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CALIBRATION PROCEDURES FOR IN-VIVO SODIUM IODIDE SPECTROMETRY OF PLUTONIUM AND AMERICIUM IN THE HUMAN LUNG

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INTRODUCTION

This paper describes the calibration techniques and associated error analysis for the in-vivo measurement by NaI spectrometry of heavy elements in the lung, specifically plutonium and americium. A very brief description of the instrumentation system is included.

INSTRUMENTATION SYSTEM

The detection system consists of twin phoswich detectors, each with a 3-mm thick front NaI (Tl) crystal and a 51-mm thick back CsI (Tl) crystal coupled to a single 5-in. phototube. The two circular detectors are covered with 0.25 mm beryllium and have a total surface area of 206 cm^2. The signals from the phototubes are amplified and routed to a pulse-shape analyzer and linear gate. Those signals with the proper rise time characteristics (NaI) are passed through the linear gate to an ADC. Shielding for the system is provided by a steel room with 20-cm thick walls, floor, and ceiling. A 3-mm thick lead lining is attached to the inner surfaces of the room for improved background reduction. The background in the room measured with the phoswich detectors (including electronic cosmic-ray overload rejection) is approximately 0.06 cpm/cm^2 of detector surface in the 14- to 25-KeV region. The high altitude of our Laboratory (2,200 meters) is a contributing factor to our high background as compared to other laboratories.

For signal processing, a minicomputer is interfaced to the ADC. The computer is used for data storage (twenty 100-channel spectra), as well as automatic data analysis. The latter includes background subtraction (both room and natural body), lung burden calculation using an internal

calibration, and calculation of errors. For routine human counting, the detector system is calibrated daily for electronic efficiency (this checks the pulse gating and timing circuitry), absolute counting efficiency in a standard counting position, and energy calibration (electronic gain).

The calibration of lung burden vs count rate is highly dependent on the thickness of tissue overlying the lungs, particularly for the 14- to 25-KeV region. Hence, the chest wall of each subject is measured ultrasonically in 12 locations, six over each lung on a 4.5-in. diameter circle tangent to the sternum and clavicle. This circle is the location for the placement of the outer edge of each detector. These 12 individual measurements are entered into the computer which calculates an exponential average of the 12. This average chest wall thickness is then used to calculate the plutonium and americium calibrations and, hence, burdens for the subject. The subjects are normally counted for 30 minutes, while the room background is counted for 60 minutes. The two counts are taken within a few hours of each other. The room backgrounds are measured with the detectors placed on a Lucite phantom to simulate the self-shielding by the subjects. During the actual count of each subject, the detectors are held to the chest of the prone person with elastic straps to prevent movement of the detectors away from the subject.

SYSTEM CALIBRATION

For the measurement of ^{241}Am, we monitor its 60-KeV gamma ray using an energy band of 51 to 70 KeV. For plutonium, both ^{238}Pu and ^{239}Pu, we monitor their L X-ray region in a band from 14 to 25 KeV. (Due to the absence or uncertainty in the ^{241}Am content of our plutonium, we are not allowed the luxury of monitoring the ^{241}Am for plutonium determination as is done at some other laboratories.) To account for the natural human backgrounds (^{40}K and ^{137}Cs), we monitor the region 75 to 100 KeV which has no interference from plutonium or americium. Using a large group of known "clean" LASL workers, we obtained natural body background strip-out factors for the contributions in the 17- and 60-KeV regions. Once room backgrounds are subtracted from the gross spectrum of each subject, these body background strip-out factors are then applied to that spectrum. The

remaining counts (net) in the 17-KeV region are then considered to be due entirely to L X-rays from both ^{241}Am and plutonium. The net counts in the 51- to 70-KeV region are considered due only to ^{241}Am. In the event that ^{241}Am is present with the plutonium, which is nearly always the case with ^{239}Pu workers, a final correction must be made to the counts remaining in the 17-KeV region due to ^{241}Am. These last remaining counts are then considered due only to plutonium. This americium strip-out factor is determined using tissue transmission measurements of the relative intensity of the americium 17-KeV region to the 60-KeV region.

