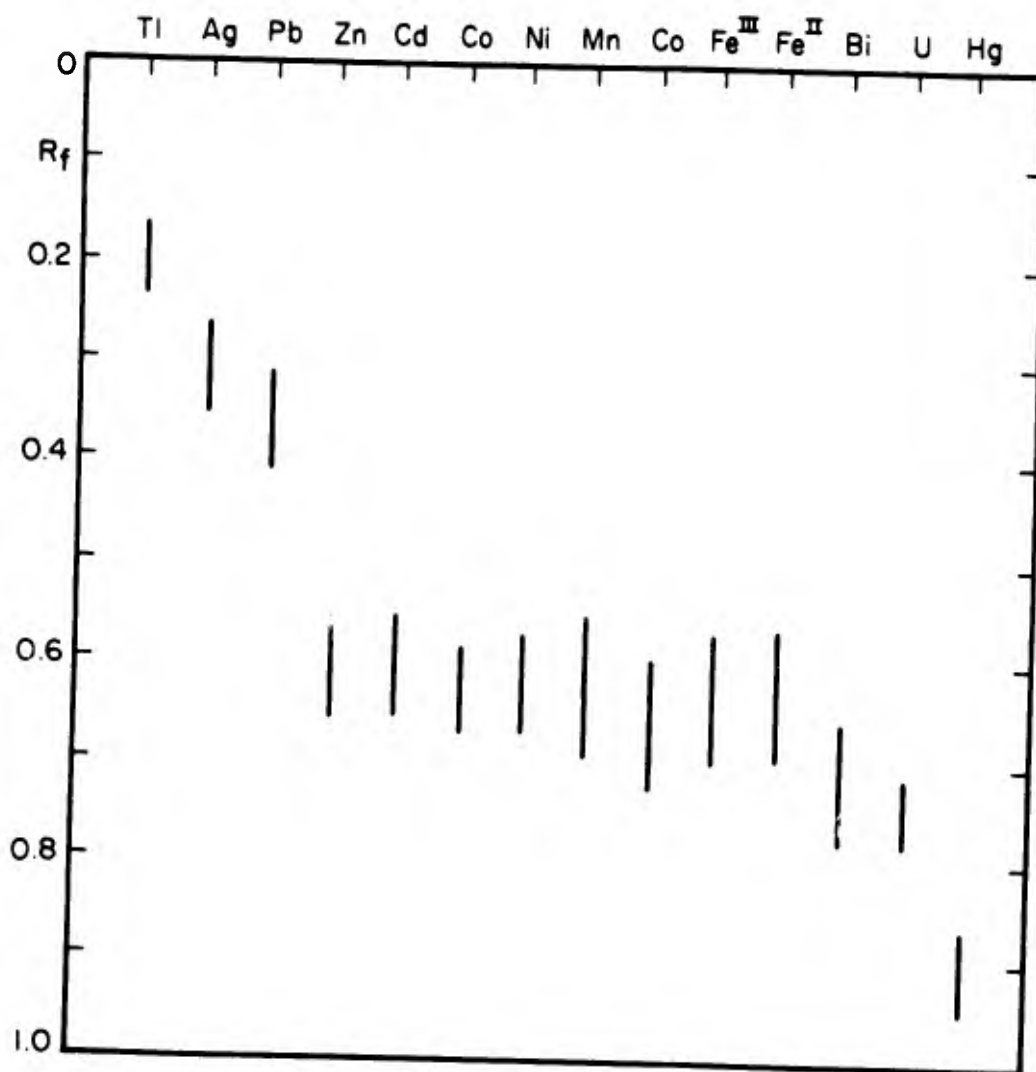


FIGURE 45. THE R_f VALUES OF 14 CATIONS IN DESCENDING ELUTION WITH THE SOLVENT SYSTEM $\text{Et}_2\text{O}-\text{MeOH}-\text{H}_2\text{O}-\text{HNO}_3 = 50:30:20:2$ (Solvent A)

