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MANUAL OF BIOASSAY PROCEDURES FOR RADIONUCLIDES

MANUEL DE TECHNIQUES DE BIODOSAGE DES RADIONUCLIDES

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**Etablissement de recherches
nucléaires de Whiteshell**

Pinawa, Manitoba R0E 1L0

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RÉSUMÉ

On décrit un programme de surveillance permettant de mesurer la contamination interne, par les radionuclides, sur le lieu de travail, des travailleurs sous rayonnement nucléaire. Ce programme comporte des techniques analytiques pour la mesure de la radioactivité alpha, bêta et gamma dans les échantillons biologiques, généralement les échantillons d'urine. On identifie les radionuclides par leur rayonnement caractéristique, au moyen de détecteurs à scintillateurs liquides et de spectromètres alpha, bêta et gamma. On donne des exemples de calcul de la radioactivité minimale décelable pour des radionuclides particuliers et on se sert de ces exemples pour tirer la fréquence d'appel pour examen suivant laquelle les divers groupes de travailleurs sont soumis à un contrôle tous les mois.

l'Énergie Atomique du Canada, Limitée
Établissement de recherches nucléaires de Whiteshell
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ABSTRACT

A monitoring program is described by which atomic radiation workers are monitored for internal contamination with radionuclides in the workplace. The program involves analytical procedures for measuring alpha, beta and gamma activity in biological specimens, usually urine. Radionuclides are identified by their characteristic radiation using liquid scintillation counting, and alpha, beta and gamma spectrometry. Examples of calculating the minimum detectable activity for specific radionuclides are given and used to derive call-in-criteria in accordance with which the different groups of workers are monitored each month.

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1. INTRODUCTION

The Bioassay Laboratory at the Whiteshell Nuclear Research Establishment (WNRE) routinely monitors atomic radiation workers (ARW) for possible internal contamination with radionuclides. This monitoring program involves analytical procedures for measuring radioactivity in human excreta, such as urine.

Radionuclides are identified by their characteristic alpha, beta or gamma radiation, using equipment that is designed to detect these different types of radiation. However, most bioassay samples must be chemically processed before counting for radionuclide activity can be done reliably.

This manual describes various bioassay procedures with specific reference to

- (1) Methods for separating alpha, beta and gamma activities from human urine,
- (2) Methods for counting radionuclides in bioassay samples,
- (3) Calibration procedures,
- (4) The sensitivity of methods used,
- (5) The call-in-criteria and
- (6) The reporting of results.

2. METHODS FOR MEASURING ALPHA ACTIVITY

Alpha particles ($^4\text{He}_2$) are absorbed by small amounts of matter. Therefore, to measure alpha activity in urine, it must first be extracted quantitatively and deposited as a near weightless source on a clean stainless steel surface (planchet) of dimensions that fit the counting position in an alpha counter. Preparation of such weightless sources involves chemical procedures that vary with the nature of the alpha activity. The methods depend on specific program activities, which are

- (1) Screening for gross alpha activity,
- (2) Assay of natural uranium and thorium by neutron activation, and
- (3) Assay of plutonium, americium, uranium and thorium by electroplating.

2.1 PROCESSING OF URINE SAMPLES FOR GROSS ALPHA SCREENING MATERIALS

- (a) $0.9 \text{ mol} \cdot \text{dm}^{-3}$ reagent grade calcium phosphate standard (Fisher Scientific)
- (b) Reagent grade 16 mol/L HNO_3 and 15 mol/L NH_4OH (Fisher Scientific)
- (c) Reagent grade fuming HNO_3 (20 mol/L) (Fisher Scientific)
- (d) 100-200 mesh chromatographic grade silica gel (Sigma Chemical Co)
- (e) 2-cm diameter by 25-cm-long quartz glass column (WNRE glass blower)
- (f) 10-mL polystyrene pipettes (Fisher Scientific)
- (g) One-inch-diameter (nominal) stainless steel counting planchets (Fisher Scientific)

- (h) High-density polyethylene activation capsules (Urige Universiteit, Biologisch laboratorium de Boelelaan 1087, Amsterdam Buitenvelder, The Netherlands)
- (i) 1.3-cm³ transfer capsules machined from super-pure iron (WNRE Machine Shop)
- (j) Natural uranium standard, prepared by electroplating natural uranium onto a highly polished stainless steel disc
- (k) Uranium stock standard prepared by dissolving 1 mg uranyl nitrate (Research Organic/Inorganic Chemical Corp.) in 100 mL of 1 mol/L HNO₃
- (l) Thorium stock standard prepared by dissolving 1 mg of thorium nitrate (May and Baker) in 100 mL of 1 mol/L HNO₃.

Methods

- (1) Transfer 300 mL of urine to a 500-mL beaker.
- (2) One mL of calcium phosphate standard is added to coprecipitate the actinides.
- (3) Add 50 mL of 16 mol/L HNO₃ and heat on a hot plate at 80 to 90°C for 2 h to mineralize the urine.
- (4) Add 7 mol/L NH₄OH in sufficient quantity until precipitate starts to form. Heat an additional 30 min at 80 to 90°C.
- (5) Cool, decant, and then transfer the precipitate with ammonia wash to 150 mL centrifuge bottles.
- (6) Centrifuge for 5 min and transfer the precipitate with 5 mol/L HNO₃ to a 150-mL beaker.
- (7) Evaporate to dryness under a heat lamp and burn off the residual organics in a muffle furnace at 525°C for 5 min. If the residue is still discoloured, add 2 mL of fuming HNO₃ and heat for an additional 5 min at 525°C.
- (8) Cool and then add 100 mL of 1 mol/L HNO₃ to dissolve the precipitate.
- (9) Boil the 100 mL solution for 20 min. Use a Teflon stirring bar to prevent spattering. Place a Teflon cover plate over the beaker during boiling. The boiling is required to hydrolyze the phosphates [1].
- (10) Cool and make up to 200 mL with double-distilled water.
- (11) Transfer the 200 mL solution to a Teflon beaker and adjust the pH between 5 and 6 with 7 mol/L NH₄OH. The use of Teflon rather than glass beakers facilitates quantitative recovery by preventing errors due to adsorption of actinides to glass at near neutral solutions.
- (12) With a 10-mL pipette, pass the 200 mL solution through a column containing 6 g of chromatographic grade (100-200 mesh) silica gel supported on glass wool. Prewash the column with 5 mol/L HNO₃ and then with double-distilled water to neutral pH.
- (13) After the 200-mL sample is passed through the column, wash the column with two equivalent volumes of distilled water.
- (14) Elute the actinides with 45 mL of double-distilled 5 mol/L HNO₃ into a 50-mL plastic beaker.
- (15) Evaporate the eluate to dryness, dissolve with 0.5 mL of 5 mol/L HNO₃ and transfer either to planchets for direct counting, or to activation vials for neutron activation.
- (16) Samples transferred to planchets are dried under a heat lamp for 1 h and then counted for alpha activity.
- (17) Samples to be neutron activated are dried under a heat lamp for 2-3 h. These activation vials are heat sealed and then placed in transfer capsules for activation.

2.1.1 Description of Alpha-Beta Planchet Counter

The counter is a Model 1105 Spectro/Shield manufactured by Nuclear Chicago Corporation. It is comprised of: (a) an automatic planchet sampler changer for transferring samples to the counting chamber; (b) a sample changer controller that provides programming and control functions; (c) a gas flow sample detector for counting either alpha or beta activity; (d) a plastic scintillation guard detector for coincidence background rejection; (e) two single-channel analyzers, one for net sample counts and one for sample plus background counts; and (f) a PDP-5 computer for data storage and processing.

The counter is operated in Geiger mode for beta counting and in the proportional mode for alpha counting. For alpha counting, the gas used is 90% argon, mixed with 10% methane. For beta counting, the gas used is 95% helium mixed with 5% isobutane.

2.1.2 Calibration of Alpha-Beta Planchet Counter

Calibration of this counter with a standard beta source involves measuring the voltage plateau and establishing the correct operating voltage. Figure 1 shows that the operating voltage of 1100 V is within the range of the voltage plateau. A standard natural uranium source is used to check the alpha counting efficiency each time samples are being counted for alpha activity. This 4.7-Bq source gives a counting efficiency of 45%. Planchets are individually counted for a 24 h background before use. The planchet backgrounds vary from 0.1 to 0.2 counts per minute (cpm) over this time period. These background counts are subtracted from the gross count rates of samples.

2.2 NEUTRON ACTIVATION ANALYSIS OF URINE FOR NATURAL URANIUM AND THORIUM

The chemical extraction procedure is identical to that described in Section 2.1. The eluate from individual samples (step 16) is transferred to separate 0.5-mL polyethylene activation vials supplied by the Analytical Science Branch. The transferred eluates are dried and the vials sealed with heat. The vials are inserted into individual 1.3-cm³ transfer capsules. Personnel from the Analytical Science Branch do the neutron activation analysis of the samples. The sample is irradiated for 5 min at a neutron flux of 1×10^{14} n cm⁻²·s⁻¹. The vial temperature over this 5-min irradiation time is less than 80°C.

Four samples can be neutron activated simultaneously, one of which is usually a standard. After activation the samples are collected immediately and counted with a gamma spectrometer (described later in Section 4.1 of this report) for neutron activation products from natural uranium and thorium. Table 1 shows the expected yield and energies of the gamma activity from a 5-min irradiation of 1 µg each of natural uranium and thorium.

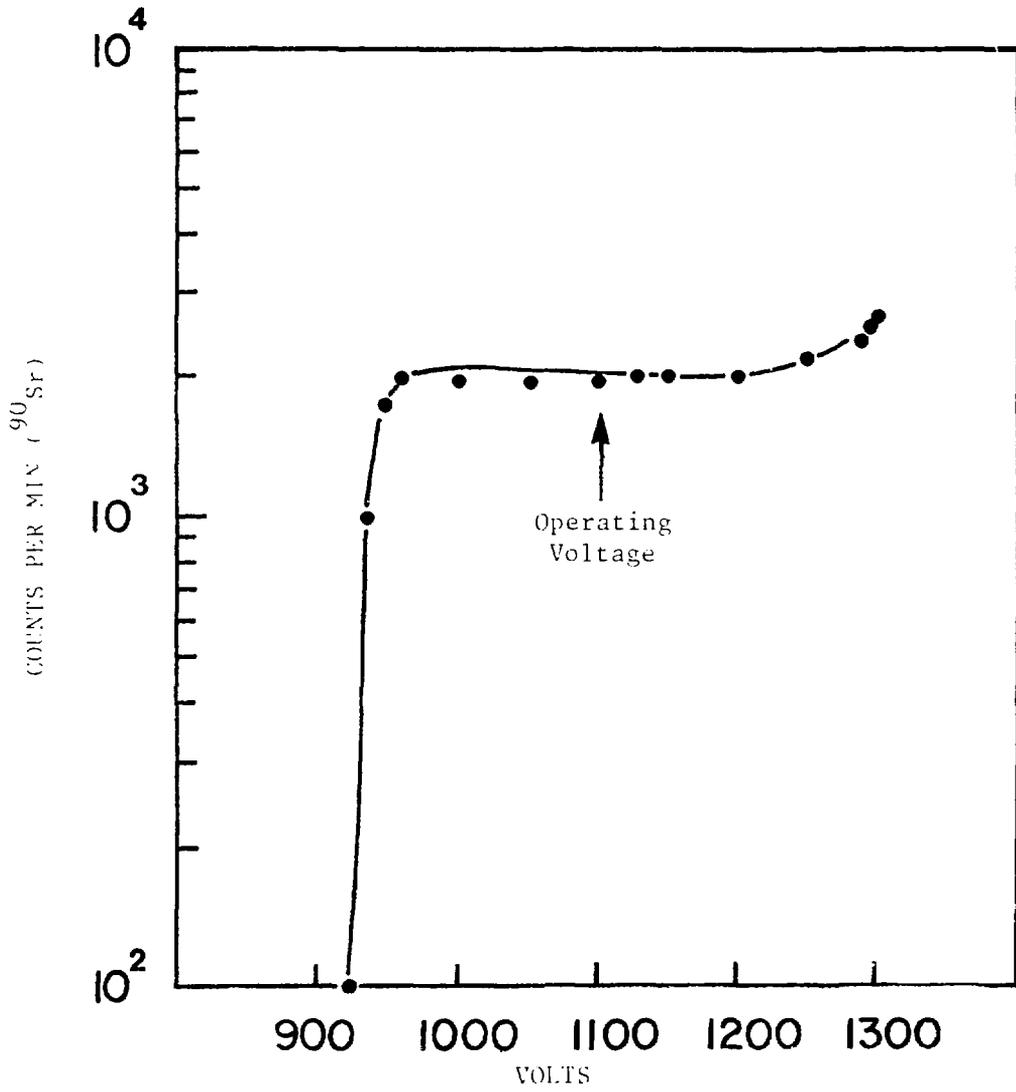


FIGURE 1: Voltage Plateau for Beta Counting

TABLE 1
AMOUNT OF ^{239}U AND ^{233}Th PRODUCED FROM A 5-MIN IRRADIATION OF
1 μg EACH OF NATURAL URANIUM AND THORIUM

Reaction	Energy (keV)	Abundance (%)	T 1/2 (min)	$\gamma\text{s}^{-1} \cdot \mu\text{g}^{-1}$
$^{238}\text{U} (n, \gamma) ^{239}\text{U}$	74.7	51.0	23.5	4.83×10^4
$^{232}\text{Th} (n, \gamma) ^{233}\text{Th}$	86.9	2.7	22.2	7.49×10^3

Figure 2 shows 3 spectra obtained from a 3-min count of neutron-activated urine samples, counted within 10 min after activation. Spectrum A is from 100 mL of a control specimen. Spectrum B is from 100 mL of the control specimen spiked with 2 μg each of thorium and uranium standard. Spectrum C is from 2 μg of the thorium and uranium standard.

The 74.7 and 86.9 keV peaks are well resolved and can be integrated for quantitative measurement after correcting for decay over the time period from immediately after irradiation to beginning of counting. Samples are always activated concurrently with known standards so that a quantitative estimate of the actinide(s) present in a sample can be calculated by comparing it to the known standard.

2.3 ACTINIDE ANALYSIS OF URINE BY ELECTROPLATING

Electroplating is a procedure where the actinides are plated on to a highly polished stainless steel disc and then counted with an alpha spectrometer. The plated source produces a high resolution spectrum by virtue of its weightlessness. This permits identification of the energy of the alpha particle and allows reliable measurements to be made, provided the alpha activity is quantitatively extracted beforehand.