The tissue transmission measurements were made using mock lungs uniformly loaded with ^{239}Pu, ^{238}Pu, or ^{241}Am. The absorption medium was lean beefsteak cut into 1-cm thicknesses. Each transmission measurement was made with one or more pieces of beefsteak placed directly on top of the mock lung, with the phoswich detector (only one was used for these measurements) directly in contact with the beefsteak. Using this method, the "r^2" dependence of the transmission data was automatically included as the tissue thickness was changed. Compared with the conventional method of using fixed detector-source geometry in such transmission measurements, the non-fixed geometry method eliminated a potential $\approx 16\%$ error in the overall lung burden calibration.

The tissue transmission measurements provide a relative calibration as a function of chest wall thickness. To provide an absolute calibration, a REMAB phantom was loaded with the distributed source mock lungs mentioned above (RANDO lung stock; Alderson Research Laboratories, Inc.). The average tissue-equivalent chest thickness of the phantom was determined by a series of ultrasonic measurements coupled to the relative transmission of the 17- and 60-KeV regions from the distributed ^{241}Am mock lung in the phantom. The chest wall of the phantom thus determined was 3.2 ± 0.15 cm. For the three isotopes of interest here, the phantom data provided their respective number of counts per second per nCi of activity (cps/nCi) at a tissue thickness of 3.2 cm. The relative transmission data then allow scaling of that absolute measurement to other chest wall thicknesses.

Figure 1 shows the plutonium L X-ray transmission data (14 to 25 KeV) as a function of tissue thickness. Note that the data are normalized to 1.00 at 1 cm of tissue. Both ^{238}Pu and ^{239}Pu transmission data are plotted on the same graph. The error bars for the 1-, 2-, and 3-cm data are within their plotted symbols. Since the data for the two isotopes of plutonium agreed so well, a composite least-squares fit to all of the data was performed. This final fit is given in Table 1.

Figure 2 shows similar data for the 60-KeV gamma ray (51- to 70-KeV region) from ^{241}Am. Also included on the figure are the data for ^{103}Pd (17 to 27 KeV) which is being used in plutonium counting intercalibration studies. Table 1 gives the computer-fit equations for the two sets of transmission data. Error bars for the data of Figure 2 are within the symbols. Figure 3 shows the relative transmission data for the 14- to 25-KeV and the 51- to 70-KeV regions using the ^{241}Am distributed source. These data provide the ^{241}Am strip-out factor, P, when it is desired to determine the plutonium burden in the presence of ^{241}Am. The computer fit for these data is also shown in Table 1.

The natural body background strip-out factors, K, were obtained from a group of 92 "clean" LASL plutonium workers. These factors are just the ratios of the 14- to 25-KeV region counts or the 51- to 70-KeV region counts (room background subtracted) to the counts in the 75- to 100-KeV region, respectively. These factors and their standard deviations are shown in Table 1. As more "clean" workers are counted, these factors will be updated.

The final absolute calibration factors were determined using the phantom as described above. This factor, F_i (nCi/cps), was measured at 3.2-cm tissue thickness for the three nuclides, as well as for ^{103}Pd. By solving the equation $C_i = F_i T_i$ for each isotope, where C is the calibration constant and T is the transmission equation as given in Table 1 and evaluated at 3.2 cm, the final calibration equations, F_i, were obtained. These are listed in Table 1. Figure 4 shows these calibration factors drawn as a function of tissue thickness. Note that the curves are just the inverse of the transmission curves (Figures 2 and 3) normalized to the phantom results

at 3.2 cm. Using these calibration factors, Table 2 defines the terms and gives the final expression for the lung burden determination in nCi. As implied by the various equations for the burden calculation, we have no way to differentiate between ^{238}Pu and ^{239}Pu in the actual measurement. We must have isotopic information on the exposure before a correct burden can be calculated. The ^{241}Am interference, however, can be determined in the measurement as described above. Note that the ratio of F_{238}/F_{239} is 2.1 and not ~ 2.5 as would be indicated by the data found in the tabulation of Swinth.[1] We have measured this ratio (distributed sources in phantoms) numerous time with the same result. We have no explanation for this apparent discrepancy.