Two-dimensional chromatographic separations are accomplished by first allowing a migration in one direction along the filter paper with one solvent and then, after turning the paper 90°, allowing a second migration using another solvent. The location of each component can be determined by spraying a color-producing reactant on the chromatogram. If the components are radioactive, scintillation or Geiger counter methods can be used to locate the constituents. Table 11 lists separable elements and a reactive spray for color identification.⁽⁶⁵⁾

A paper chromatographic fission product separation scheme has been devised, using an eluting solvent consisting of methyl isobutyl ketone and various concentrations of Bakers Analyzed nitric acid.⁽⁶⁶⁾ The fission product elements spotted on filter paper are listed below:

$\text{Th}(\text{NO}_3)_4$	$\text{UO}_2(\text{NO}_3)_2$	$\text{Ce}(\text{NO}_3)_3$	$\text{Ce}(\text{NO}_3)_4$	$\text{Zn}(\text{NO}_3)_2$
$\text{Sr}(\text{NO}_3)_2$	AgNO_3	$\text{In}(\text{NO}_3)_2$	$\text{Ba}(\text{NO}_3)_2$	$\text{La}(\text{NO}_3)_3$
$\text{Sn}(\text{NO}_3)_2$	$\text{Y}(\text{NO}_3)_2$	$\text{ZrO}(\text{NO}_3)_2$	$\text{Cd}(\text{NO}_3)_2$	$\text{Sm}(\text{NO}_3)_3$
$\text{Pr}(\text{NO}_3)_3$	$\text{Gd}(\text{NO}_3)_3$	RbCl	CsCl	RuCl_3
RhCl_3	PdCl_2	SbCl_3	GeF_4	AsCl_3

Only thorium, uranyl, tin, antimony, zirconyl, rubidium, cesium, arsenic, and iodine gave R_f values other than zero. Other chromatographic schemes exist for separation of ferrous and ferric iron as well as various ionic species of polyvalent metals, such as antimony, arsenic, chromium, cobalt, copper, iron, mercury, molybdenum, platinum, plutonium, thallium, uranium and vanadium.⁽⁶⁷⁾

TABLE 11

METALION	SPRAY	COLOUR OF SPOT	R _F VALUE (HEAD AND TAIL OF SPOT)
Tl(I)	0.5% w/v 8-hydroxyquinoline in absolute alcohol. Hold over ammonia and u.v. light.	Yellow-green fluorescence	0.25-0.17
Ag(I)	5% w/v tannic acid in warm 60% v/v aqueous methylated spirits. Warm the damp strip.	Brown stain	0.36-0.27 Dark hydrolysis products also present at starting line
Pb(II)	Aqueous solution of rhodizonic acid and held over-ammonia.	Red	0.41-0.22
Zn(II) Cd(II)	0.5% w/v 8-hydroxyquinoline in absolute alcohol. Hold over ammonia and under u.v. light.	Yellow fluorescence Yellow fluorescence	0.66-0.55 0.66-0.55
Cu(II) Co(II) Ni(II)	0.1% w/v rubeanic acid in absolute alcohol. Hold over ammonia.	Green Yellow-brown Blue	0.66-0.56 0.68-0.55 0.68-0.55
Mn(II)	4% v/v salicylaldehyde in 50% aqueous ethyl alcohol. Hold over ammonia and under u.v. light	Dark spot against a yellow-green fluorescent background	0.70-0.56
Fe(II) Fe(III)	0.5% w/v aqueous potassium ferrocyanide.	Blue Blue	0.70-0.58 0.70-0.58
Bi(III)*	10% w/v freshly prepared aqueous sodium dithonite. Warm.	Brown	Hydrolysed all down the paper to 0.8
U(VI)	0.5% w/v aqueous potassium ferrocyanide.	Brown	0.78-0.72
Hg(II)	0.05% w/v dithizone in chloroform.	Pale pink	0.95-0.87

* Bismuth may be chromatographed successfully only if the original solution contained 5% v/v aqueous nitric acid, then no hydrolysis products are formed, and the R_F value of Bi is 0.78-0.67.