The following procedure is used to prepare a urine sample for electroplating:

Materials

- (a) Hydrochloric acid (HCl), reagent grade, Fisher Scientific
- (b) Nitric Acid (HNO_3), reagent grade, Fisher Scientific
- (c) Hydrogen peroxide (H_2O_2), reagent grade, Fisher Scientific
- (d) 2.5-cm-diameter x 28-cm-long glass column
- (e) AG 1-X8 ion exchange resin, 50-100 mesh, analytical grade, Bio Rad Laboratory
- (f) Reagent containing 1 mol/L HNO_3 in 93% methanol, reagent grade, Fisher Scientific

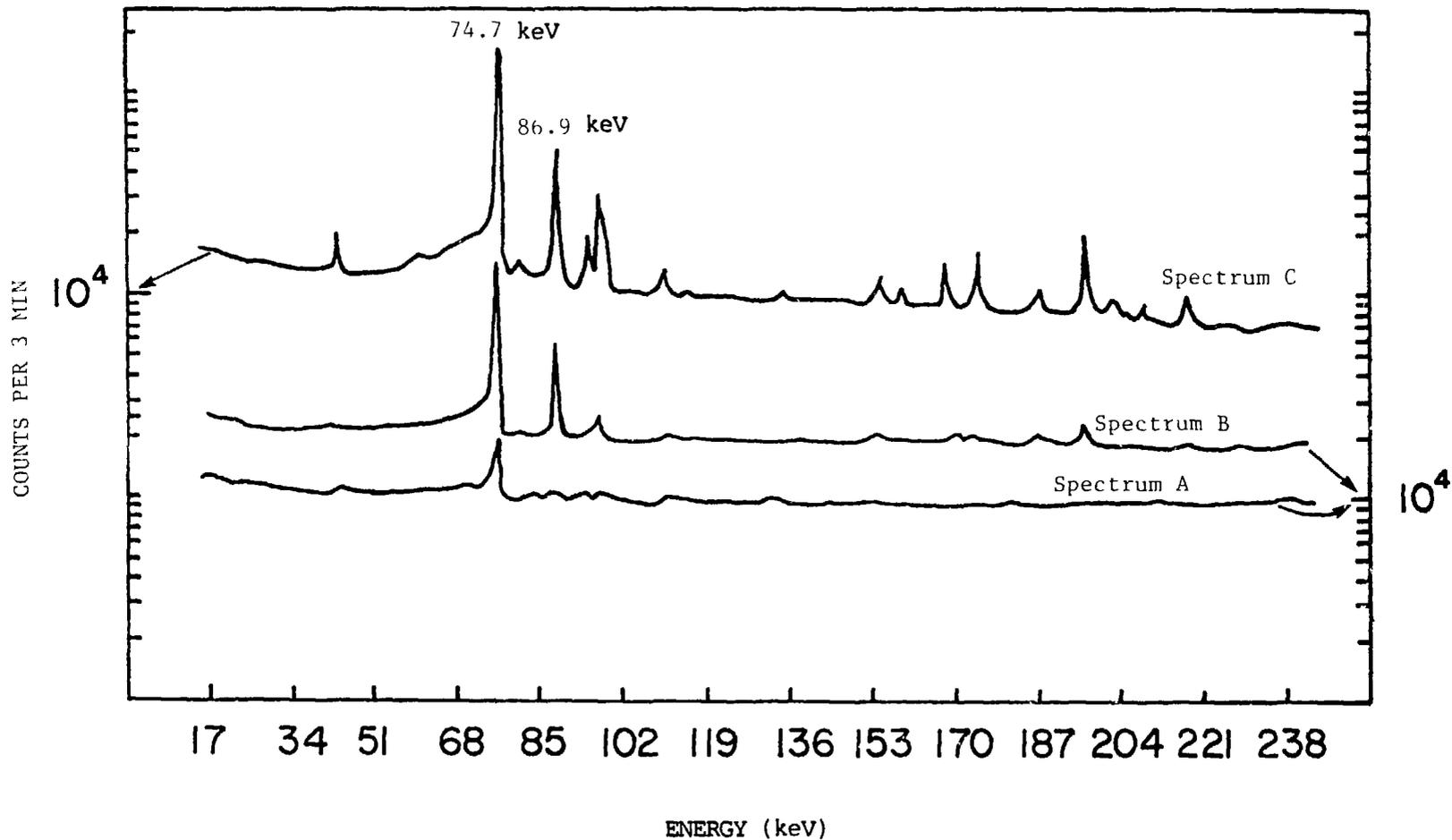
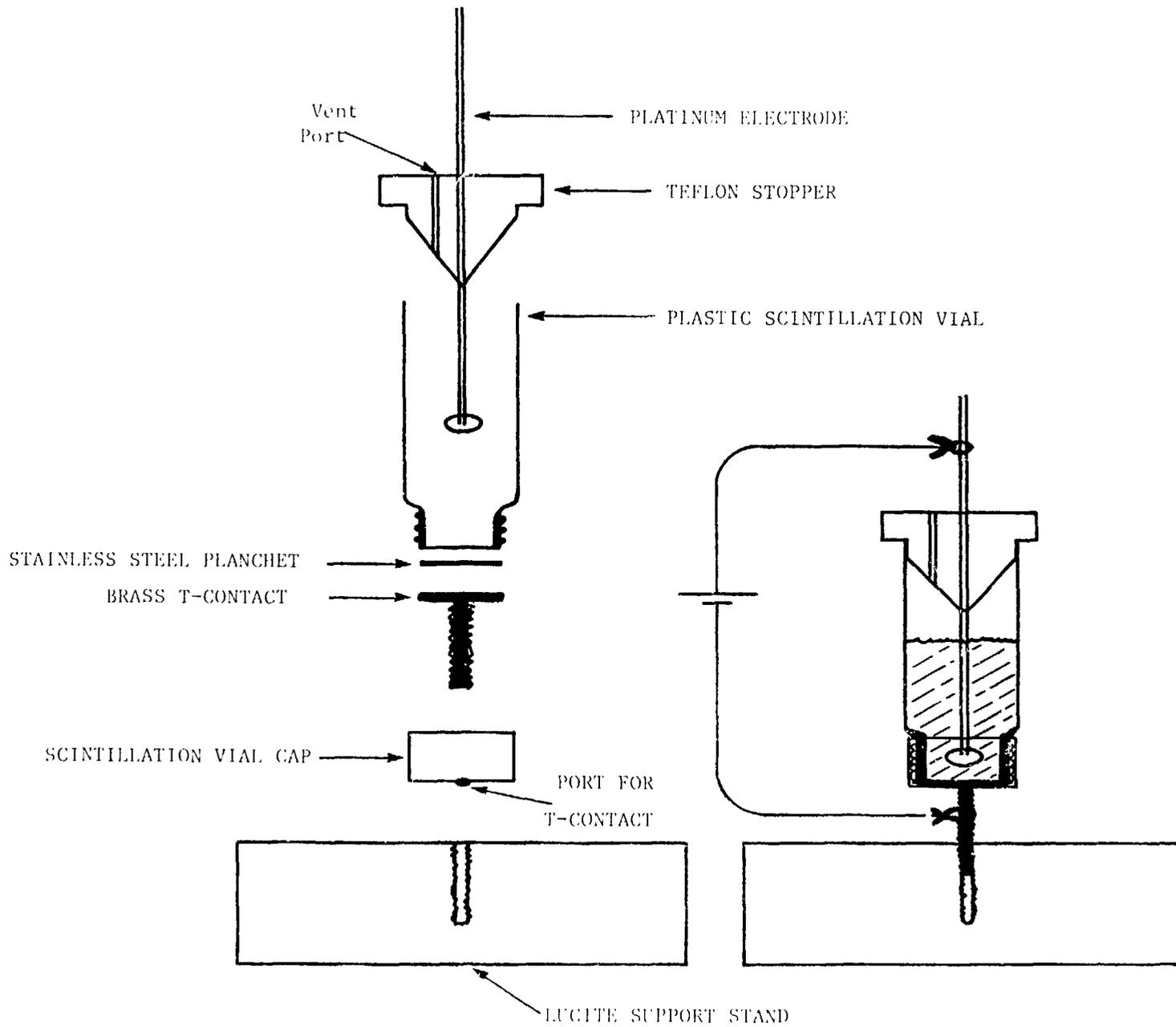


FIGURE 2: Spectra After a 3-min Count of Neutron-Activated Urine Samples, Counted Within 10 Min of the Activation. Spectrum A is from 100 mL of the control specimen. Specimen B is from 100 mL of the control specimen spiked with 2 μ g each of a Th and U standard. Specimen C is from 2 μ g each of the Th and U standard.



- (g) Reagent containing 1.5 mol/L HCl in 86% methanol
- (h) Electroplating cell (Figure 3) made from a 20-mL plastic vial. The cathode is made from a 1.9-cm-diameter highly polished stainless steel disc and the anode is made from a 1-mm diameter platinum wire. A 7-V power source (Acopian, Model K76200) supplies 1 A of current for the electrolysis process.
- (i) Sulphuric acid (H_2SO_4), analytical grade, J.T. Baker Chemical Company
- (j) Thymol blue, 0.05% solution, Fisher Scientific
- (k) Ammonia gas (lecture bottle), Canadian Liquid Air Ltd.
- (l) Ammonium hydroxide (NH_4OH) (reagent grade), Fisher Scientific
- (m) Solution containing $0.13 \text{ mol} \cdot \text{dm}^{-3}$ ammonium nitrate (NH_4NO_3) (analytical grade), Fisher Scientific, in 0.15 mol/L ammonium hydroxide (NH_4OH)
- (n) Uranium-232 standard in 1 mol/L HNO_3 (0.1 Bq/mL), New England Nuclear

Methods

- (1) The urine is mineralized according to the procedure outlined in steps 1-7 of Section 2.1. To check for total uranium recovery, 50 μL of the ^{232}U standard is added to the urine before mineralization.
- (2) The white salts are dissolved in 100 mL of 9 mol/L HCl and 10 drops of H_2O_2 are added.
- (3) Heat at 80-85°C for 1 h. Cool to room temperature and at this point, the solution is ready for separation on an ion exchange chromatography column [2,3].
- (4) Prepare the chromatography column by supporting 20 mL AG 1-8X resin on a glass wool in a 2.5-cm-diameter x 28-cm-long glass column.
- (5) Wash the column with 100 mL 9 mol/L HCl containing 1 drop of 30% H_2O_2 .
- (6) Add the sample from step 2 to the column.
- (7) Wash the column with 50 mL 9 mol/L HCl. The eluent will contain thorium, americium and curium.
- (8) Elute the uranium with 200 mL of 7.2-mol/L HNO_3 . Evaporate the eluent to dryness and proceed with the electroplating procedure (Section 2.3.1).
- (9) Wash the column with 5 mL of 1.2 mol/L HCl and elute the plutonium with 200 mL 1.2 mol/L HCl containing 2 mL of 30% H_2O_2 . Evaporate the eluent to dryness and proceed with electroplating procedure (Section 2.3.1).
- (10) The eluent from step 6 containing thorium, curium, and americium is evaporated to dryness and taken up in 100 mL of 7-8 mol/L HNO_3 . The column is washed with 100 mL of 7-8 HNO_3 and the sample is put through the column. The column is washed with 100 mL of 7 to 8 mol/L HNO_3 . The eluent contains the Am and Cm. Thorium is eluted with 100 mL of 9.0 mol/L HCl. The thorium eluent is dried and is then ready for the electroplating procedure (Section 2.3.1).
- (11) The eluent containing the americium and curium is dried and taken up in 100 mL of 1 mol/L HNO_3 /93% Cu_3OH . The sample is then put through the column and the curium and americium are eluted with 100 mL of 1.5-mol/L HCl/86% CH_3OH . The eluent containing the americium and curium eluent is dried and ready for the electroplating procedure (Section 2.3.1).

2.3.1 Electroplating Procedure

The electroplating procedure is as follows:

- (1) The dried actinide samples are quantitatively transferred with 5 mol/L HNO_3 to 5-mL pyrex beakers.
- (2) Add 15 drops of concentrated $[\text{H}_2\text{SO}_4]$.
- (3) Slowly evaporate to approximately 0.5 mL.
- (4) Cool the sample and add 3 mL distilled H_2O .
- (5) Add 2 drops 0.05% thymol blue and expose the sample to NH_4 gas until it turns a salmon-pink or straw-yellow colour. This indicates a pH of 2.5.
- (6) Pour the sample into the electroplating cell, rinsing with 0.1 mol/L H_2SO_4 solution.
- (7) Again expose the sample to NH_4 until color changes to pH 2.5.
- (8) Electroplate for 1 h at 7 V and 1 A of current.
- (9) Add 10 mL of 1.5 mol/L NH_4OH to the cell solution and continue electroplating for a further 1 min. Disconnect sample from power supply.
- (10) Flush cell with 1% NH_4NO_3 /0.15 mol/L NH_4OH .
- (11) Remove disc and rinse with 5 mL of a solution made up of 1 L $\text{C}_2\text{H}_5\text{OH}$ plus 4 drops NH_4OH . The disc is dried and ready for counting.

Figure 4 shows a spectrum obtained by spiking 300 mL of urine with 0.35 Bq of natural uranium, 0.20 Bq of ^{239}Pu and 0.02 Bq of ^{241}Am . The urine was processed and electroplated as per the above procedures and counted with a 450-mm²-surface barrier detector (see Section 2.3.2).

2.3.2 Surface Barrier Detector

Figure 5 identifies the components of the alpha spectrometry system.

The weightless source produced by electroplating is placed 50 mm beneath the surface barrier detector, both of which are housed in an evacuated chamber. This combination of a weightless source and vacuum allows the emitted alpha particles to interact with the detector with very little energy degradation. The detector response produces a signal of size that is proportional to the energy of the incident alpha particles. Therefore, spectrometric measurements of alpha-emitting isotopes can be made with the associated electronics.

2.3.3 Calibration

The energy per channel (see Section 4.1.1 for description of the multichannel analyzer) is set by using a weightless standard containing approximately 550 Bq each of ^{239}Pu , ^{241}Am and ^{244}Cm . A 1000-channel energy region is used for alpha counting. The channel locations of the alpha peaks from these three isotopes are given in Table 2. This energy calibration is equivalent to 2.7 keV/channel, which yields a sufficiently good separation of most of the alpha emitters. Figure 6 shows the spectra of these three isotopes.

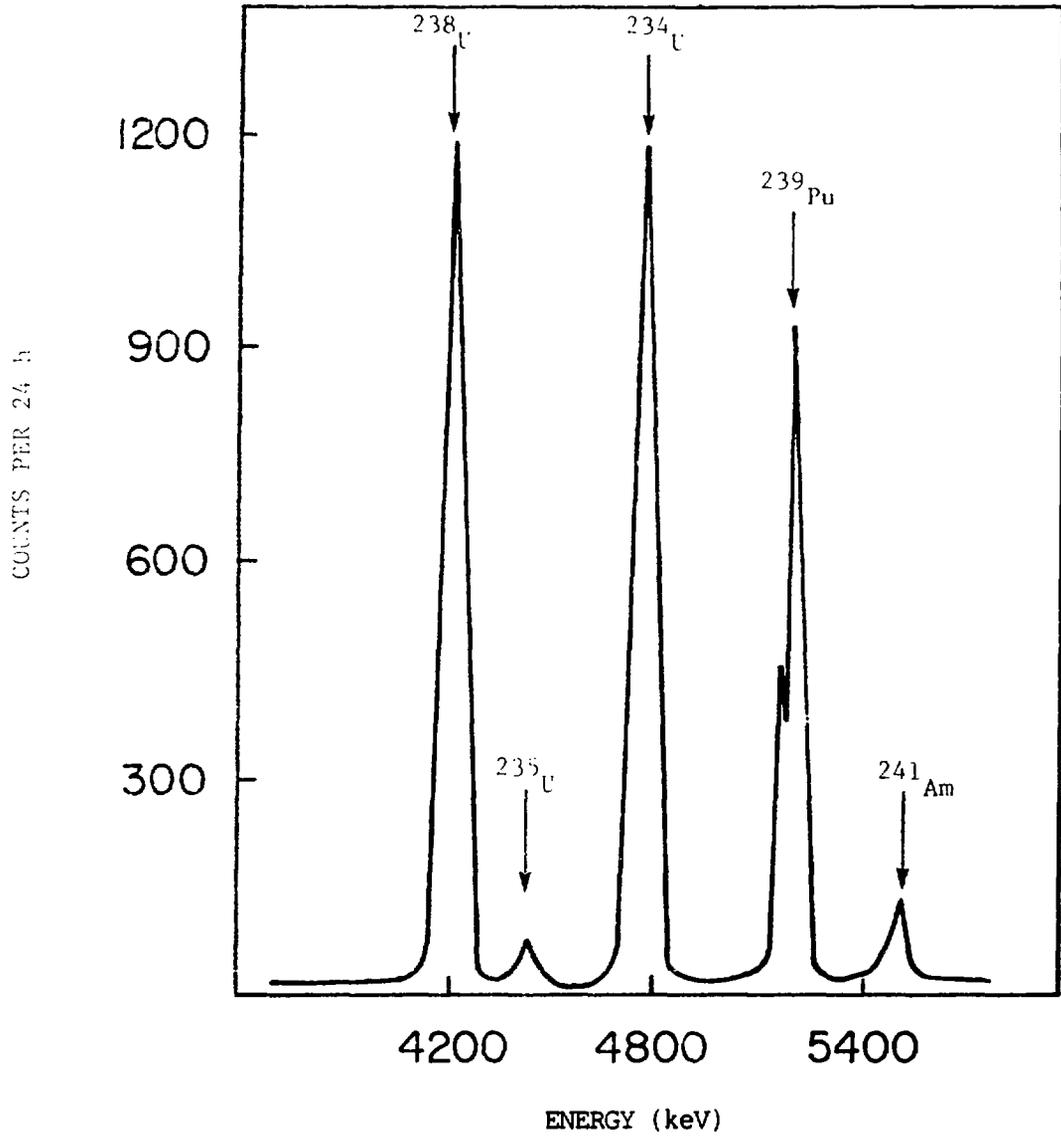


FIGURE 4: Electroplated Natural Uranium, ^{239}Pu and ^{241}Am from a Spiked Urine Sample