ERROR ANALYSIS

Errors associated with the calculated lung burdens, B, are determined by first assuming that no measurement biases are present (this is not necessarily true and will be discussed later) and that the errors in all of the terms are completely random in nature and not correlated. Since all of the burden calculations are of the form $B = N \cdot F$, we use the general expression:

$$\Delta B/B = \left[(\Delta N/N)^2 + (\Delta F/F)^2\right]^{1/2}, \qquad \text{(Eq. 1)}$$

where Δ signifies a random error.

Considering the second term first, the general form of the calibration factor, F, is the same for all cases, namely:

$$F = Ce^{a + bt + ct^2} = Ce^{f(t)}. \qquad \text{(Eq. 2)}$$

Therefore, for all cases, we have

$$\Delta F/F = \left[(\Delta c/c)^2 + (\Delta e^{f(t)}/e^{f(t)})^2\right]^{1/2}, \qquad \text{(Eq. 3)}$$

where

$$\Delta e^{f(t)}/e^{f(t)} = \Delta f(t) = \Big\{(\Delta a)^2 + \Big[(\Delta b/b)^2 + (\Delta t/t)^2\Big](bt)^2 + (ct^2)^2\Big[(\Delta c/c)^2 + 2(\Delta t/t)^2\Big]\Big\}^{1/2}. \qquad \text{(Eq. 4)}$$

Using the estimates of the errors in the parameters obtained in the computer fits of the transmission data, the values obtained for $\Delta f(t)$ are:

$$^{238,9}\text{Pu} \cong 26\%$$
$$^{241}\text{Am} \cong 6\%.$$

Due to the slow variation of $\Delta f(t)$ with chest thickness, t, these values are approximately constant ($\sim$7% relative) over the range $1.5 \leq t \leq 3.0$ cm.

For $\Delta C/C$, we have the following estimated uncertainties:

^{238,9}Pu mock lung loading	10%
Variation in area covered by ribs	5%
Phantom chest wall measurement	14%
Phantom-mock lung measurement	5%
Total error $\Delta C/C$ =	19%

^{241}Am mock lung	10%
Phantom chest wall measurement	4.3%
Phantom-mock lung measurement	5%
Total error $\Delta C/C$ =	12%

Therefore, using Eq. 3, we have for $\Delta F/F$:

$$^{238,9}\text{Pu} \cong 32\%$$
$$^{241}\text{Am} \cong 13\%,$$

which are approximately constant as a function of tissue thickness, t.

For $\Delta N/N$ in Eq. 1, we have for ^{238,9}Pu:

$$N_{17} = R_1 - K_1R_3 = G_1 - B_1 - K_1 (G_3 - B_3), \qquad \text{(Eq. 5)}$$

where

G_i = gross subject count rate in region i

B_i = gross room background count rate in region i

$$\therefore \Delta N_{17}/N_{17} = \left\{ G_1/T_c + B_1/T_b + (K_1G_3)^2 \left[(\Delta K_1/K_1)^2 + 1/T_cG_3 \right] + (K_1B_3)^2 \left[(\Delta K_1/K_1)^2 + 1/T_bB_3 \right] \right\}^{1/2} \Big/ N_{17}. \quad \text{(Eq. 6)}$$

where

$$\Delta G_i = (G_iT_c)^{1/2}/T_c \text{ cps and } \Delta B_i = (B_iT_b)^{1/2}/T_b \text{ cps}$$

and T_c = subject count time (sec) and

T_b = room background count time.

