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The Self-Absorption Effect of Gamma Rays in ^{239}Pu

by

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ABSTRACT

Nuclear materials assay with gamma-ray spectrum measurement is a well-established method for safeguards. However, for a thick source, the self-absorption of characteristic low-energy gamma rays has been a handicap to accurate assay. I have carried out Monte Carlo simulations to study this effect using the ^{239}Pu α-decay gamma-ray spectrum as an example. The thickness of a plutonium metal source can be considered a function of gamma-ray intensity ratios. In a practical application, gamma-ray intensity ratios can be obtained from a measured spectrum. With the help of calculated curves, scientists can find the source thickness and make corrections to gamma-ray intensities, which then lead to an accurate quantitative determination of radioactive isotopes in the material.

Introduction

Gamma-ray spectrum measurement is a well-established assay method for nuclear material safeguards. From a gamma-ray spectrum, investigators can identify different radioactive isotopes in the material. In principle, they can also determine quantities of each isotope in the material. However, for quantitative measurement this procedure requires a well-calibrated detector system whose use is restricted to an ideal thin and point radioactive source.

In many practical cases, the source could be a slab of unknown thickness. In such a case, the self-absorption of gamma rays in the material will distort the spectrum and may lead to inaccurate quantitative determinations.

Monte Carlo Simulations

I used the computer code CYLTRAN of the integrated TIGER series of Coupled Electron/Photon Monte Carlo Transport Codes[1] for simulations of the self-absorption effect in a plutonium decay gamma ray spectrum. CYLTRAN combines a condensed-history electron Monte Carlo technique with a conventional single-scattering photon Monte Carlo technique. The code simulates the transport of all generations of electrons and protons with energy between several megavolts and 1.0 keV. The electron transport includes energy-loss straggling, multiple electron scattering, production of knock-on electrons, continuous bremsstrahlung, characteristic x rays, and annihilation radiation. The photon transport includes photoelectric, Compton, and pair-production interactions.

Slabs of bare plutonium metal with varying thicknesses and a density of 19.8 gm/cm^3 were modeled as sources. In the model a detector was located 10 cm from the source surface, and a 2-cm-thick lead collimator between the source and the detector limited the source area to 7.5 cm in diameter.

Gamma rays with selected energies were originated uniformly throughout the source volume and transported isotropically toward the detector. I tallied the spectrum at the detector front surface to avoid alteration of the spectrum by the detector responses. If there were no self-absorption, the number of gamma rays tallied at the detector front surfaace would be proportional to the source thickness. For a given detector, a tally of the pulse-height distribution at the detector can also be compared with the real measured spectrum.

Figure 1 is a plot of gamma-ray intensities as a function of source thickness. The straight line represents the gamma-ray intensity with no self-absorption effect. The deviation from the straight line, as shown for three different gamma ray energies, indicates clearly the energy-dependent self-absorption effect. These curves start to level off at different thicknesses. Because all principal gamma rays in ^{239}Pu decay are low energy (< 413.7 keV), plutonium metal thicker than 5 cm is essentially considered to have infinite thickness to its own gamma rays and hence is not amenable to quantitative gamma ray assay.

The intensity ratios between different gamma rays changes as a function of source thickness up to 5 cm because of differential absorption characteristics. I calculated 6 gamma-ray intensity ratios in ^{239}Pu for 17 different thicknesses ranging from 1.0×10^{-4} cm to 5 cm. The ratios were 375 keV/413 keV, 203 keV/413 keV, 146 keV/413 keV, 129 keV/413 keV, 51 keV/413 keV, and 38 keV/413 keV.

Results and Discussion

I carried out the calculations for gamma-ray intensity ratios as functions of source thickness. A practical applicaiton would be to derive source thickness given the appropriate measured gamma-ray intensity ratios. Figures 2–7 show plots of the source thickness as functions of different gamma-ray intensity ratios. In each plot, ◇ are calculated values that can be fit by a function of the form

$$\ln T = \Sigma a_i (\ln R)^i,$$

where T is the thickness and R is the gamma-ray intensity ratio. The dots (●) in the plots are 50 interpolations of the function. Calculations are available from the author.