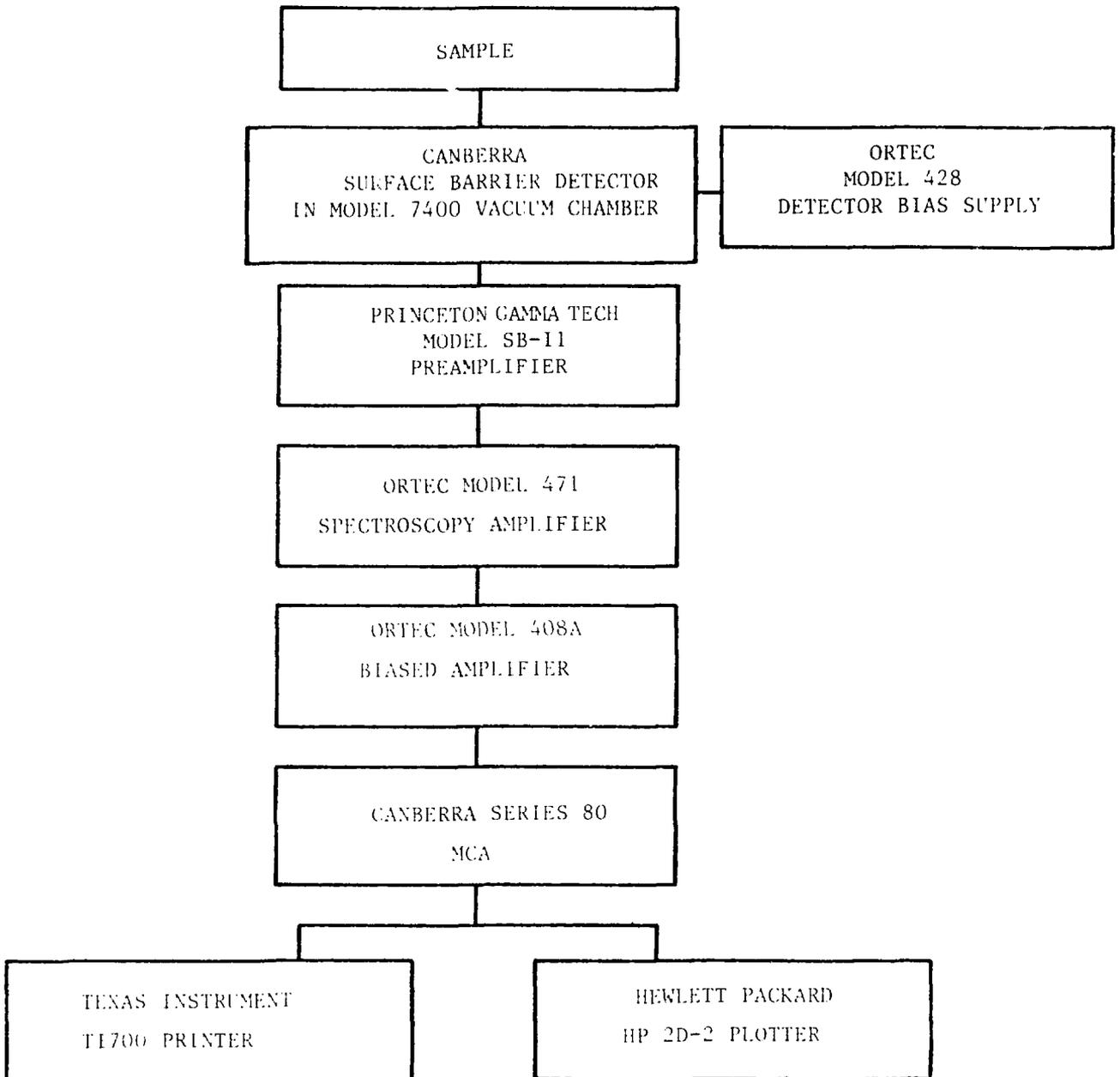


FIGURE 5: Alpha Spectroscopy System

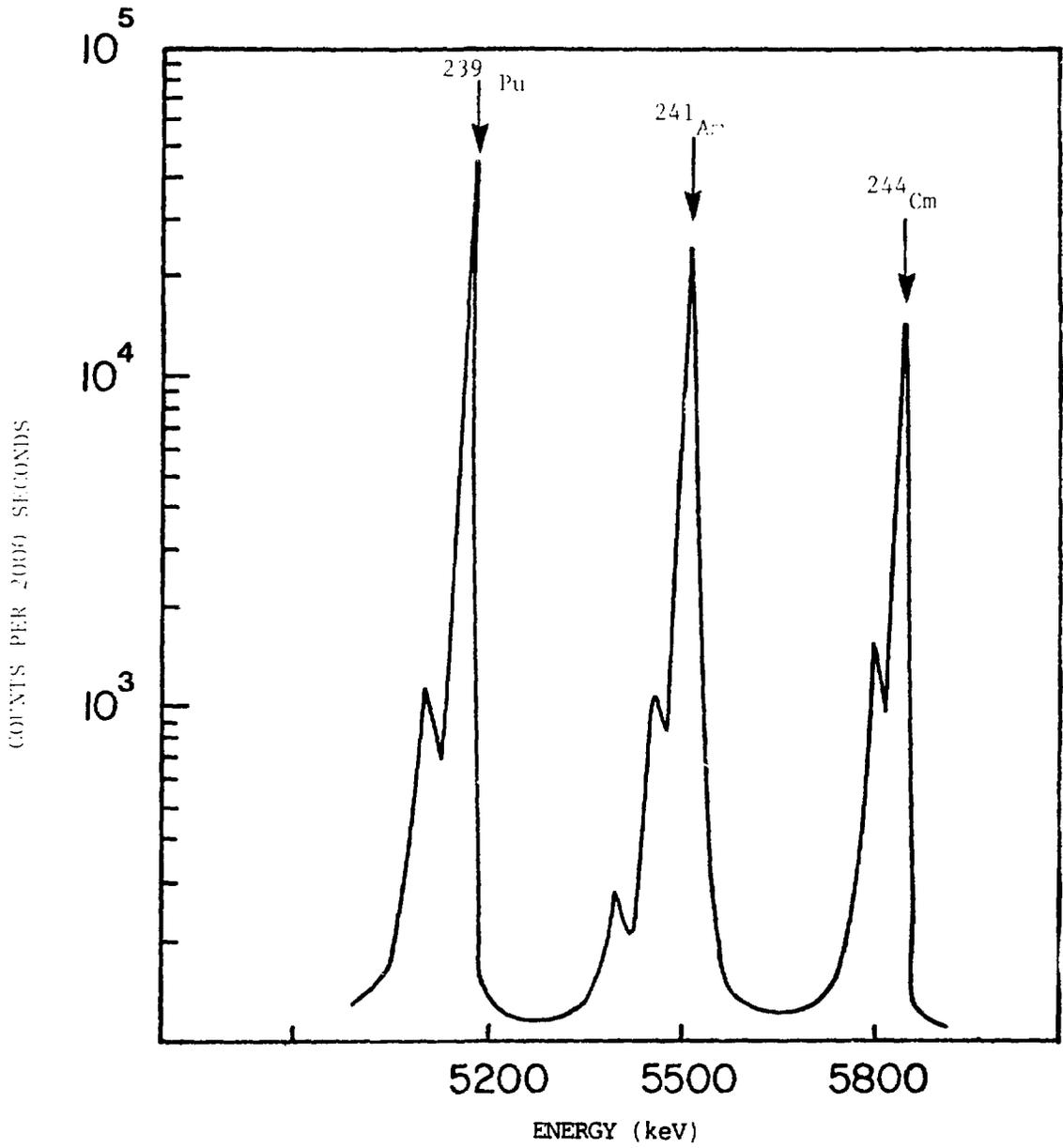


FIGURE 6: Alpha Energy Calibration Standard

TABLE 2

CHANNEL LOCATION FOR ^{239}Pu , ^{241}Am AND ^{244}Cm ALPHA PEAKS

Isotope	Energy (keV)	Channel Number of Alpha Peak
^{239}Pu	5157	331.4
^{241}Am	5486	454.8
^{244}Cm	5806	571.6

The resolution of alpha peaks by the surface barrier detector is recorded automatically by the Canberra Series 80 microprocessor. The energy calibration is done before every sample count. Typically, the resolution of the detector at full width, half maximum (FWHM) is 25 keV.

The counting efficiency of the detector increases as the source to detector distance decreases. Although the highest efficiency occurs when the sample is nearest the detector, this close distance can produce recoil contamination of the detector by some radionuclides [4]. In cases where recoil contamination is likely, the source is placed at a distance of 1.5 cm from the detector. The data in Table 3 and Figure 7 illustrate how the counting efficiency changes with distance for ^{238}Pu (1.04×10^4 Bq) alpha particles.

TABLE 3

DISTANCE BETWEEN SOURCE AND DETECTOR VERSUS EFFICIENCY FOR ^{238}Pu

Distance from face of detector (cm)	cps	Counting Efficiency %
0.5	2559.4	24.5
1.0	1368.3	13.1
1.5	780.9	7.5
2.0	506.8	4.9

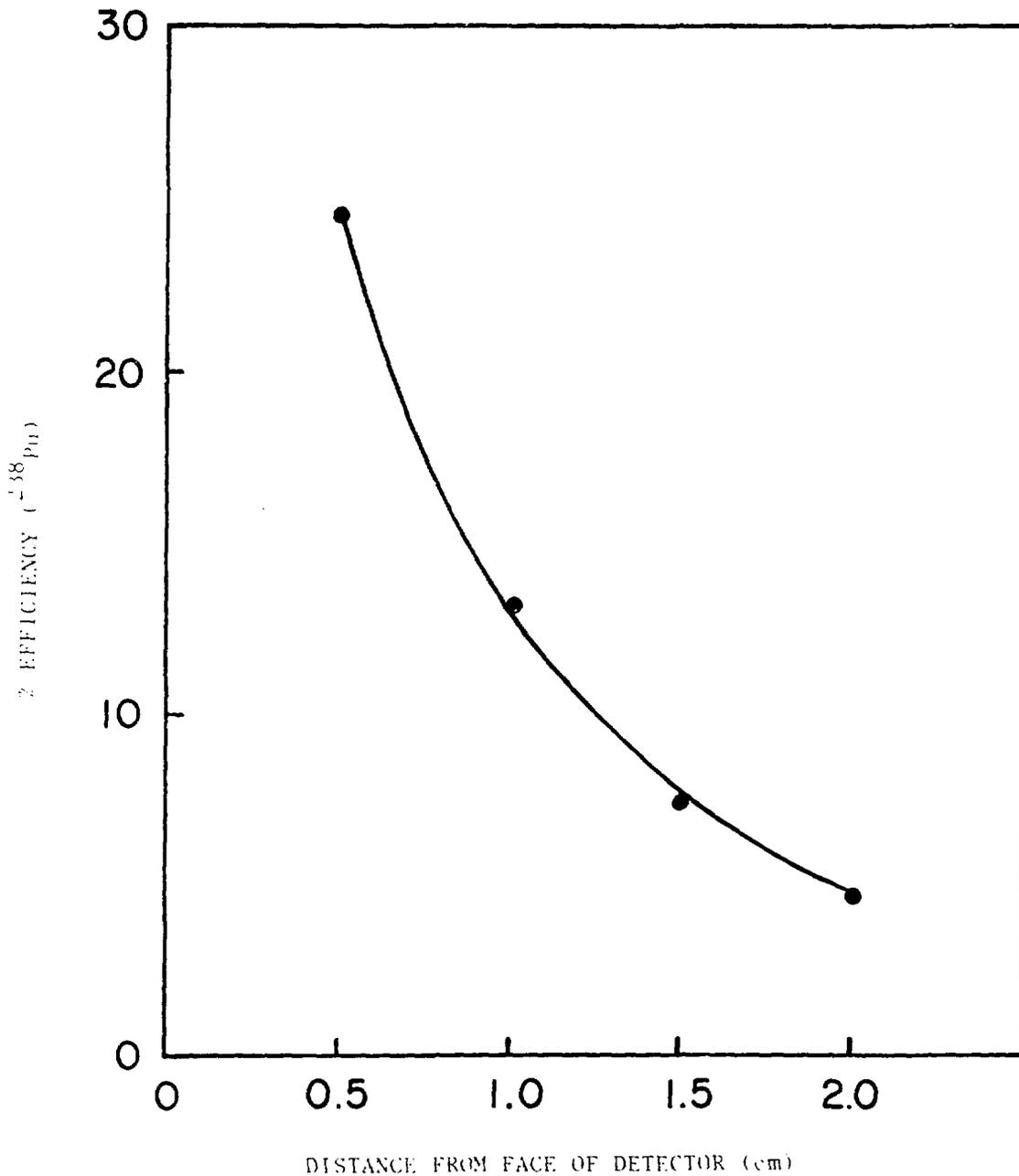


FIGURE 7: Alpha Counting Efficiency Versus Distance Between Source and Detector

The recovery efficiency is a measure of the amount of unknown radionuclide extracted from the sample and is determined by spiking a sample with a known amount of standard. For the gross alpha screening and neutron activation procedures, the recoveries for natural thorium and uranium are $80 \pm 10\%$ and $80 \pm 20\%$, respectively [10]. For the electroplating procedure, precise recoveries for each sample can be determined by spiking the sample with appropriate internal standards [2]. Typically, internal standards such as ^{236}Pu , ^{230}Th and ^{232}U are used for the respective recovery determinations of ^{238}Pu and ^{239}Pu , natural thorium and natural uranium.

2.4 MINIMUM DETECTABLE ACTIVITY (MDA)

The MDA [4] can be calculated by knowing the true average background and the sample detection sensitivity (S) for a given detection system.

The minimum detectable activity (MDA) at the 95% confidence level is given by

$$\text{MDA} = 2(\text{BG})^{1/2}/\text{S} \quad (1)$$

BG is the total background count and S is the detection sensitivity, which includes counting and recovery efficiencies.