ELECTROCHROMATOGRAPHY AND ELECTROPHORESIS

Electrochromatography relies on the electromigration of ions in a solution in which electrodes provide a dc potential. The ions migrate towards the electrodes, and those having different migration rates are separated. When a support for the solution, such as filter paper, gelatin, or an ion exchange resin is placed between the electrodes, the separation is stabilized, and a permanent separation is obtained by removing the potential and drying the support. Applied potentials vary between 100 and 400 volts dc with currents between 50 and 100 ma.

Electrochromatographic separations of rare earth metals and potassium isotopes have been accomplished in columns filled with sand, glass, wool, etc. (68) Similar procedures have been used to separate inorganic ionic mixtures of Rb^+/Na^+ ; Li^+/Na^+ ; Rb^+/Cs^+ ; $\text{Li}^+/\text{Na}^+/\text{K}^+$ and inorganic ions such as acetates and citrates. Two-dimensional pherograms with cations of the copper group (Cu^{++} ; Cd^{++} ; Hg^{++} ; Sb^{++} ; Bi^{++}) have been developed by using 0.1 M lactic acid. Subsequently, the mixture was made alkaline with NH_4OH and pherographed again at right angles. By a similar method, the metals of the ammonium sulfide group (Al^{+++} ; Fe^{++} ; Mn^{++} ; Co^{++} ; Ni^{++} ; Zn^{++} ; CrO_4^-) can be separated.

ELECTRODEPOSITION

Metals, oxides, and carbide coatings on metal or graphite have been made using electrophoretic deposition. Materials, such as B, Dy, Zr, Au, Nb, Mo, W, Re, Nb_3Sn , ZrH_2 , ThC, UC, NpC, PuC, (ZrU)C, UWC_2 , UO_2 , UNOC_2 , Ta_2O_5 and W- UO_2 and Mo UO_2 composites have been successfully deposited. (69) In solutions containing more than one element depositable on the same electrode, separation can sometimes be affected by controlling the dc potential, ionic concentration, complexing agents, etc.

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PART III

APPENDICES

REFERENCES

BIBLIOGRAPHY

Appendix A
U. S. HELICOPTER CAPABILITIES*

BELL MODEL 204 - U. S. ARMY HU1A

Carries an external sling for a 3000 pound load
Weight empty - 3860 lb
Weight loaded - 7200 lb
Cruising speed - 115 mph

BOEING VERTOL MODEL 107 - U. S. MARINE CORPS HRB-1

Carries 23-25 passengers or approx. 3500 pounds
Weight empty - 9298
Take off weight - 18,450
Cruising speed - 150 mph

BOEING VERTOL MODEL 114 - U. S. ARMY HC-1B CHINOCK

Carries a normal payload of 6000 pounds
Maximum loaded weight is 33,000 pounds
Cruising speed - 175 mph

KAMAN SEASPRITE - U. S. NAVY HU2K-1

Carries an external cargo hook with a 4000 pound capacity
Weight empty - 5052 lb
Weight loaded - 7378 lb

KAMAN MODEL 600-3 - USAF H-43 B HUSKIE

Carries a load of 1000 pounds plus two passengers
or approx. 1300 pounds
Weight empty - 4469 lb
Weight loaded - 5969 lb
Cruising speed - 109 mph

*This data is from Jane's All the World's Aircraft, 1961-1962, McGraw Hill.