Similarly for ^{241}Am:

$$N_{60} = R_2 - K_2R_3 = G_2 - B_2 - K_2 (G_3 - B_3) \quad \text{(Eq. 7)}$$

$$\therefore \Delta N_{60}/N_{60} = \left\{ G_2/T_c + B_2/T_b + (K_2G_3)^2 \left[(\Delta K_2/K_2)^2 + 1/T_cG_3 \right] + (K_2B_3)^2 \left[(\Delta K_2/K_2)^2 + 1/T_bB_3 \right] \right\}^{1/2} \Big/ N_{60}. \quad \text{(Eq. 8)}$$

For N'_{17}, the case where ^{241}Am interferes with plutonium, we have:

$$N'_{17} = N_{17} - PN_{60} \quad \text{(Eq. 9)}$$

$$\therefore \Delta N'_{17}/N'_{17} = \left\{ (\Delta N_{17})^2 + (PN_{60})^2 \left[(\Delta P/P)^2 + (\Delta N_{60}/N_{60})^2 \right] \right\}^{1/2} \Big/ N'_{17}. \quad \text{(Eq. 10)}$$

But $P = e^{(-0.909 - 0.748t + 0.094t^2)}$ [see Table 1]

and $$\Delta P/P = \Delta e^{f(t)}/e^{f(t)} = \Delta f(t) =$$

$$\left\{ (\Delta a)^2 + (bt)^2 \left[(\Delta b/b)^2 + (\Delta t/t)^2 \right] + (ct^2)^2 \left[(\Delta c/c)^2 + 2(\Delta t/t)^2 \right] \right\}^{1/2}.$$

Using the appropriate values in $\Delta P/P$, we find that

$$\Delta P/P \sim 14\%; \; 1.5 \leq t \leq 3. \quad \text{(Eq. 11)}$$

. Using the results of Eqs. 6, 8, and 11 and substituting into Eq. 10, we can readily solve for $\Delta N'_{17}/N'_{17}$.

Returning now to Eq. 1, we have for the final errors on B:

$$^{238,9}\text{Pu:}\quad \Delta B/B = \left[(\Delta N_{17}/N_{17})^2 + (0.32)^2\right]^{1/2} \qquad \text{(Eq. 12)}$$

$$\Delta B'/B' = \left[(\Delta N'_{17}/N'_{17})^2 + (0.32)^2\right]^{1/2} \qquad \text{(Eq. 13)}$$

$$^{241}\text{Am:}\quad \Delta B/B = \left[(\Delta N_{60}/N_{60})^2 + (0.13)^2\right]^{1/2}. \qquad \text{(Eq. 14)}$$

Although involved, Eqs. 12, 13, and 14 are handled in a straightforward manner by using the results of Eqs. 6, 7, and 10. It should be noted that the errors represent 1σ error limits. Also, no matter what the lung burden, a plutonium burden has at least a 32% absolute error and an ^{241}Am burden has at least a 13% error.

Recent autopsy data at LASL[2] indicate that plutonium distributional effects are real and large in a limited number of cases examined. Measurements with phantoms at LASL have also shown that reasonable placement of point plutonium sources can easily lead to errors of 200 to 300%, compared to the uniform calibration discussed here. Therefore, it is paramount that we recognize the possibility of large biases (both positive and negative) in in-vivo lung counting. Repeated positive counts indicate a real plutonium burden, albeit with a potentially large error. However, a repeated null result, in reality, may be false if the plutonium is "hiding."

DETECTABILITY

Any treatment of plutonium lung counting should include a discussion of detection limits. This complicated subject has been discussed in detail by Currie.[3] Instead of the 2σ or 3σ levels above background commonly used in trace element analysis, we choose to define detectability (Currie's L_c, critical level) as the 1.64σ level, where σ refers here to the total uncertainty in the net count rate measurement (body background also subtracted). In Currie's treatment, the deviation in background from a well characterized blank was used in his critical level definition. Our "blank"

background is a much more complicated function due to varying natural human body radioactivity.

For determining our detection limits, we assume that the frequency distribution of net counts above background (N_{17} or N_{60}) is normal. We now find N such that $N = 1.64\ \Delta N$. This gives a 95% probability of a real burden giving a positive net count. Hence, we solve $N_{17} = 1.64\ \Delta N_{17}$ and $N_{60} = 1.64\ \Delta N_{60}$ using Eqs. 6 and 8, respectively, for ΔN with G_1 or G_2 replaced by Eqs. 5 or 7, respectively. The resulting equations are quadratic in N_{17} or N_{60} but can easily be solved. By application of the appropriate calibration factor, F, we can obtain the detection limits in nCi for any particular count.