(a) MDA for Gross Alpha Screening

For a background of 0.1 cpm over a 24-h counting period, a 45% counting efficiency and 80% sample recovery, the minimum detectable activity is given by

$$\text{MDA} = 2 \frac{(0.1 \times 24 \times 60)^{1/2}}{0.45 \times 0.8} = 66.7 \text{ alphas/24 h or } 0.05 \text{ alphas/min}$$

(b) MDA for the Electroplating Procedure

For a background of 0.01 cpm over a 24-h counting period, 20% counting efficiency and 80% sample recovery, the minimum detectable activity by the electroplating procedure is given by

$$\text{MDA} = 2 \frac{(0.01 \times 24 \times 60)^{1/2}}{0.2 \times 0.8} = 47.43 \text{ alphas/24 h or } 0.03$$

alphas/min

(c) MDA for Neutron Activation Analysis

A precise, generally applicable value is not calculable since the background changes with each sample that is activated. However, from Figure 2, it can be seen that this technique is sensitive enough to measure urinary-uranium levels that result from natural amounts in the body.

3. BETA DETECTION

Some radionuclides such as ^{90}Sr and ^3H are pure beta emitters, while others decay predominantly by beta emission. The presence of such radionuclides in urine samples is assayed by measuring the beta radiation directly or in chemically extracted precipitates.

The procedure for doing gross beta screening and tritium analyses will now be described.

3.1 SCREENING SAMPLES FOR GROSS BETA ACTIVITY

In general, this procedure involves coprecipitation of the alkaline earth elements, plating the precipitated samples on to planchets and then counting the samples in a beta counter.

Employees submit urine samples in 1-L plastic bottles. Generally, the urine is collected either during working hours or overnight. The volume of urine submitted is usually about 500 mL.

The step by step procedure for screening urine samples for gross beta activity is as follows:

Materials

(a) Sulkovitch Reagent

Preparation:

Equal 50-g weights of ammonium oxalate (Reagent grade, Fisher Scientific) and oxalic acid (Reagent grade, Fisher Scientific) are dissolved in 1200 mL of distilled water. Heat to 50-60°C for complete dissolution and add 100 mL of glacial acetic acid (Reagent grade, Fisher Scientific). Add distilled water to make up to 1500 mL.

- (b) One-inch-diameter (nominal) Stainless Steel Planchets, Fisher Scientific
- (c) Ammonia wash solution (0.15 mol/L NH_4OH), Fisher Scientific
- (d) Centrifuge, IEC size 2, Model K, Van Waters: Rogers, Inc.
- (e) Conical centrifuge tubes (40 mL), Fisher Scientific
- (f) Vortex Mixer, Fisher Scientific
- (g) Heat lamp with two 250-W lamps, Fisher Scientific
- (h) Air stirrers, Fisher Scientific
- (i) Plastic bottles (1 L), Fisher Scientific
- (j) Strontium nitrate ($^{90}\text{Sr}(\text{NO}_3)_2$) in 1 mol/L HNO_3 (3.7 kBq/mL), Amersham Corporation.

Methods

- (1) The submitted samples are identified and labelled. A 20-mL aliquot is taken for gamma counting and also for measuring of creatinine, if necessary.
- (2) The total volume of the submitted urine sample is measured and recorded.
- (3) An aliquot of Sulkovitch Reagent, equivalent to 10% of the urine volume, is added to the urine to precipitate alkaline earth elements.

This reagent is mixed with the urine for 15 min with an air stirrer. Ten such samples can be done simultaneously.

- (4) After 15 min of stirring, the samples are removed while carefully washing down the stirrers into the urine with 0.15 mol/L NH_4OH .
- (5) The samples are then centrifuged at 2500 RPM for 15 min using an IEC size 2 Model K centrifuge.
- (6) The supernatant is discarded and the precipitate is transferred with 0.15 mol/L NH_4OH to 40-mL conical centrifuge tubes.
- (7) The precipitates are again centrifuged at 3500 RPM for 5 min using an O.H. Johns Minor centrifuge.
- (8) The supernatant formed is discarded and the nonwhite precipitate is washed with 0.15 mol/L NH_4OH , vortexed and centrifuged again at 3500 RPM for 5 min.
- (9) The precipitate is made into a slurry with 0.15 mol/L NH_4OH by vortex mixing, and then plated out onto one-inch-diameter (nominal) stainless steel planchets.
- (10) The precipitate is dried for a half hour under a heat lamp at 70°C and then counted in the alpha-beta planchet counter for gross beta activity.
- (11) The recovery efficiency of the alkaline earth element(s) is determined by spiking a urine sample with 2 Bq of ^{90}Sr and extracting it as outlined in the above methods.

3.1.1 Creatinine Measurement

The amount of radionuclide activity excreted in urine is expressed on the basis of the daily urinary volume. The ideal volume of urine with which to do the analysis is on a 24-h sample, generally 1.4 L for males and 1.0 L for females [11]. However, for convenience of ARW, the usual practice is to submit smaller samples that can be collected overnight or during work. The fractional daily excretion, represented by the volume of urine submitted, is best estimated from the amount of creatinine present in the sample, expressed as a fraction of the daily amount of creatinine excreted by an individual [5].

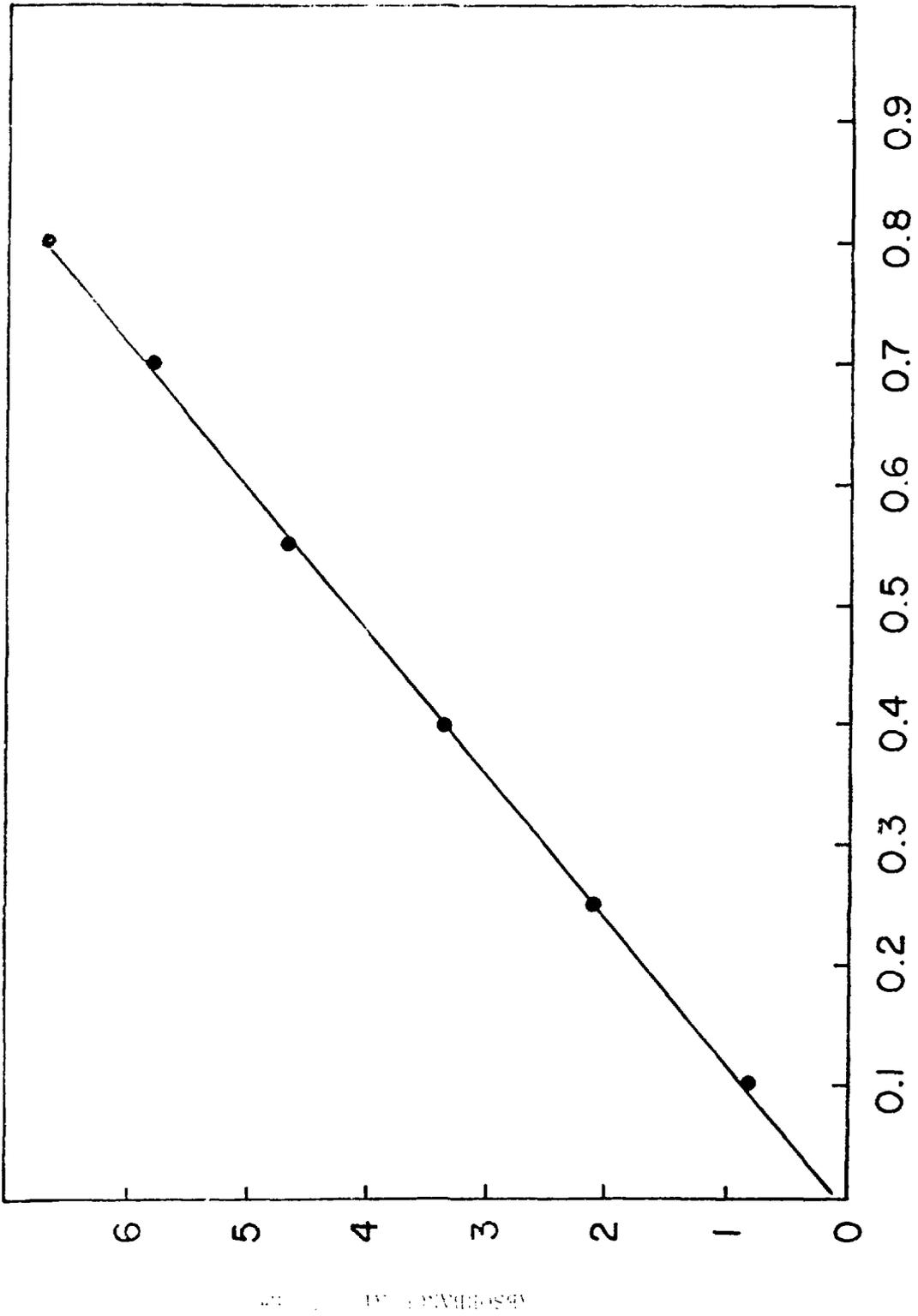
The procedure for measuring creatinine in urine is as follows:

Materials

- (a) Sodium Hydroxide (NaOH), reagent grade, Fisher Scientific
- (b) Picric Acid, reagent grade, Fisher Scientific
- (c) Creatinine, reagent grade, Matheson, Coleman and Bell
- (d) Spectrophotometer, Carey 15, Applied Physics Corporation

Methods

- (1) Prepare a creatinine standard at 1 mg/mL.
- (2) Place 300 μL each of: urine sample, distilled water, and creatinine standard into 3 separate 100 mL volumetric flasks.
- (3) Add 5 mL of a saturated solution of picric acid to each flask.
- (4) At known intervals of between 15 to 30 s, add 1 mL of 1 mol/L NaOH to each flask.
- (5) After 10 min have elapsed, the solutions are made up to 100 mL with distilled water and shaken thoroughly to ensure even mixing.



CREATININE (mg/dl)

FIGURE 8: Creatinine Concentration Versus Absorbance

- (6) Five minutes after mixing, the absorbance at 515 nm is measured in the Carey 15 spectrophotometer.
- (7) The concentration of the sample is obtained by comparing its absorbance at 515 nm with that of the standard.

Measurements have shown that, in the standard, the creatinine concentration versus absorbance is linearly related (Figure 8).

The following example illustrates a typical calculation to determine a person's daily creatinine excretion:

Absorbance reading from 300 μ L of urine = 3.4.
From Figure 8, an absorbance of 3.4 is equivalent to 0.4 mg creatinine/300 μ L.
Daily urine excretion = 1400 μ L.
Therefore:
Daily creatinine excretion = $\frac{0.4 \text{ mg} \times 1400 \text{ mL}}{0.3 \text{ mL}}$
= 1866 mg

The creatinine concentration in a urine sample is useful in several ways:

- (1) When a follow-up of an internal contamination occurs, several 24-h urine samples are submitted, from which a true daily creatinine excretion rate for the individual is derived. Subsequent measurements of the daily creatinine excretion, based on the fractional daily urine volumes, can be compared with the actual 24-h value. Conversely, the creatinine content in a sample, representing a fractional daily urine volume, is used to estimate the daily urine volume. The latter value is useful in calculating the daily excretion rate of radionuclides by individuals.
- (2) For screening purposes, the creatinine in a sample representing a fractional daily urine volume is compared with the established mean value [11] of 1.7 g/day for males and 1.0 g/day for females. Creatinine determination is also a check for abnormal characteristics.
- (3) The amount of creatinine excreted daily can be correlated with the individual's total body potassium as measured with the whole body counter [12]. Figure 9 shows the curvilinear relationship between the daily creatinine excretion versus total body potassium for a number of employees at WNRE.

3.1.2 Counter Description

The operation of this counter is described in Section 2.1.1 of this report. The Geiger Mueller mode is used for beta counting.

3.1.3 Calibration

The counter efficiency for a weightless $^{90}\text{Sr}/^{90}\text{Y}$ calibration source is 43%. Due to self-absorption in the precipitate, the efficiency decreases as the weight of the precipitate in the planchet increases. To illustrate how the efficiency varies with precipitate weight, known amounts of KCl can be added to a planchet and the beta activity from ^{40}K counted.