SIKORSKY S-61

Carries a load of 28 passengers or approx. 5000 pounds

Weight empty - 9270 lb

Normal loaded weight - 18,700 lb

Cruising speed - 136 mph

SIKORSKY HSS-2 - U. S. NAVY

Carries a load capacity of 6000 lb

Weight empty - 10,854 lb

Normal loaded weight - 17,300 lb

Maximum speed - 150 mph

Appendix B
EQUIPMENT LIST

PRELIMINARY GAMMA RADIATION MONITOR
FOR NO. 10 CONTAINER

Harshaw 8S4/2 NaI(Tl) Integral Line Scintillation Detector

Number Required: 1

Weight: ~ 5 lbs

Price: \$225

RIDL Model 10-17 Scintillation Preamplifier

Number Required: 1

Weight: ~ 1.1 lbs

Price: \$110

RIDL Model 30-19 Amplifier-Discriminator

Number Required: 1

Power Consumption: 3 watts

Weight: 3 lbs

Price: \$325

RIDL Model 35-7 Linear/Log Count Rate Meter

Number Required: 1

Power Consumption: 9 watts

Weight: 3 lbs

Price: \$495

RIDL Model 29-1 Instrument Case and Power Supply

Number Required: 1

Power Consumption: < 60 watts

Weight: 35 lbs

Price: \$195

RIDL Model 40-9B High Voltage Power Supply

Number Required: 1
Power Consumption: < 30 watts
Weight: 4 lbs
Price: \$445

System Totals for Preliminary Gamma Radiation Monitor

Power Consumption: 102 watts
Weight: 51 lbs
Price: \$1,795

PLASTIC SCINTILLATOR GROSS GAMMA
COUNTING SYSTEM

Plastic Scintillation Phosphor NE102

Number Required: 1
Weight: ~ 0.9 lbs
Price: ~ \$800

RIDL Model 10-17 Scintillation Preamplifier and Photomultiplier

Number Required: 8
Total Weight: 48 lbs
Total Price: ~ \$2,000

RIDL Model 40-11 High Voltage Distribution Panel

Number Required: 2
Total Weight: 11.2 lbs
Total Price: \$550

RIDL Model 30-19 Amplifier-Discriminator

Number Required: 1
Power Consumption: 3 watts
Weight: 3 lbs
Price: \$325

RIDL Model 49-43 6 Decade Printing Preset Scaler

Number Required: 1
Power Consumption: 8.4 watts
Weight: ~ 3.4 lbs
Price: \$795

RIDL Model 54-8 5 Decade Printing Electronic Timer

Number Required: 1
Power Consumption: 8.3 watts
Weight: 3.4 lbs
Price: \$695

Hewlett Packard 7 Column Printer

Number Required: 1
Weight: 33 lbs
Price: \$1,995

RIDL Multiple Printing Scaler Programmer

Number Required: 1
Power Consumption: ~ 10 watts
Weight: 5 lbs
Price: \$425

RIDL Model 29-1 Instrument Case and Power Supply

Number Required: 2
Total Power Consumption: 120 watts
Total Weight: 70 lbs
Total Price: \$390

System Totals for Gross Gamma Counting System

Power Consumption: 390 watts
Weight: 178 lbs
Price: \$7,975

NaI(Tl) GAMMA SCINTILLATION SPECTROSCOPY SYSTEM

Harshaw 83F8/2 (Std. Well) Integral Line Scintillation Detector

Number Required: 1

Weight: ~ 5 lbs

Price: ~ \$400

RIDL Model 10-17 Scintillation Preamplifiers and Photomultiplier

Number Required: 2

Total Weight: ~ 12 lbs

Total Price: ~ \$500

RIDL Model 10-17 Scintillation Preamplifier

Number Required: 1

Weight: ~ 6 lbs

Price: \$110

TMC Model 404 Four Input Analyzer

Number Required: 1

Power Consumption: 36 watts

Weight: ~ 37 lbs

Price: \$12,975

TMC Model 560 Visual Readout Indicator

Number Required: 1

Weight: 2 lbs

Price. \$375

TMC Model 500 Paper Tape Printer

Number Required: 1

Power Consumption: 55 watts

Weight: ~ 33 lbs

Price: \$1,875

Tektronix Model 503 RM 5" Oscilloscope Model 171

Number Required: 1
Power Consumption: 107 watts
Weight: 27 lbs
Price: \$705

TMC Model 522 Spectrum Resolver-Integrator

Number Required: 1
Power Consumption: 19 watts
Weight: ~ 28 lbs
Price: \$2,400

System Totals for Gamma Spectroscopy

Power Consumption: 217 watts
Weight: 150 lbs
Price: \$19,340

ALPHA AND BETA DETECTOR ELECTRONIC COMPONENTS

RIDL Model 10-17 Scintillation Preamplifier and Photomultiplier

Number Required: 2
Total Weight: ~ 12 lbs
Total Price: \$500

RIDL Model 40-9B High Voltage Power Supply

Number Required: 2
Total Power Consumption: ~ 60 watts
Total Weight: 8 lbs
Total Price: \$890