Using the above method and typical data, we find the following average detection limits for a single 30-minute count, a 60-minute background count, and a subject having a 2.3-cm chest wall thickness (about average for LASL workers):

^{238}Pu: $D_{1.64\sigma} = 10$ nCi

^{239}Pu: $D_{1.64\sigma} = 21$ nCi

^{241}Am: $D_{1.64\sigma} = 0.24$ nCi

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2. J. F. McInroy, Los Alamos Scientific Laboratory, private communication (1976).
3. L. A. Currie, Limits for Qualitative Detection and Quantitative Determination -- Application to Radiochemistry, Anal. Chem. 40:586 (1968).

TABLE 1. Computer Fits and Other Factors

I. Transmission Equations

A. $^{238,9}Pu$: $T = e^{(1.438 - 1.545t + 0.096t^2)}$

B. ^{241}Am: $T = e^{(0.301 - 0.303t)}$

C. ^{103}Pd: $T = e^{(0.873 - 0.883t)}$

t = tissue thickness, cm

II. ^{241}Am Strip-Out Factor, P

$P = e^{(-0.909 - 0.748t + 0.094t^2)}$

III. Body Background Strip-Out Factor, K

$K_1 = 0.136 \pm 0.026$; 17-KeV region

$K_2 = 0.908 \pm 0.060$; 60-KeV region

IV. Calibration Factors: F_i (nCi/cps) = C_i/T_1, C constant

A. ^{238}Pu: $F_{238} = 41e^{-(1.438 - 1.545t + 0.096t^2)}$

B. ^{239}Pu: $F_{239} = 86e^{-(1.438 - 1.545t + 0.096t^2)}$

C. ^{241}Am: $F_{241} = 1.4e^{-(0.301 - 0.303t)}$

D. ^{103}Pd: $F_{103} = 1.7e^{-(0.873 - 0.883t)}$

TABLE 2. Definitions and Lung Burden Equations for Pu and ^{241}Am

I. Regions of Interest: Count Rates, R (cps)

14 to 25 KeV: R_1 (room background removed)
51 to 70 KeV: R_2 (room background removed)
75 to 100 KeV: R_3 (room background removed)

II. Net Rates, N (cps): Room and Body Backgrounds Removed

$N_{17} = R_1 - K_1R_3$; without ^{241}Am present

$N'_{17} = N_{17} - PN_{60}$; with ^{241}Am present

$N_{60} = R_2 - K_2R_3$

III. Lung Burden Calculations: Burden, B (nCi)

$B_{239} = N_{17} \cdot F_{239}$; No ^{238}Pu, No ^{241}Am

$B'_{239} = N'_{17} \cdot F_{239}$; No ^{238}Pu, With ^{241}Am

$B_{238} = N_{17} \cdot F_{238}$; No ^{239}Pu, No ^{241}Am

$B'_{238} = N'_{17} \cdot F_{238}$; No ^{239}Pu, With ^{241}Am

$B_{241} = N_{60} \cdot F_{241}$

Fig. 1. Distributed source transmission data for plutonium with beefsteak medium and using the photon energy region 14 to 25 KeV. The line is the composite computer-fit to both the ^{238}Pu and ^{239}Pu data.

Fig. 2. Distributed source transmission data for ^{241}Am and ^{103}Pd. Energy regions are 51 to 70 KeV and 17 to 27 KeV, respectively. The lines are the respective computer fits.

Fig. 3. Signal ratio (17 KeV/60 KeV) for ^{241}Am distributed source and beefsteak medium.

Fig. 4. Absolute calibration factors (F nCi/cps) for the in-vivo detection of ^{238}Pu, ^{239}Pu, ^{103}Pd, and ^{241}Am as a function of tissue thickness.