Although each gamma-ray intensity ratio exhibits sensitivity within certain limits of thickness, using the six ratios permits determination of thicknesses ranging from 5×10^{-4} to 5 cm. For an illustration, the measured intensity ratio of 129 keV/413 keV is 2.0 ± 0.1 and that of 203 keV/413 keV is 0.305 ± 0.020. The interpolations of the plots show that the values of the thickness are $(5.06 \pm 0.46) \times 10^{-[illegible]}$ cm and $(4.90 \pm 0.15) \times 10^{-[illegible]}$ cm respectively. The results are consitent with each other.

Figure 8 is a plot of the intensity correction factor for the 413.7-keV gamma ray as a function of plutonium sample thickness. In the plot ◇ are calculated values and ● are 50 interpolations. Calculations are available from the author. The curves in Figure 8 allow corrections for gamma-ray intensities and determination of the ^{239}Pu quantity in the source. For a thickness of $(5.00 \pm 0.10) \times 10^{-[illegible]}$ cm, the intensity of the 413.7-keV gamma ray should be increased by a factor of 1.072 ± 0.008 to obtain the true source intensity.

Plutonium-239 samples usually contain other plutonium isotopes: ^{238}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am. Determining the amounts of these isotopes is possible by knowing their decay rate and measuring the gamma-ray intensity ratios: 125 keV/146 keV for $^{241}Am/^{239}Pu$, 148 keV/146 keV for $^{241}Pu/^{239}Pu$, 152 keV/146 keV for $^{238}Pu/^{239}Pu$, and 160 keV/146 keV for

$^{240}Pu/^{239}Pu$. However, for samples thicker than 0.01 cm, the differential absorptions will change these ratios, and corrections factors will be required to obtain the true ratios and to calculate the true contents of these isotopes.

Figure 9 shows plots of these correction factors for gamma-ray intensity ratios as a function of sample thickness. No correction is needed for the 148 keV/146 keV ratio. These correction factors approach constant values for sample thickness larger than 0.25 cm; that is, 0.25 cm is approaching the infinite thickness for gamma rays with energy $\leq$ 160 keV. For the example above, the measured intensity ratios 125 keV/146 keV, 152 keV/146 keV, and 160 keV/146 keV require multiplication by factors of 1.256 $\pm$ 0.020, 0.941 $\pm$ 0.005, and 0.890 $\pm$ 0.006, respectively, to obtain the true ratios.