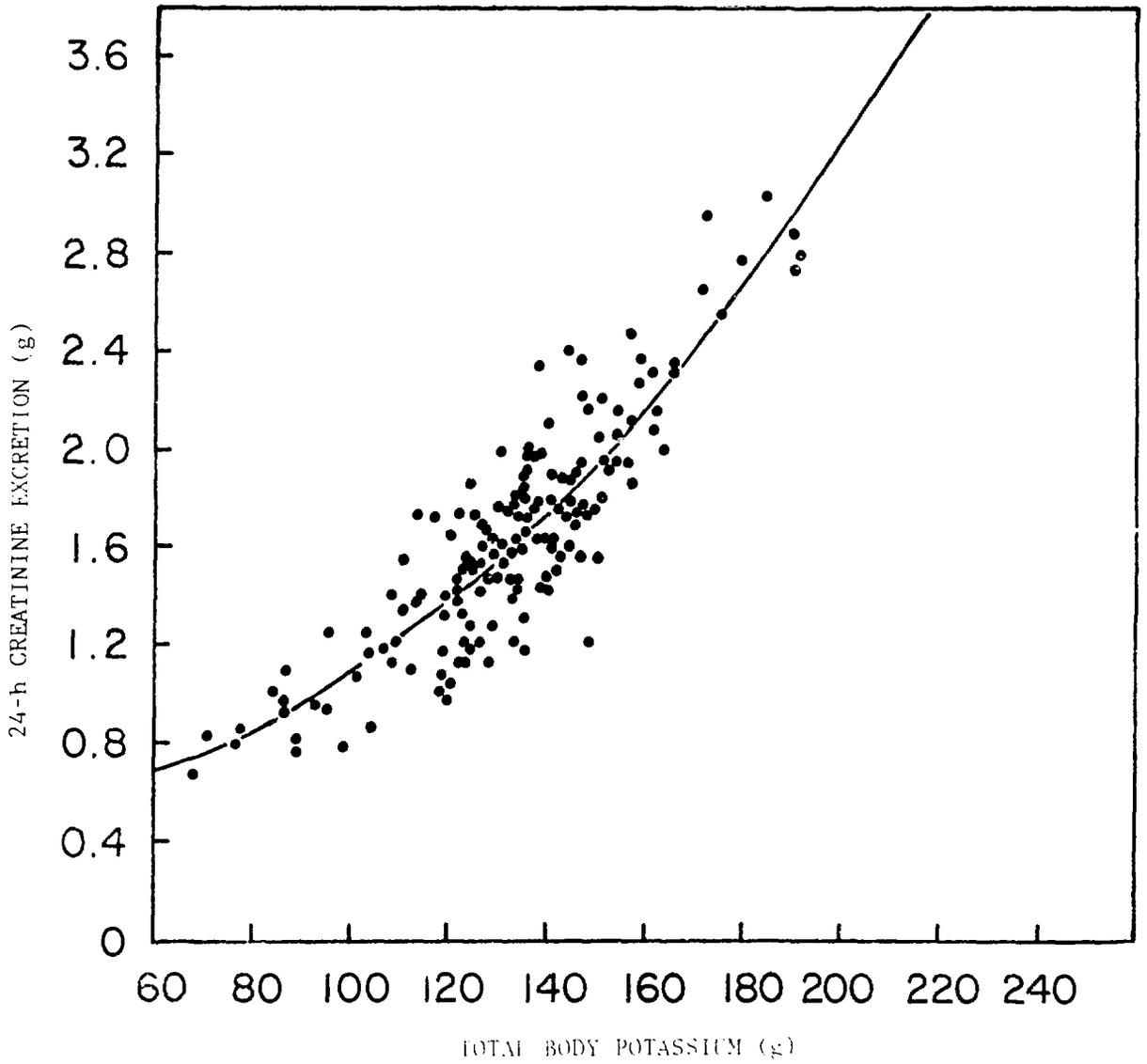


FIGURE 9: Daily Urinary Creatinine Excretion Versus Total Body Potassium

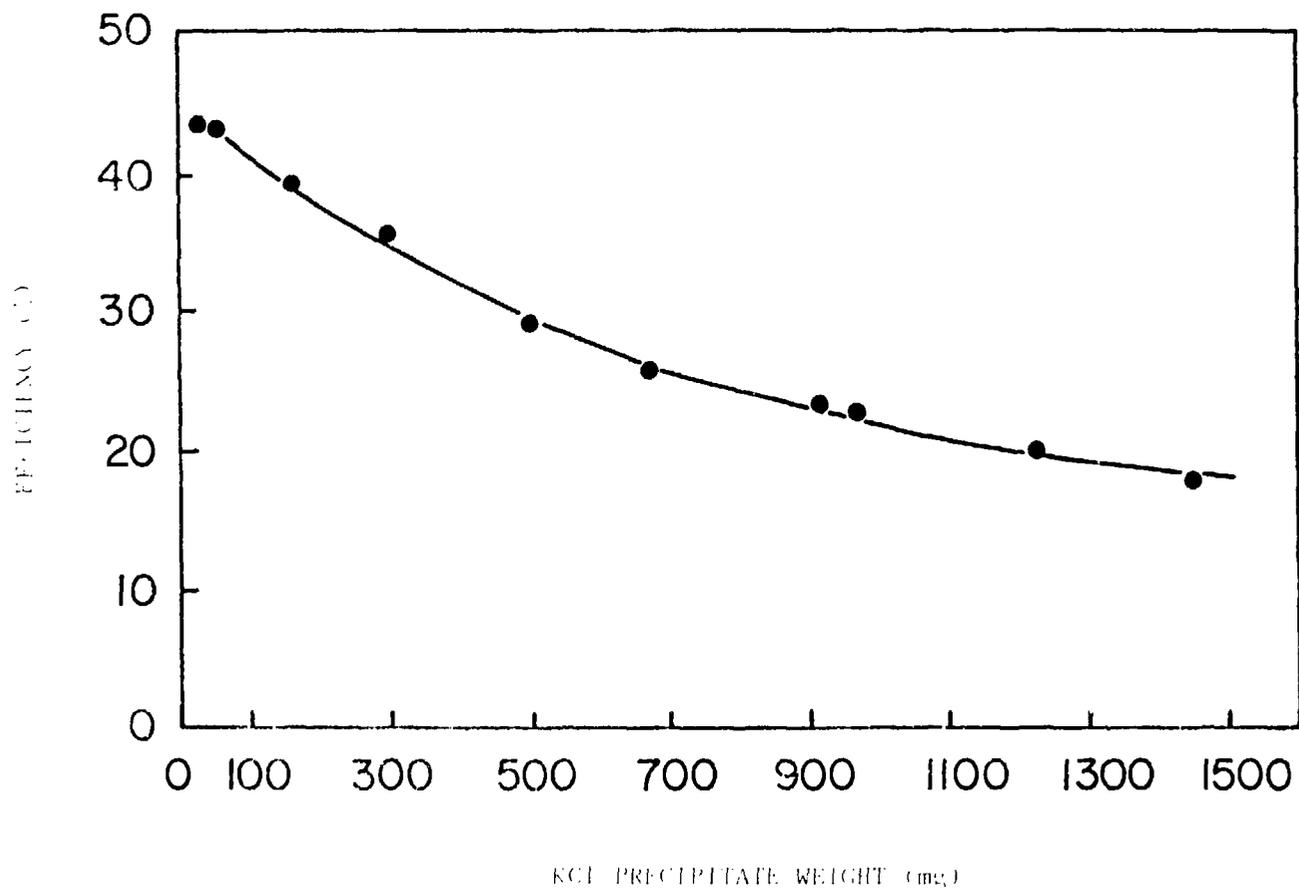


FIGURE 10: Beta Counting Efficiency Versus Precipitate Weight

Table 4 and Figure 10 illustrate that, due to absorption of the beta activity by KCl, the counting efficiency decreases nonlinearly as the amount of KCl is increased. The calibration curve of Figure 10 is used routinely to determine the correct counting efficiency for precipitates of given weights.

The beta activity per gram of KCl is calculated as follows:

$$\begin{aligned} 1 \text{ g KCl} &= 0.524 \text{ g K} \\ 1 \text{ g K} &= 0.0118\% \text{ } ^{40}\text{K} \end{aligned}$$

$$\begin{aligned} \text{Therefore } 1 \text{ g KCl} &= 0.524 \times 0.000118 \text{ g } ^{40}\text{K} \\ &= 6.184 \times 10^{-5} \text{ g } ^{40}\text{K} \\ 1 \text{ g } ^{40}\text{K} &= 1.557 \times 10^7 \text{ DPM} \\ \text{DPM per gram KCl} &= 6.184 \times 10^{-5} \times 1.557 \times 10^7 \\ &= 962.8 \end{aligned}$$

$$\begin{aligned} \text{Beta intensity (average energy} &= 0.585 \text{ meV)} \\ &= 89.3\% (13) \end{aligned}$$

$$\begin{aligned} \text{Therefore, } \beta/\text{m gram KCl} &= 962.8 \times 0.89 \\ &= 857 \end{aligned}$$

Table 4 gives the cpm and counting efficiency of ^{40}K for varying weights of KCl.

TABLE 4
cpm AND COUNTING EFFICIENCY OF ^{40}K FOR VARIOUS WEIGHTS OF KCl

WEIGHT (mg)	cpm	COUNTING EFFICIENCY %
25.0	9.4	43.7
56.2	21.1	43.7
120.7	40.1	38.6
300.0	91.8	35.6
504.7	126.4	29.1
675.9	148.7	25.6
909.7	179.4	22.9
964.2	185.2	22.3
1223.0	209.9	20.0
1448.3	222.0	17.8

3.1.4 Minimum Detectable Activity (MDA)

Gross beta screening is referenced to a ^{90}Sr assimilation. According to ICRP 10 [9], 150 DPM/day (2.5 Bq/day) in urine 30 days after an assimilation of 1480 Bq (1 investigation level) of transportable ^{90}Sr is referred to as one derived investigation level (DIL).

The urinary excretion equation describing transportable ^{90}Sr is given by [9]:

$$Y(t) = 0.12 e^{-0.693t/2.4} + 0.08e^{-1.2 \ln(t)}$$

where $Y(t)$ = daily fractional excretion of ^{90}Sr
 t = days after intake

For a DIL of 150 DPM/day (2.5 Bq/day) ^{90}Sr , the expected count rate (cpm) from a processed urine sample is given by:

$$\text{cpm} = \frac{150 \times R \times E \times V_s}{1400}$$

Where R = recovery efficiency,
 E = counting efficiency,
 V_s = volume of urine submitted (mL),
and 1400 is the average daily excretion rate in mL.
For an average sample where
 $R = 75\%$,
 $E = 30\%$,
 $V_s = 500$,
the $\text{cpm} = \frac{150 \times 0.75 \times 0.30 \times 500}{1400} = 12$

The minimum detectable activity (see Section 2.4), when $BG = 1.5$ cpm over a 30-min period, $S = 0.75$ (recovery efficiency) and $E = 0.30$ is given by

$$\text{MDA} = 2 \frac{(1.5 \times 30)^{1/2}}{0.75 \times 0.30} = 59.6 \text{ counts/30 min or } 2 \text{ cpm}$$

3.2 TRITIUM DETECTION

Existing Canadian research and power reactors are moderated with heavy water (deuterium oxide). Some of this moderator becomes radioactive with tritium (HTO) as a result of neutron activation of the deuterium atoms. This tritiated water (~450 G Bq/L of moderator in WR-1, WNRE) poses a potential contamination hazard to reactor workers. Therefore, reactor workers are frequently monitored for internal uptake of tritium by checking for tritium activity in their urine. The assay uses the following materials and methods.

Materials

- (a) Aquasure - water soluble liquid scintillation cocktail, New England Nuclear Nuclear Nuclear
- (b) 20 mL liquid scintillation vials, Fisher Scientific
- (c) A set of 7 tritium standards containing varying amounts of carbon tetrachloride quenching material, Amersham Searle

Methods

One mL of urine is added to 10 mL of liquid scintillation cocktail (LSC). At this ratio of sample to LSC, the counting efficiency is

about 35%. Higher ratios yield lower efficiencies. The tritium activity is measured directly in a liquid scintillation counter.

3.2.1 Counter Description

The tritium counter is a Nuclear Chicago Model 6844 Mark II series. It contains an automatic 300 sample chamber. A photon monitor rejects photon events that are caused by non-nuclear disintegration such as chemiluminescence, phosphorescence or phototube thermal noise. Counting of a sample is automatically terminated if these events contribute 5% or more to the gross count rate of a sample.

The counter has three channels with automatic window settings for ^3H , ^{14}C and ^{32}P . Routinely, channel A is used for tritium counting and channel B is used for ^{14}C counting.

3.2.2 Calibration

In liquid scintillation counting, the efficiency is dependent on the physical-chemical characteristics of the sample. These characteristics, such as pH, colour, sample volume and salinity, interfere with the scintillation process, generally referred to as a quenching effect. Quenching results in a decrease in the amount of light produced by each beta particle and therefore in the height of the pulse produced by the phototubes. In severe cases, the pulses may be reduced to the noise level or lost entirely. For smaller quenching effects, the beta ray energy spectrum should be only slightly shifted toward lower energies.

The change in counting efficiency due to varying degrees of quenching is routinely determined by the channel ratio method. The beta spectrum is divided into two energy regions, defined by the discriminator settings. For an unquenched sample, Channel A discriminator levels are set to include approximately 60% of the lower region of the beta spectrum while Channel B is set to include the remaining 40% for B/A of 0.67. By counting standards, with varying amounts of quenching, a relationship between the B/A ratio and counting efficiency in Channel A is determined.

A typical quench curve of counting efficiency versus B/A ratio is shown in Figure 11.

The data in Figure 11 are fitted to a linear regression equation:

$$Y = 0.85X - 2.15 \quad r = 0.99$$

Where

$$Y = \% \text{ counting efficiency and } X = \text{B/A.}$$
$$r = \text{correlation coefficient}$$

The counting efficiency for a sample is calculated from the regression equation by using the appropriate value for X.

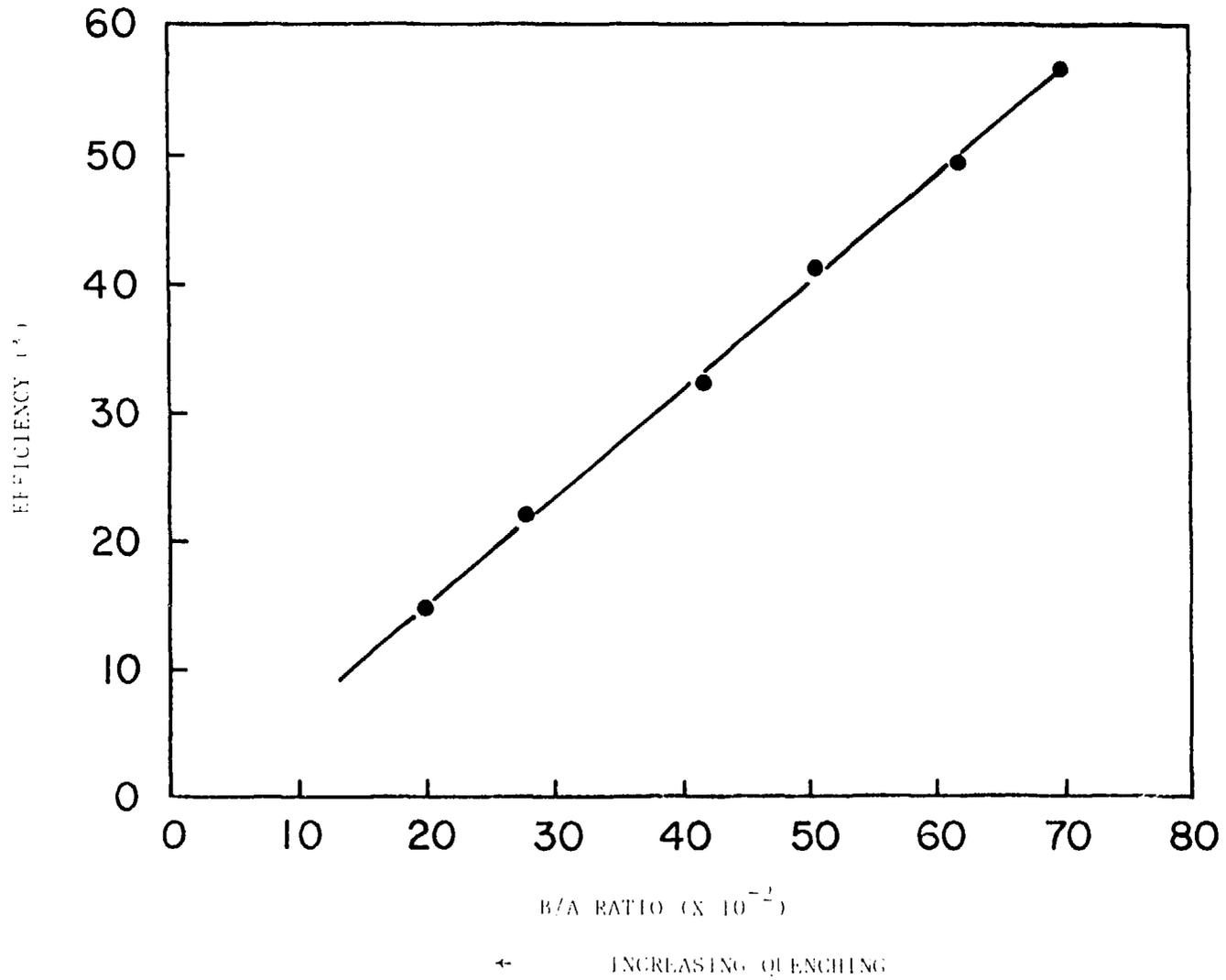


FIGURE 11: Effect of Quenching on B/A Ratio and Counting Efficiency of Tritium

3.2.3 Minimum Detectable Activity

The 30-day DIL (Section 3.3) in urine is 1.5×10^5 Bq/L [9]. At 40% counting efficiency for 1 mL of urine, this level of activity would give 3600 cpm.

The MDA (see Section 2.4) at the 95% confidence level is calculated using the following equation:

$$MDA = 2 \frac{(BG)^{1/2}}{S}$$

Where BG = 25 cpm over 20-min duration

S = 40% counting efficiency.

Therefore $MDA = 2 \frac{(25 \times 20)^{1/2}}{0.4} = 111.8$ d/20 min/mL or 4.5 DPM/mL.

3.2.4 Total Body Dose Calculation

For an occasional acute exposure, the dose calculation is based on the one recommended by the Federal/Provincial Working Group on Bioassay and In Vivo Monitoring [14].

The following formula is used:

$$D = 6.05 \times 10^{-11} C/\lambda$$

Where

D = the infinity dose in sieverts

C = concentration of tritium in urine in Bq/L and the

decay constant $\lambda = \frac{\ln 2}{T_{1/2}}$ with half-life ($T_{1/2}$) equal to 10 days.

Based on this equation, the relationship between the count rate per mL of urine, the tritium activity (Bq/L of urine) and the dose commitment (sieverts) are linearized on semi-log plots as illustrated in Figure 12.

4. GAMMA DETECTION

Radioactive isotopes, such as ^{60}Co , ^{137}Cs , ^{131}I , and others that emit energetic gamma rays, are detected directly by gamma ray spectrometry. Urine samples, as submitted by atomic radiation workers, are screened for these gamma emitting isotopes. Two gamma counting systems are used: (1) an intrinsic germanium detector with automatic sample changing and (2) a Well type NaI(Tl) with manual sample changing, used mainly as a backup for the germanium detector.