Fig. 1

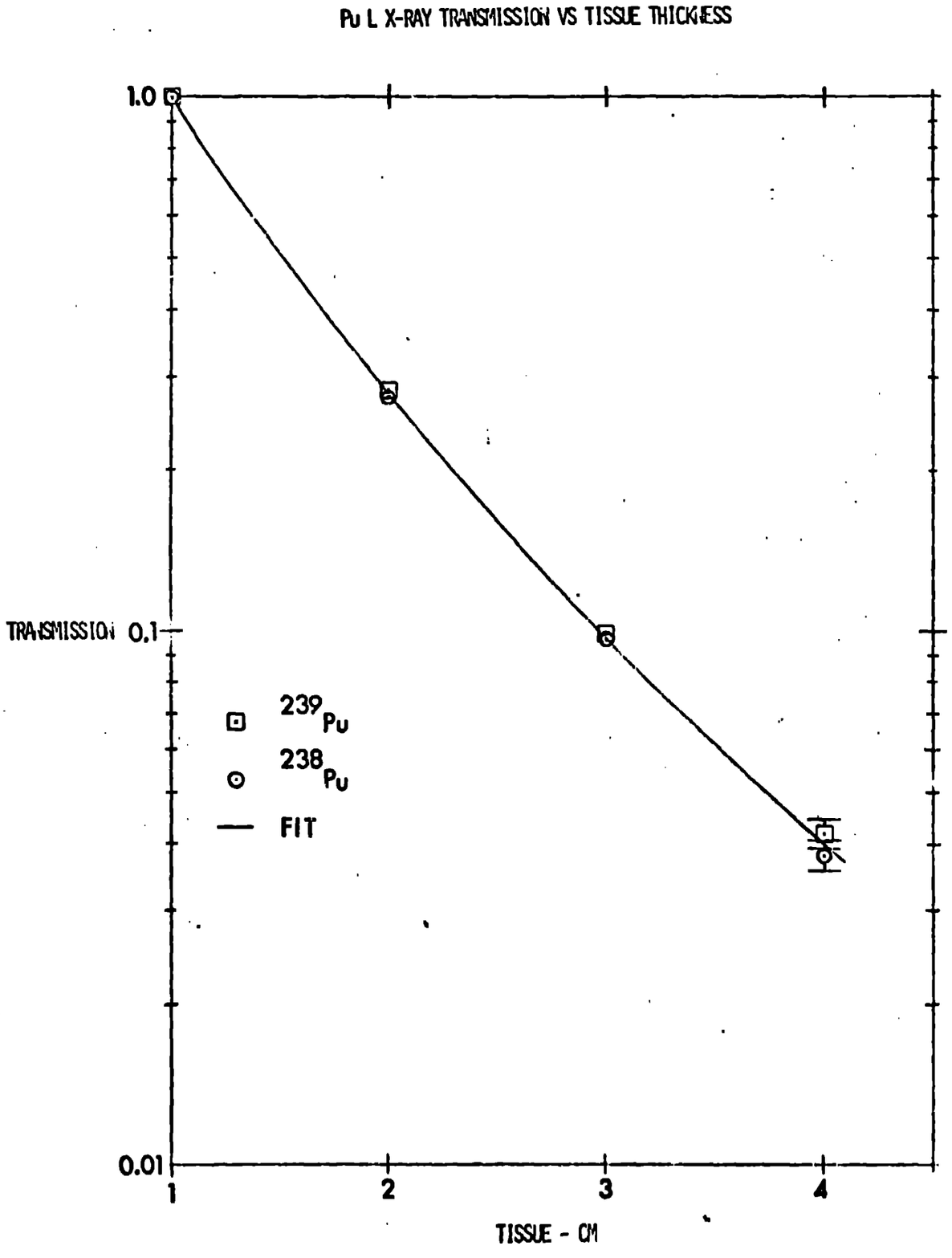

Pu L X-RAY TRANSMISSION VS TISSUE THICKNESS
1.0
0.1
0.01
TRANSMISSION
239Pu
238Pu