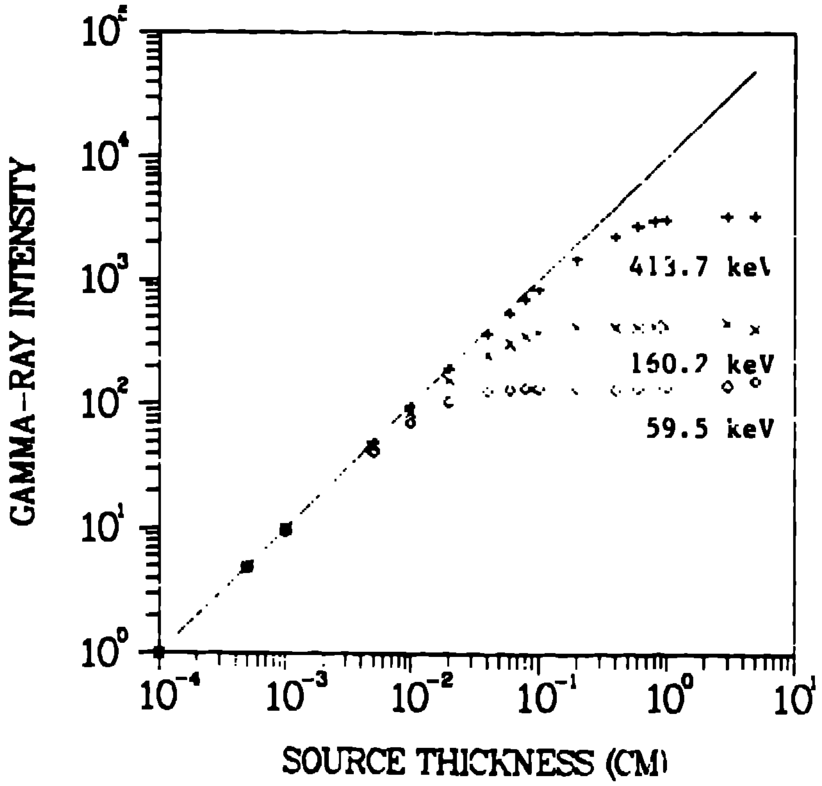
Conclusion

The self-absorption effect, usually considered a handicap in radioactive isotope quantitative assay from gamma-ray spectra, can instead in many cases be used to determine the source thickness with the help of careful Monte Carlo simulations. Such simulations, however, **must** incorporate the actual geometry of the experiments.

REFERENCES

1. J. A. Halbleib and T. A. Mehlhorn, "ITS: The Integrated TIGER Series of Coupled Electron/Photon Monte Carlo Transport Codes," Sandia National Laboratories report SAND84-0573 (1984).

2. M. R. Schmorak, "Nuclear Data Sheets for A = 231, 235, 239," Nuclear Data Sheets, Vol. 40 (1), 1983.



GAMMA-RAY INTENSITY
SOURCE THICKNESS (CM)
413.7 keV
160.7 keV
59.5 keV

Figure 1.