4.1 INTRINSIC GERMANIUM DETECTOR

This unit is an intrinsic germanium coaxial detector with vertical dipstick cryostat and a 30-L dewar for liquid nitrogen. This detector responds to gamma ray interaction by producing a charge signal of size proportional to the energies of the incident gamma rays. The associated

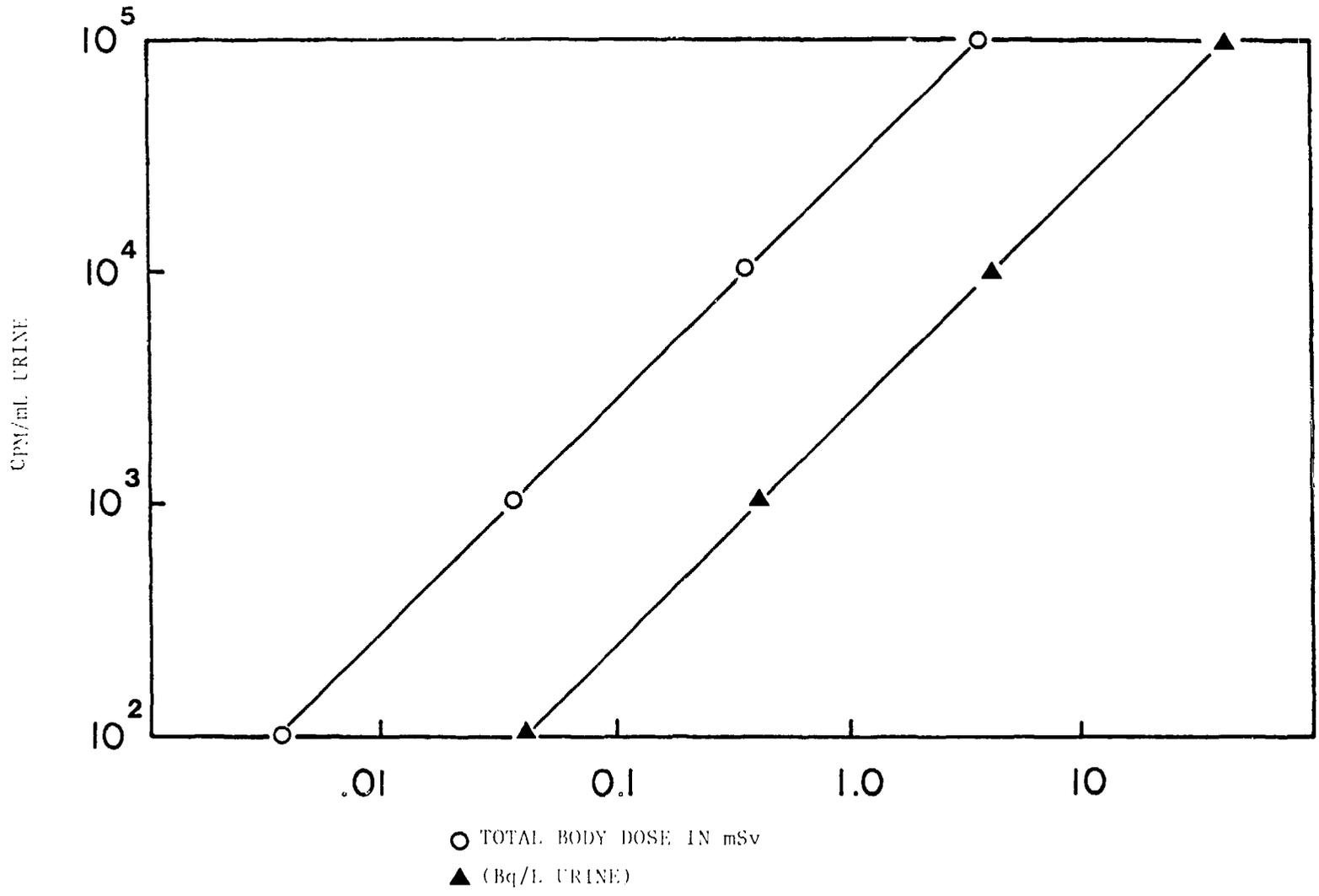


FIGURE 12: Count Rate Versus Tritium Activity and Dose Commitment

electronics (Figure 13) amplify and shape these pulses for gamma ray spectrometry.

4.1.1 Canberra Series 80 Multichannel Analyzer (MCA) and Analogue to Digital Converter (ADC)

This unit is an 8K Multichannel Analyzer with microprocessor control. The ADC is a plug-in signal processing module with 4 mixer router inputs. Each input has its own linear gate and separate live and true time channels. A direct ADC input is provided and used when Mixer-Router capability is not necessary.

The microprocessor has built-in firmware for many spectral analysis functions. The unit can count a sample, analyze the spectrum and provide a voltage signal to a sample changer controller. The Canberra series 80 has the ability to store in its memory a sequence of instructions, initiated by pressing appropriate buttons on the front panel, and executing this sequence on demand. In such a programmed task, the analyzer can count up to 20 samples, analyze the spectrum of each sample and print out the relevant region(s) of interest.

4.1.2 Counting and Spectrum Analysis

The Canberra Series 80 MCA is capable of counting a sample in the total 8192 channels or in any 256 channel segment. For routine counting using the intrinsic germanium detector, 1012 channels are used. The counting is initiated by pressing the "collect" button and terminated by previously entered preset values for either total time, counts or % error (Series 80 operator manual).

The following features are included in a typical spectrum analysis:

- (1) Automatic peak search with calculated values for peak centroid (centre channel) location and resolution at full width, half maximum (FWHM);
- (2) Spectrum smoothing by averaging 3 or 5 consecutive channels;
- (3) Background subtracting or spectrum stripping with the use of a normalizing factor;
- (4) Calculation of net and gross peak area in counts per second (cps) with 2 sigma error;
- (5) Identification of the peak energy from a stored reference calibration equation;
- (6) The printing or plotting of a full or partial spectrum. Also, the printing of net and integral counts, and the peak energy with peak limits.

4.1.3 Digital Energy Calibration

Activated by the button ENERGY CALIB, the energy calibration function allows the user to enter the gamma energies of a counted calibration source. This function automatically finds the channel number that these gamma rays are in, and by least square analysis, fits this data

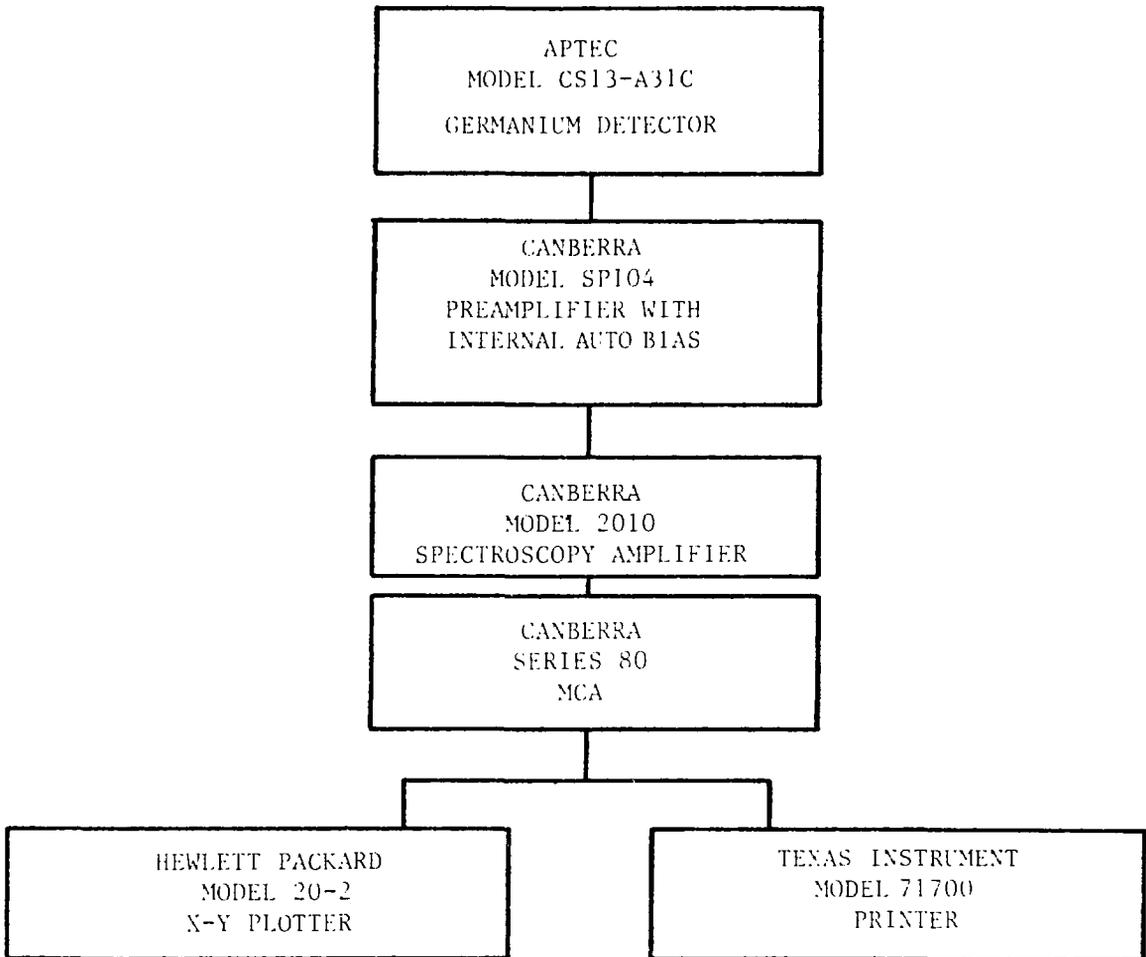


FIGURE 13: Germanium Detector Gamma Counting System

(Channel number versus gamma energy) to a first- or second-order equation. The equation is stored in memory and can be used to identify peak energy of samples. Four separate equations can be stored, one for each mixer/router input.

The fitted equations are of the form:

$y = ax + b$ or
 $y = a_1x^2 + a_2x + b$, where y is the energy for a given channel number x .

Routinely, the calibration data are fitted to a first order equation, typically calculated as $y = 1.76x + 15.6$. A ^{152}Eu calibration source is used for calculating the above equation. This source is useful because of its many well-defined gamma rays (Figure 14) and long half-life (12.7 years).

4.1.4 Task Programming

A task is a series of push button functions (instructions) activated in a sequence to perform a certain gamma ray analysis procedure. A total of 16 push-button functions are available. These are listed and described in the Series 80 operators manual.

The following sequence of instructions are used routinely to count urine samples. The sequence is identified as TASK I.

- (1) CLEAR DATA - clears data in the first 1024 channels.
- (2) CLEAR ROI - clears all regions of interest (ROI) in the first 1024 channels.
- (3) PRESET TIME - sets the counting time to seconds.
- (4) COLLECT - starts the counting and terminates at preset time.
- (5) PEAK - searches for peaks in the spectrum and sets ROI limits on all peaks.
- (6) READOUT - prints out:
 - (a) ROI and PEAK in keV,
 - (b) Integral and net area in counts and counts/second,
 - (c) % error of net area.
- (7) CLEAR ROI - clears all ROIs established in step 6.
- (8) ENTER ROI - enters new ROI between Channel 2 and 1023.
- (9) READOUT - prints out the count in the entire spectrum.
- (10) PAUSE - generates a 5-V pulse on a rear panel connector to advance the sample changer and causes the task to wait until a 'CHANGER READY' signal is received.
- (11) GO TO (1) - at the completion of step 10, the task returns to step 1.

The task is initiated by pressing the EXECUTE button. At this point the number of samples to be counted is entered via the keyboard and is equal to the number of times TASK:I will cycle. The first sample is labelled as Sample No. 1 with unit increments for identification of subsequent samples.

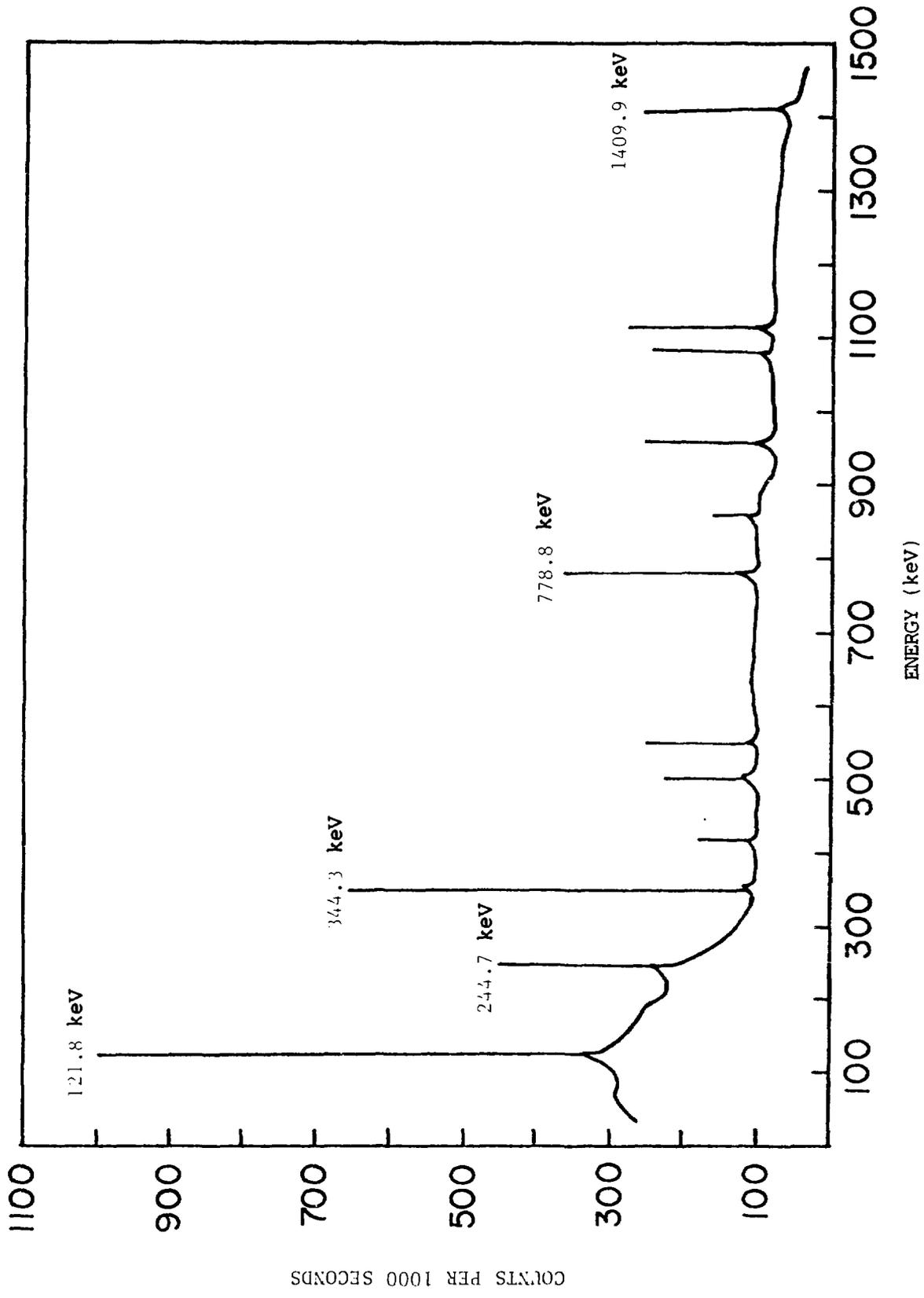


FIGURE 14: ^{152}Eu Spectrum in a 1000 Channel Region

4.1.5 Sample Changer and Controller

This unit was designed and assembled by WNRE staff and is described in detail in the Operation and Maintenance Manual [15]. A general description of this unit is as follows:

A cylindrical structure, consisting of a 10-cm lead barrier, shields the germanium detector on all sides and from the top to reduce background radiation. At a location in line with the sample changer, a pie-shaped piece of the wall of the lead shield is attached to the sample transfer assembly, leaving an open port when the sample holder is withdrawn and a closed port when it is over the detector.