RIDL Model 30-24 Dual Amplifier-Discriminator

Number Required: 1
Weight: ~ 8 lbs
Price \$624

RIDL Model 49-43 6 Decade Printing Preset Scaler

Number Required: 2
Total Power Consumption: 16.8 watts
Total Weight: 6.8 lbs
Total Price: \$1,590

RIDL Model 54-8 5 Decade Printing Electronic Timer

Number Required: 2
Total Power Consumption: 16.6 watts
Total Weight: 6.8 lbs
Total Price: \$1,390

RIDL Model 29-1 Instrument Case and Power Supply

Number Required: 2
Total Power Consumption: ~ 120 watts
Total Weight: 70 lbs
Total Price: \$390

System Totals for Alpha and Beta Detector Electronic Components

Power Consumption: ~ 214 watts
Weight: 112 lbs
Price: \$5,384

ELECTROPHORESIS AND PAPER CHROMATOGRAPHY EQUIPMENT

One Spinco Model R Paper Electrophoresis Apparatus and Auxiliary Equipment Set. (Aloe Scientific No. VSP 301911)

Power Consumption: ~ 50 watts
Weight: ~ 40 lbs
Price: \$2,182

One 10 x 18 inch Chromatography Assembly (Aloe Scientific No. V 29204 SB)

Weight: ~ 10 lbs
Price: \$139

Voltage Regulator (One Sorenson Model R 3010, 0-3000 VA at 110 V ac)

Weight: ~ 40 lbs

Price: \$622

System Totals for Electrophoresis and Paper Chromatography
Equipment

Power Consumption: ~ 50 watts

Weight: ~ 90 lbs

Price: \$2,943

Appendix C
DEVELOPMENTAL SYSTEMS

TENT STRUCTURE

Weight: ~ 130 lbs

Estimated Cost: ~ \$4000

AIR CONDITIONING AND MOTOR GENERATOR EQUIPMENT

Power Consumption: ~ 2 KW

Weight: ~ 350 lbs

Estimated Cost: ~ \$3,000

LABORATORY HARDWARE

Weight: ~ 100 lbs

Estimated Cost: \$2,000

REPACKAGING CONTAINERS (No. 10 Size)

Number: 2000

Weight: ~ 0.15 lbs/per container and lid

Weight of 1000 units: ~ 150 lbs

Estimated Cost: \$7,000

MISCELLANEOUS EQUIPMENT AND SUPPLIES

Weight: 100 lbs

Estimated Cost: \$5,000

TRANSPORT CONTAINER

Weight: ~ 500 lbs

Estimated Cost: \$5,000

CANISTER AND ELEVATOR PLATFORM SYSTEM

Power Consumption: ~ 150 watts

Weight: 204 lbs

Estimated Cost: \$23,000

DEVELOPMENTAL SYSTEMS TOTALS

Power Consumption: 4 Kilowatts

Weight: 1534 lbs

Estimated Cost: \$49,000

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13. ABSTRACT <p>A study of the laboratory requirements necessary for evaluating radioactive contamination of food, water and biological specimens has been completed. From the information obtained, a mobile laboratory facility with the capability of qualitative and quantitative analyses of millimicrocurie amounts of alpha, beta and gamma emitting radioisotope contaminants has been designed. The laboratory utilizes sufficient shielding so all analyses can be performed in a two milliroentgen per hour gamma radiation field and where the average gamma energy is one mev. The entire laboratory including all electronics, detectors, shielding, power supplies, motor generator, laboratory structure and hardware, and transport container weighs less than two thousand pounds and has a total power requirement (115 volts a.c. - 60 cycle) of four kilowatts. (Power is furnished by a self-contained gasoline driven motor generator.) The laboratory in its cubic transport container, six feet on a side, is thus readily transportable either by helicopter or truck.</p>			