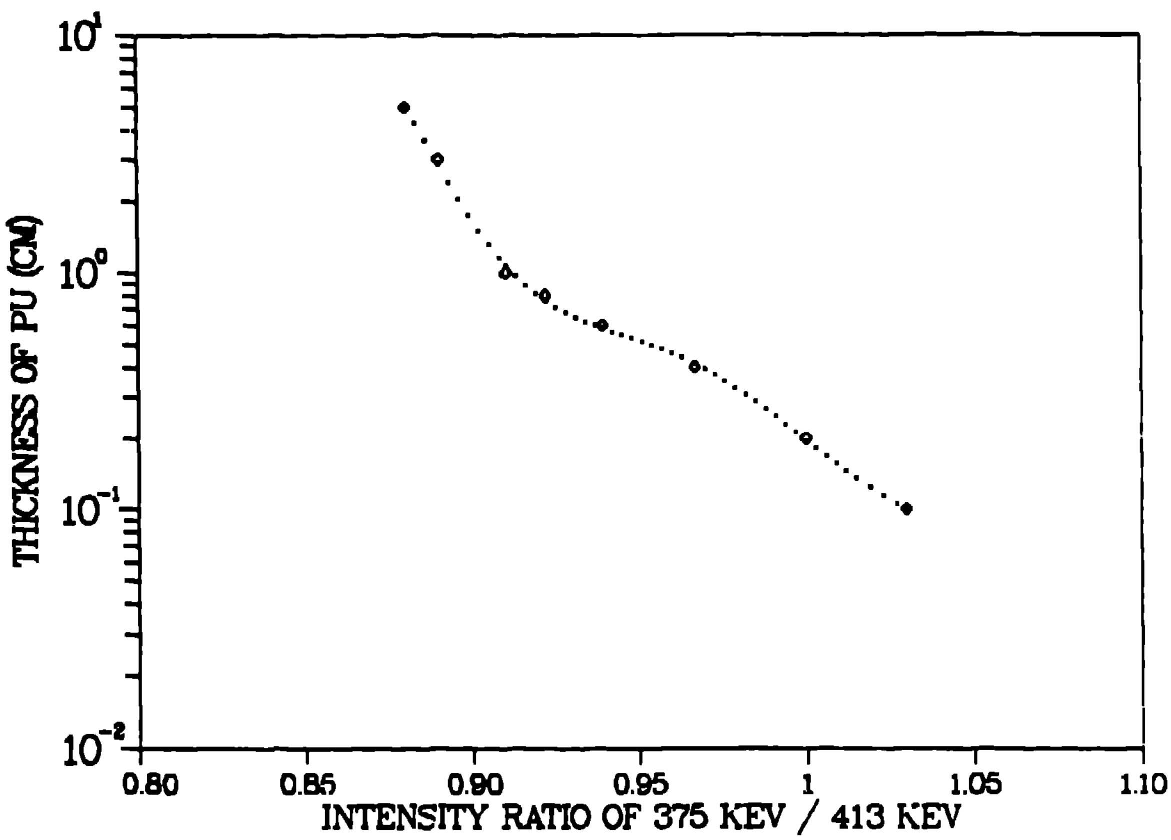


Figure 2 Sample thickness vs intensity ratio 375/413

Thin source ratio = 1.060 ± 0.008 [2]

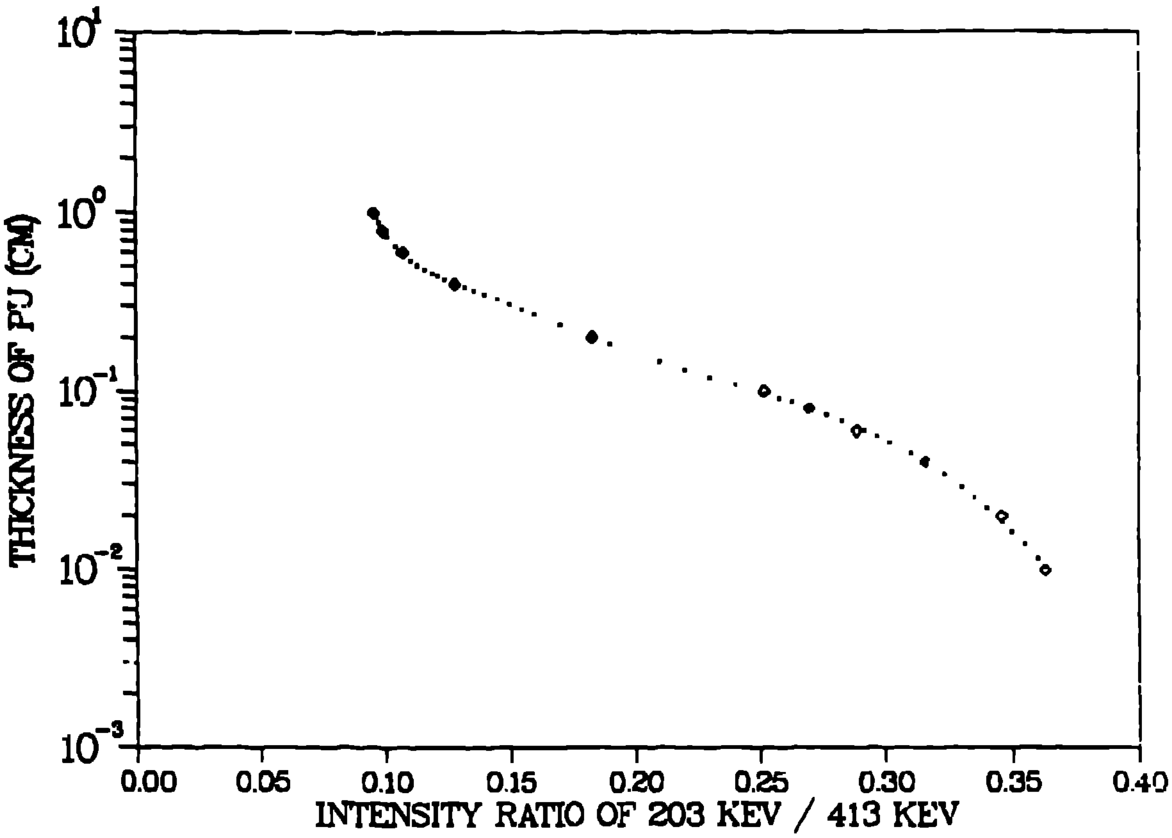


Figure 3 Sample thickness vs intensity ratio 203/413

Thin source ratio = 0.388 ± 0.003[2]