The controller, activated by the PAUSE function, is programmed to follow a certain sequence of events. The starting position of the sample changer is with the sample holder situated directly over the detector.

In the first step, the controller activates a 2-RPM motor and withdraws, via a rack and pinion assembly, the sample holder from over the detector. A second 5-RPM motor then rotates the turn table, placing the next sample in front of the sample holder. The 2-RPM motor is then activated, pushing the sample to the counting position over the detector, after which the counting begins.

4.1.6 Calibration

For routine gamma counting, 10-mL samples are placed directly above the germanium detector. The detector is therefore calibrated using 10 mL of ^{152}Eu (200 Bq/mL) and 10 mL of ^{241}Am (25 Bq/mL).

Table 5 gives the gamma ray energies of ^{152}Eu and ^{241}Am and the counting efficiencies for the different photopeaks.

Figure 15 is a plot of the data in Table 5 showing the counting efficiency in the photopeaks as a function of the gamma energy. The data were obtained on 10 mL samples, placed directly over the intrinsic germanium detector.

4.1.7 Minimum Detectable Activity

The detection limit is dependent on the counting efficiency, which, as illustrated in Figure 15, depends on the gamma ray energy. It also depends on the relative intensity of each gamma ray. An example of an MDA calculation (see Section 2.4) for ^{137}Cs is given below, where:

Background = 0.02 cps/10 800 s
Counting efficiency for 10 mL of urine = 0.005
Gamma abundance [13] = 0.898
Sample recovery = 1.0, since whole urine is used
Therefore:

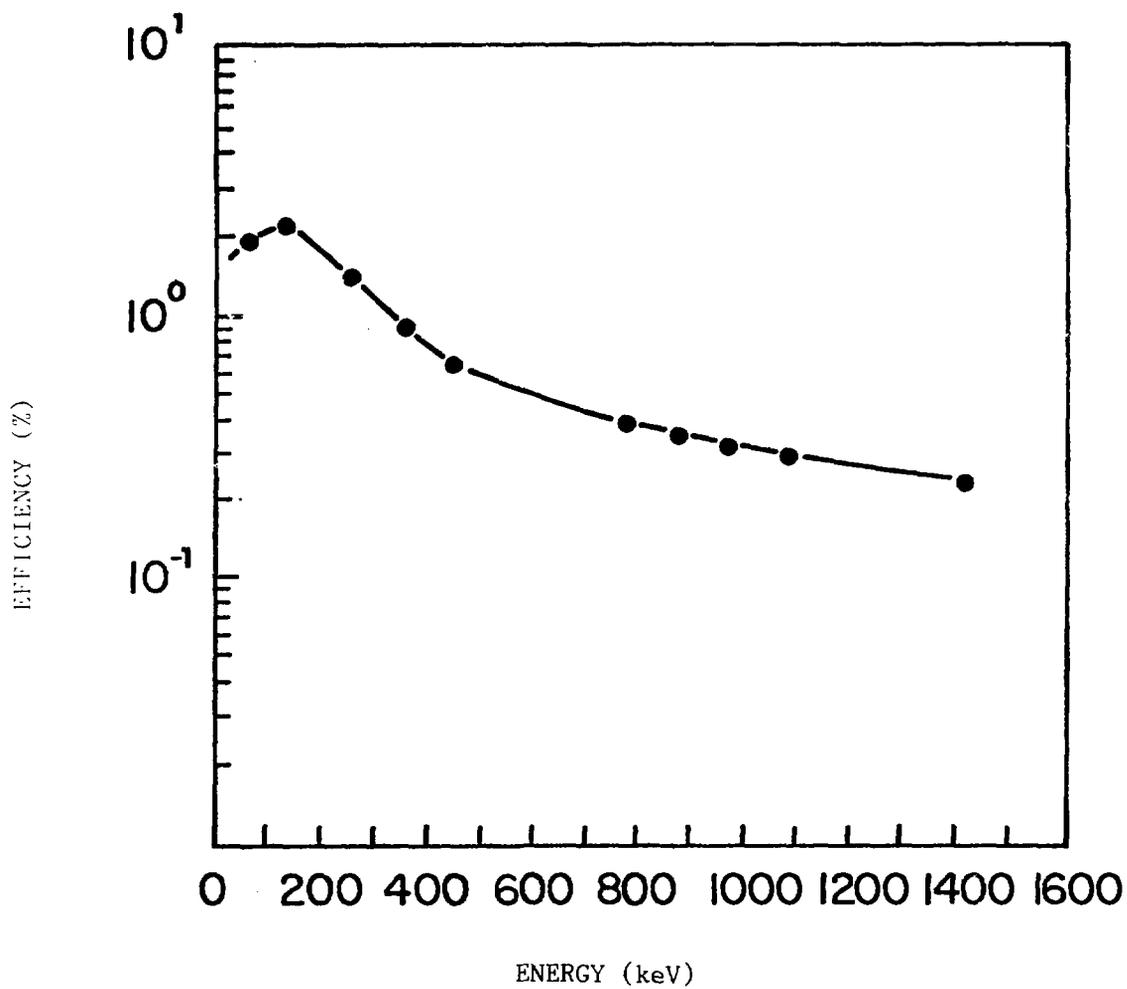


FIGURE 15: Counting Efficiency Versus Photopeak Energy

TABLE 5
EFFICIENCIES FOR VARIOUS GAMMA ENERGIES

Isotope	Energy keV	Abundance %	γs^{-1}	Peak cps	Efficiency %
²⁴¹ Am	60	35.3	88.3	1.74	1.97
¹⁵² Eu	121.8	33.2	833.0	18.6	2.23
"	244.7	7.2	180.6	2.52	1.40
"	344.3	31.4	787.8	6.83	0.87
"	443.9	3.67	92.8	0.61	0.66
"	778.8	15.2	381.4	1.43	0.38
"	867.4	5.1	128.0	0.43	0.34
"	964.0	17.3	434.1	1.37	0.32
"	1085.8	12.0	301.0	0.90	0.30
"	1407.9	24.3	609.7	1.45	0.24

$$\text{MDA} = 2 \frac{(0.02 \times 10\,800)^{1/2}}{0.005 \times 0.898}$$

= 6546.5 disintegrations/10 800 s or 0.61 DPS/10 mL urine.

For a daily urine excretion of 1400 mL, this value is equal to 0.61 x 1400/10 = 85.4 Bq/day which is well below the 30-day DIL of 481 Bq/day given in ICRP 10 [9].

4.2 DESCRIPTION OF THE SODIUM THALLIUM ACTIVATED (NaI(Tl)) WELL-TYPE DETECTOR

This detector consists of a 5 cm x 5 cm NaI(Tl) crystal, with a 2.5 cm x 3.8 cm well, that is hermetically sealed to a 5-cm-diameter photomultiplier tube. The detector assembly is in a low background shield comprised of 10 cm of lead bricks to every side, top and bottom. The inner dimension of the shield is 30 cm x 30 cm x 50 cm, with access from the top by means of a sliding door.

Figure 16 outlines the arrangement of the detector assembly and electronic components of the well-type counter.

4.2.1 Counting and Spectrum Analysis

The spectral analysis routines are the same as those discussed by Pleskach [12] on the large NaI(Tl) detector, and therefore, will not be discussed here.

The well-type detector assembly is most useful for detecting monoenergetic gamma-emitting radionuclides and where good low energy sensitivity is required.

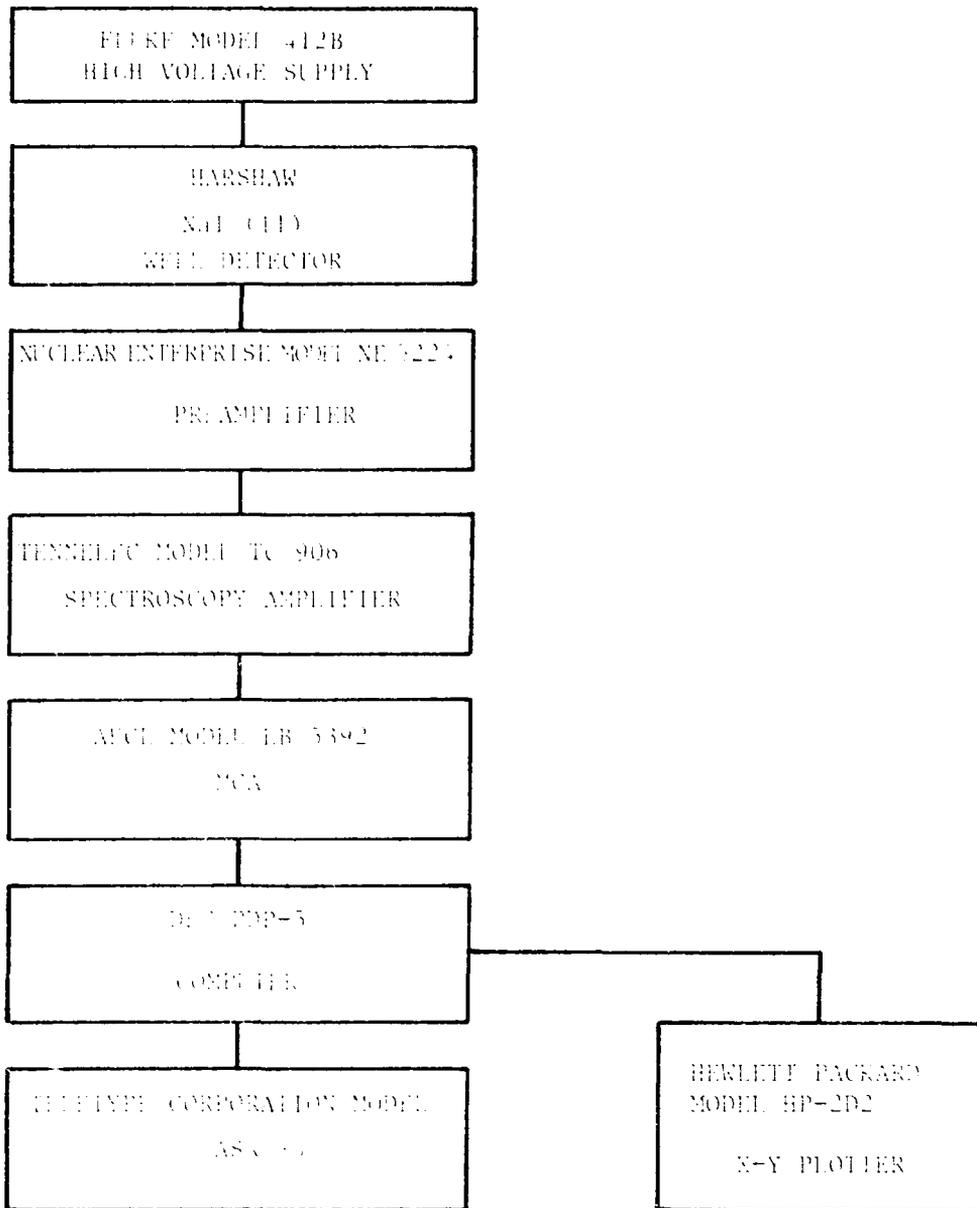


FIGURE 16: Well-Type NaI(Tl) Gamma Counting System

Sample changing is done manually, making the screening of many samples a tedious task. Therefore, this system is complementary to the intrinsic germanium detector.

4.2.2 Calibration

A 256 channel region is used for counting samples. The energy per channel is set at 10 keV using a ²²Na point source.

Photopeak counting efficiency versus gamma energy is done using 10-mL standards. Table 6 shows the photopeak efficiency for 5 different radionuclides.

TABLE 6
PHOTOPEAK EFFICIENCIES FOR VARIOUS GAMMA RAY ENERGIES

Isotope	Energy KeV	gamma/min	cpm	Photopeak Efficiency (%)
²⁴¹ Am	60	8,168	5 366	65.7
¹⁴¹ Ce	145.4	938	320	34.1
⁸⁵ Sr	514	16 827	1 225	7.3
¹³⁷ Cs	662	6 429	331	5.2
⁶⁰ Co	1,332	102 440	922	0.9

Figure 17 illustrates a typical calibration curve of gamma ray energy versus the photopeak counting efficiency for the Well-type NaI(Tl) detector.

4.2.3 Minimum Detectable Activity

An example of an MDA calculation (see Section 2.4) for ¹³⁷Cs is given below, where:

Background = 0.12/10 800 s
 Counting Efficiency for 10 mL of urine = 0.052
 Gamma Abundance (13) = 0.898
 Sample Recovery = 1.0
 Therefore:

$$MDA = 2 \frac{(0.12 \times 10\ 800)^{1/2}}{0.052 \times 0.898}$$

= 1541.9 disintegrations/10 800 s/10 mL or 0.14 DPS/10 mL.

For a daily urine excretion of 1400 mL, the above value becomes 0.14 x 1400/10 = 19.6 Bq/day which is well below the 30-day DIL of 481 Bq/day given in ICRP 10 [9].

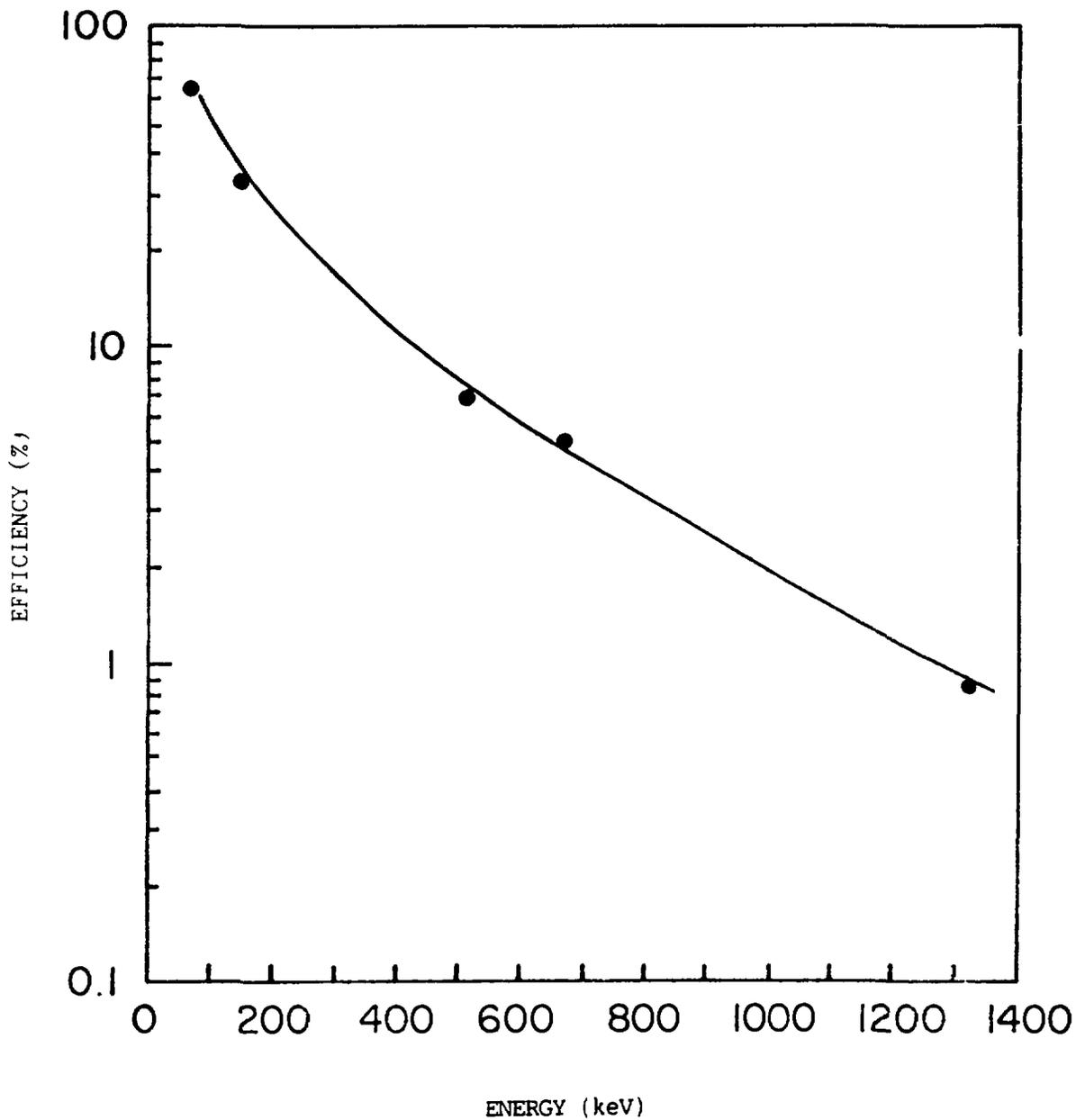


FIGURE 17: Counting Efficiency Versus Photopeak Energy for the Well-Type (NaI(Tl) Detector

5. CALL-IN CRITERIA

As outlined in the Whole Body Counting Report [12], requests for Bioassay samples are issued by special schedule or by random selection.