FIT
1
2
3
4
TISSUE - CM

Fig 2

PHOTON TRANSMISSION VS TISSUE THICKNESS

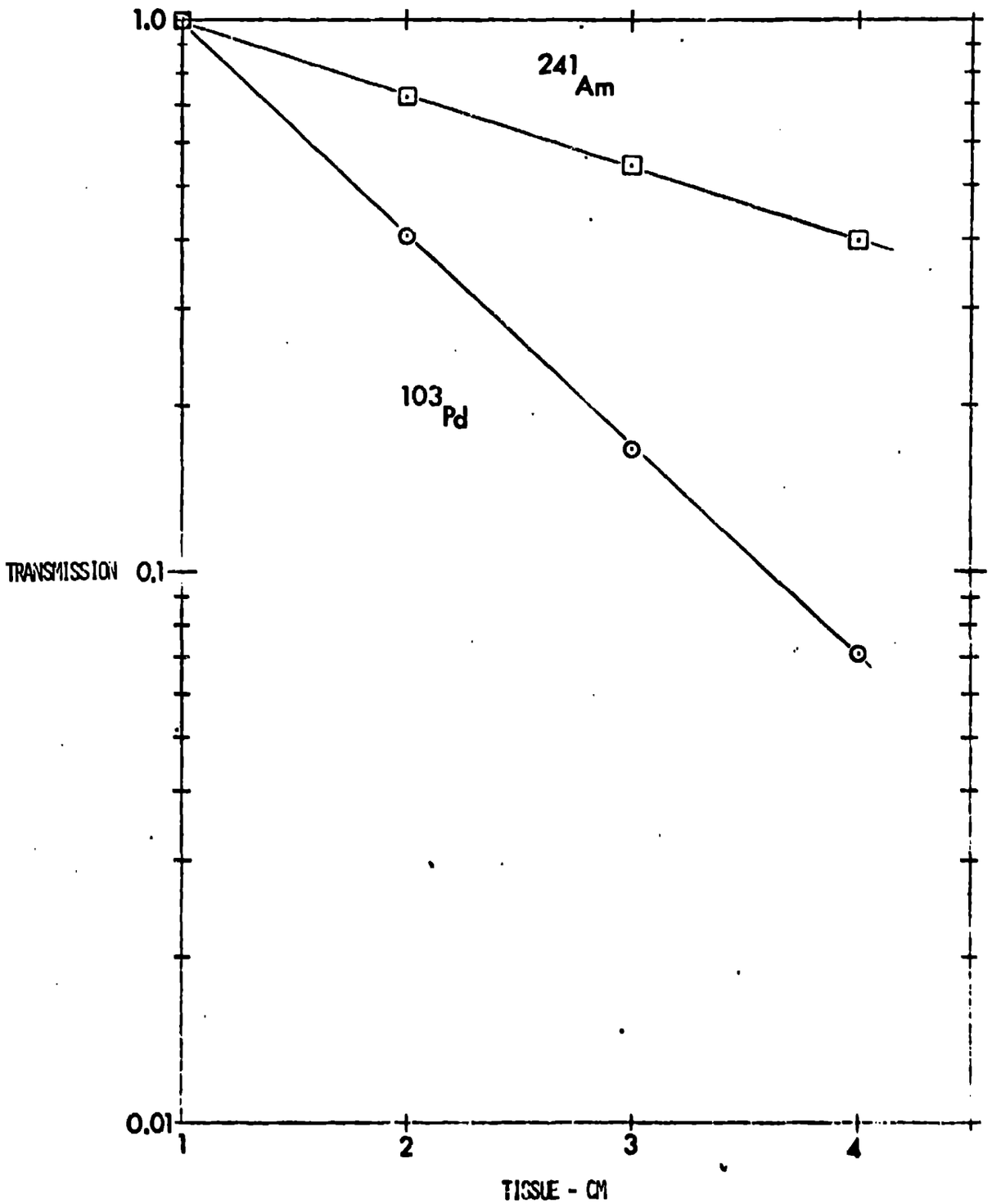
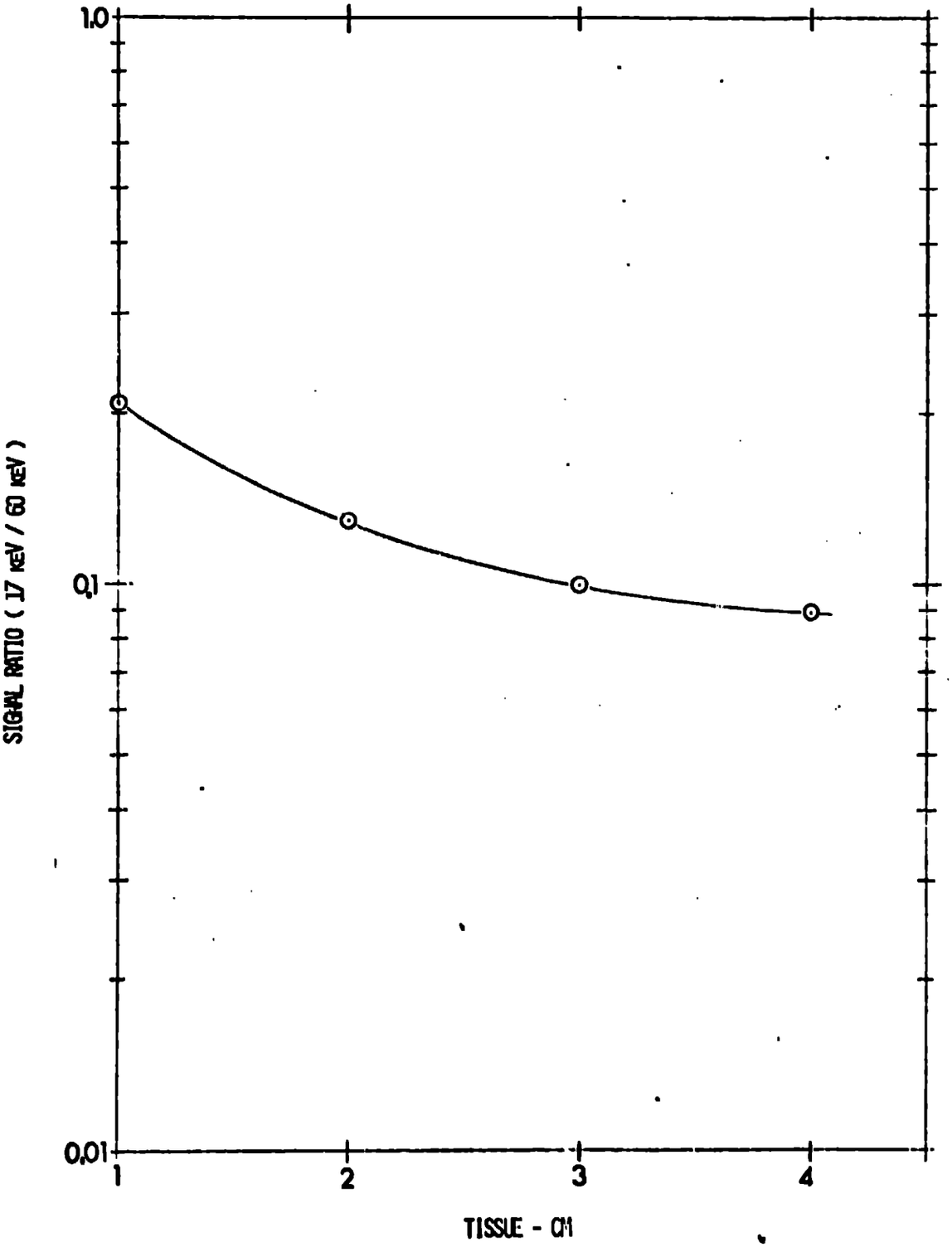


Fig 3

^{241}Am SIGNAL RATIO (17 KEV / 60 KEV) VS TISSUE THICKNESS



CALIBRATION FACTORS (F) VS TISSUE THICKNESS