5.1 SPECIAL SCHEDULE

In the special schedule category, employees are requested to submit urine samples on a regular basis depending on the nature and location of their work. Special scheduling of requests is directed at: (1) Hot cell workers being monitored bimonthly for fission products, (2) Reactor workers being monitored monthly for tritium, and (3) Other groups in accordance with the level of radioactivity that they handle. This varies as programs change.

5.2 RANDOM SELECTION

The random selection process is probabilistic in nature and is documented in Pleskach [12].

All workers are placed into groups, with members in any particular group having a similar risk of exposure to radionuclides in the work place. By randomly choosing a fraction of each group for internal monitoring at a certain sampling interval, a desired confidence level for detecting a radionuclide assimilation can be attained. Figures 18 and 19 shows how the size of the group fraction to be monitored per sampling interval, in order to achieve confidence levels of 75 or 95%, varies with the length of the sampling interval. Clearly, the longer the sampling interval, the larger the group fraction must be. Logistically, large fractions per sampling period cannot be handled if the MDA criteria given above are to be met. Therefore, a sampling interval of one month was chosen to make the size of individual group fractions to be monitored per sampling interval more manageable.

The size of the monthly group fraction for fission products is based on the urinary excretion rate ($U(t)$) of ^{90}Sr [6] given by:

$$U(t) = 0.116 e^{-0.693t/2.4} + 0.08 e^{-1.2 \ln t} \quad (2)$$

where $U(t)$ is the fraction excreted t days after assimilation.

The group fraction to be monitored is related to the sampling interval and probability of being able to detect an assimilation of radionuclides and is given by [16]:

$$(PGP) = 1 - (1-F)^m [1-F(T/I-m)] \quad (3)$$

where

$P(GP)$ = probability of being able to detect an assimilation of ^{90}Sr in a group of workers.

T = time interval in which 1% of an ALI of ^{90}Sr can be detected before the excretion rate, based on Equation (2), falls below the MDA.

I = sampling interval.

F = fraction of the group monitored during the sampling interval.

m = integer of T/I .

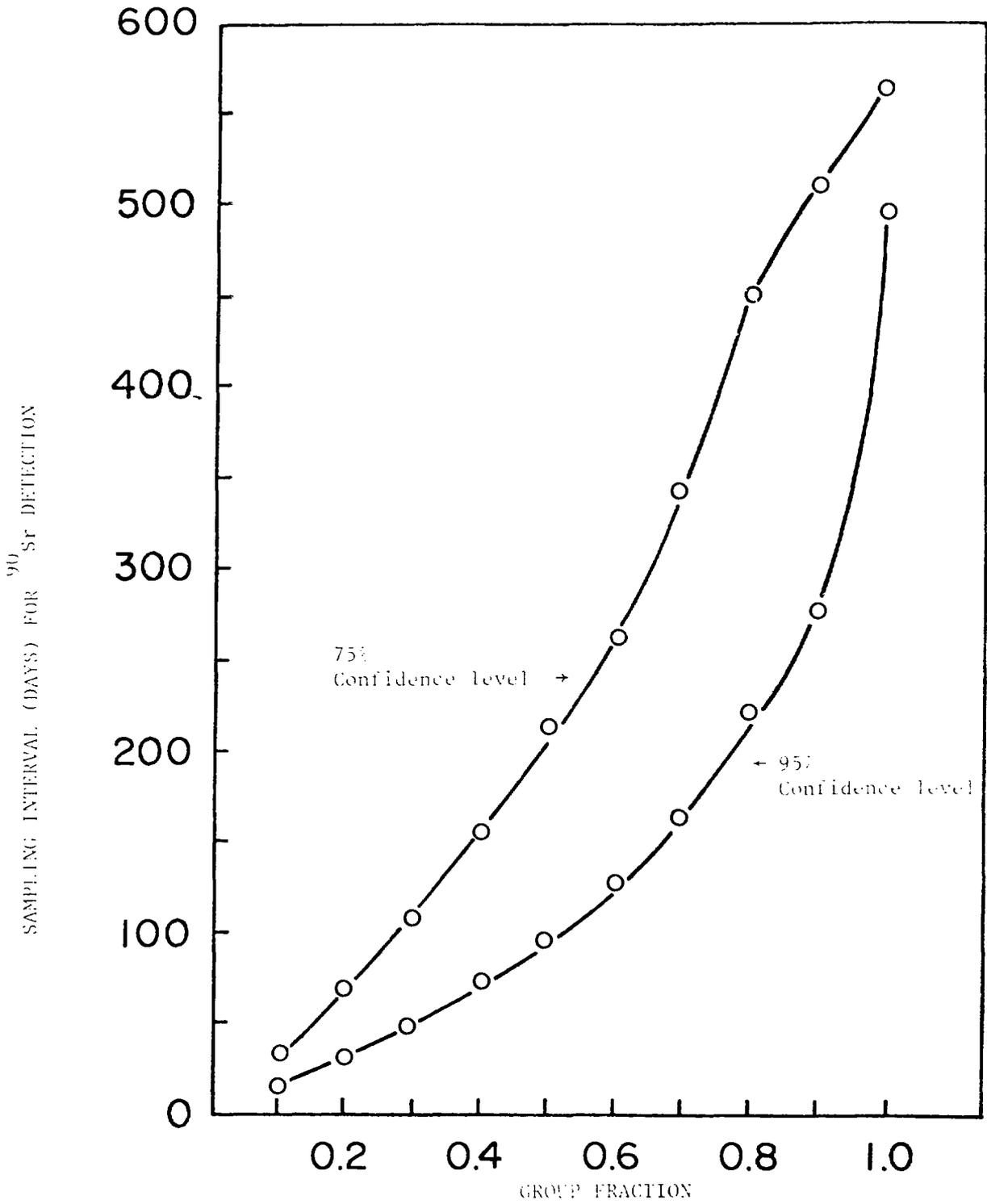


FIGURE 18: Sampling Interval Versus Group Fraction to Attain 75 and 95% Confidence Levels for Detecting a 1% Assimilation of an ALI of ^{90}Sr

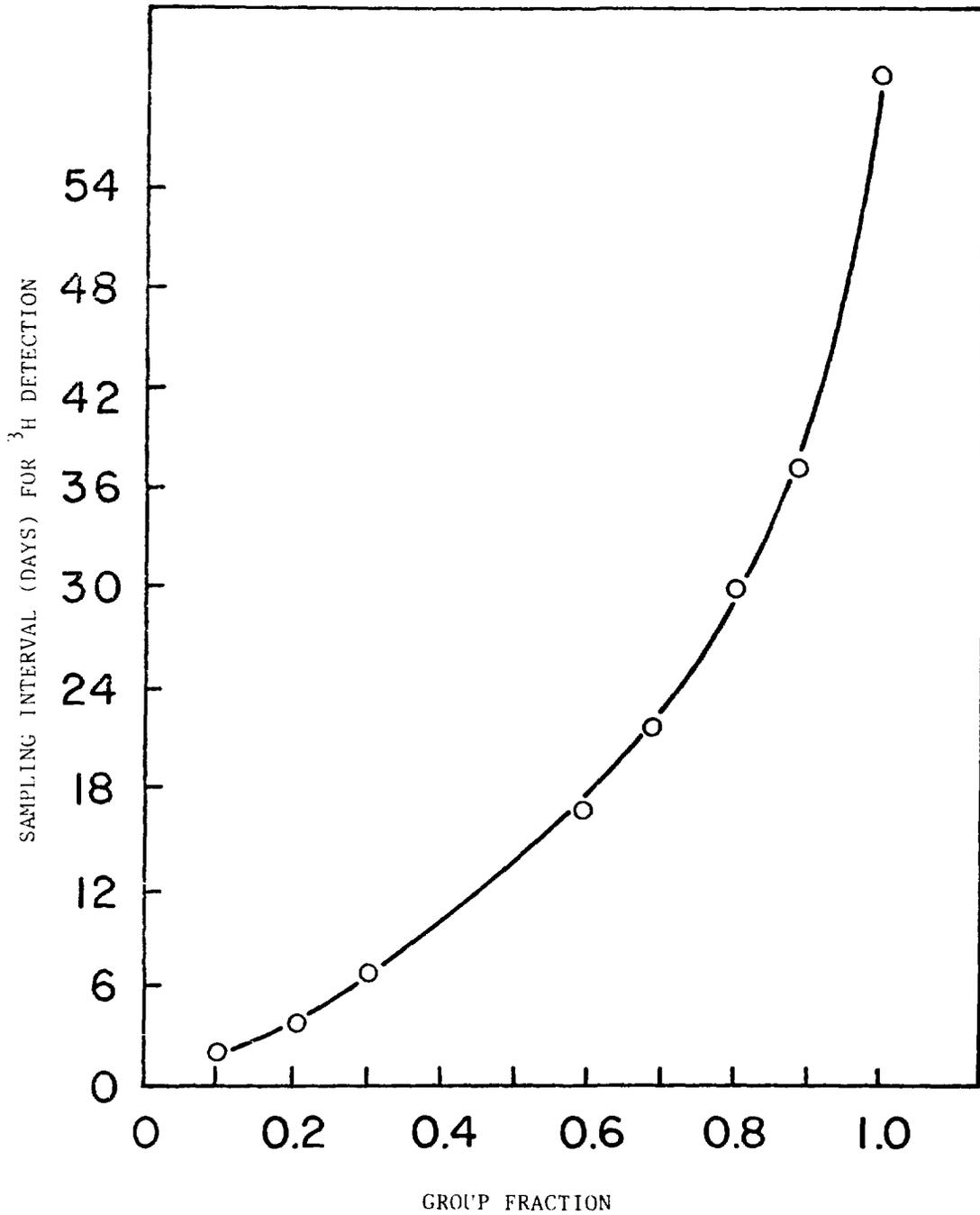


FIGURE 19: Sampling Interval Versus Group Fraction to Attain a 95% Confidence Level for Detecting a 1% Assimilation of an ALI of ^3H

From Equation (2), the length of time (T), in days, during which the excretion rate of 1% of an ALI of ^{90}Sr ($7 \times 10^3 \text{Bq}$) [17] will fall to the MDA of 0.42 Bq/day equals 400 days, and by rearranging Equation (3),

$$T/I = \frac{P(\text{GP}) - 1 + (1-F)^m + Fm(1-F)^m}{F(1-F)^m} \quad (4)$$

For given values of F and P(GP), Equation (4) is used to calculate T/I by iteratively defining the correct integer value for m. Once T/I is computed, I can be calculated since T is known from Equation (2). Table 7 and Figure 18 show how the I values vary for different values of F with P(GP) = 0.95.

The monthly group fraction for ^3H is based on the urinary excretion Equation (9) given by:

$$U(t) = 0.07e^{-0.693t/10} \quad (5)$$

where U(t) is the excretion rate after t days.

T is calculated from Equation (5) and is the length of time during which an assimilation of 1% of an ALI ($3 \times 10^7 \text{Bq}$) [17] decreases to the bioassay reporting limit of $6 \times 10^5 \text{Bq}$ (-1 mrem).

Equation (4) is used to calculate the sampling intervals for ^3H for given P(GP) and F values. These data are shown in Table 8 and Figure 19.

The goal of monitoring groups to the 95% confidence level during each sampling interval is only met when all the requests for samples are honoured promptly. Failure to do so lowers the confidence level of the specific monitoring procedure(s).

Table 9 lists the fraction of each group selected each month, the number of cases involved and the probability P(GP) of being able to detect 1% of an ALI of either ^{90}Sr or ^3H by urinalysis. The individuals in each group are selected randomly using a computer program described in another report [7].

6. REPORTING OF RESULTS

Bioassay measurements are tabulated and reported biweekly. Positive measurements are categorized and reported as:

- (1) Removal (R). For fission products, R = 150 DPM/day. This is based on an approximate 30-day derived investigation level (DIL) for soluble ^{90}Sr [9]. For ^3H , R = $1.85 \times 10^6 \text{Bq/L}$ ($50 \mu\text{Ci/L}$), approximately equal to one investigation level [9].

A person receiving an internal contamination of one R or more is removed from further handling of radioactivity until the level drops to below 1 R. The incident is investigated and the cause determined.

TABLE 7

GROUP FRACTIONS, T/I RATIOS AND SAMPLING INTERVALS FOR A 95%
CONFIDENCE LEVEL OF DETECTING ⁹⁰Sr WITH EXCRETION TIME T = 400 DAYS

GROUP FRACTION (F)	T/I RATIO	SAMPLING INTERVAL (DAYS (I)
1.0	0.95	421
0.9	1.556	257
0.8	1.938	206
0.7	2.635	152
0.6	3.365	119
0.5	4.400	91
0.4	5.892	68
0.3	8.441	47
0.2	13.453	30
0.1	28.446	14

TABLE 8

GROUP FRACTION, T/I RATIOS AND SAMPLING INTERVALS FOR A 95% CONFIDENCE
LEVEL OF DETECTING ³H WITH EXCRETION TIME T = 56.5 DAYS

GROUP FRACTION (F)	T/I RATIO	SAMPLING INTERVAL (DAYS) (I)
1.0		0.95
59		
0.9	1.556	36
0.8	1.938	29
0.7	2.635	21
0.6	3.365	17
0.5	4.400	13
0.4	5.892	10
0.3	8.441	7
0.2	13.453	4
0.1	28.446	2

TABLE 9
MONTHLY GROUP FRACTIONS AND CONFIDENCE LEVEL
FOR DETECTING 7×10^3 Bq ^{90}Sr AND 3×10^7 Bq ^3H ASSIMILATION

Group	No. in Group	Fraction of Group Selected/Month	No. Selected	P
. Hot cells	17	All are monitored bimonthly	17	0.95
. WR-1	39	1.0 (for ^3H)	39	0.95
. (a)	39	0.4	16	0.95
. Reactor Technology - Reactor Lab - Fission Product Lab - Plutonium	24	0.4	10	0.95
. Researchers	350	0.2	70	0.95
. Decontamination	16	0.4	6	0.95
. Engineering Maintenance	315	0.2	63	0.095
. Administration	322	0.1	32	0.075

- (2) Caution (C) where: $IR > C > R/2$. This level of internal contamination triggers an investigation of the incident to determine the cause.
- (3) Minor (M) where: $R/2 > M > R/3$. This level of internal contamination results in the worker being advised of the internal contamination and is urged to identify the cause.
- (4) Negligible (N) where $N < R/3$. No action is taken.

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