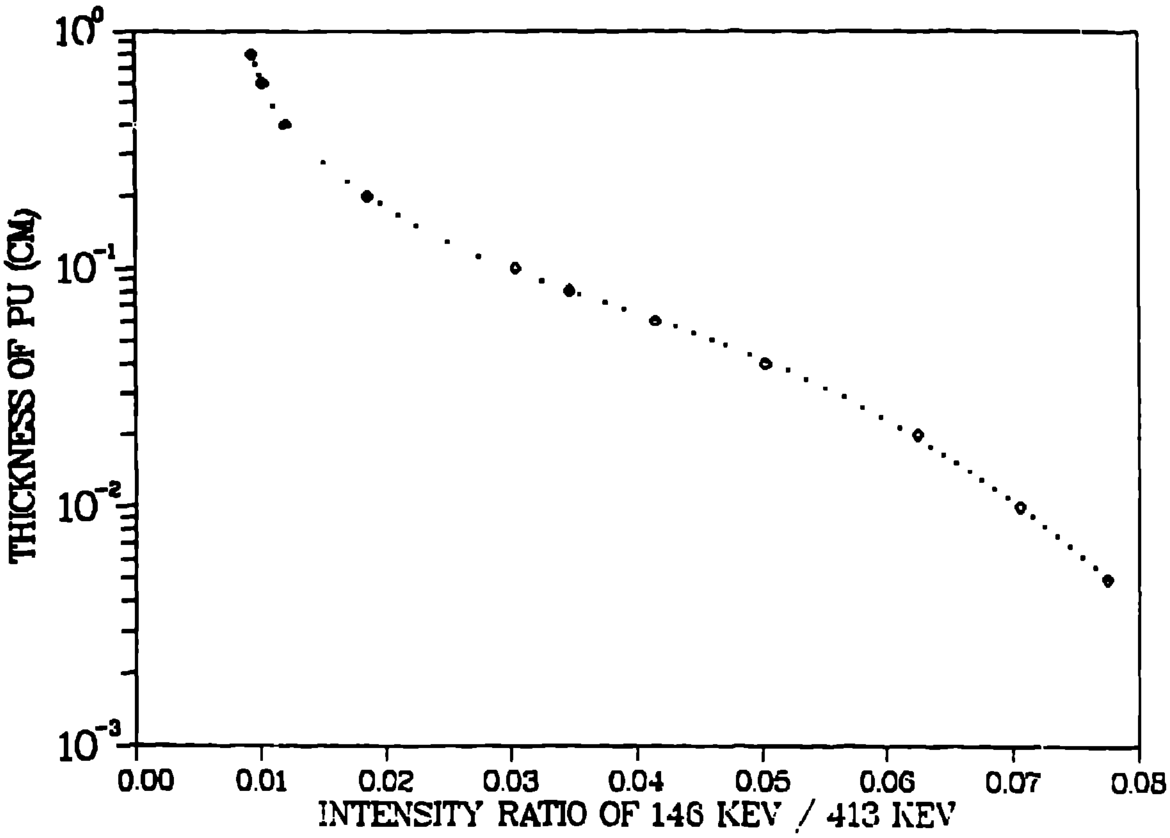


Figure 4 Sample thickness vs intensity ratio 146/413.

Thin source ratio = 0.08/2 ≈ .0018 (2)

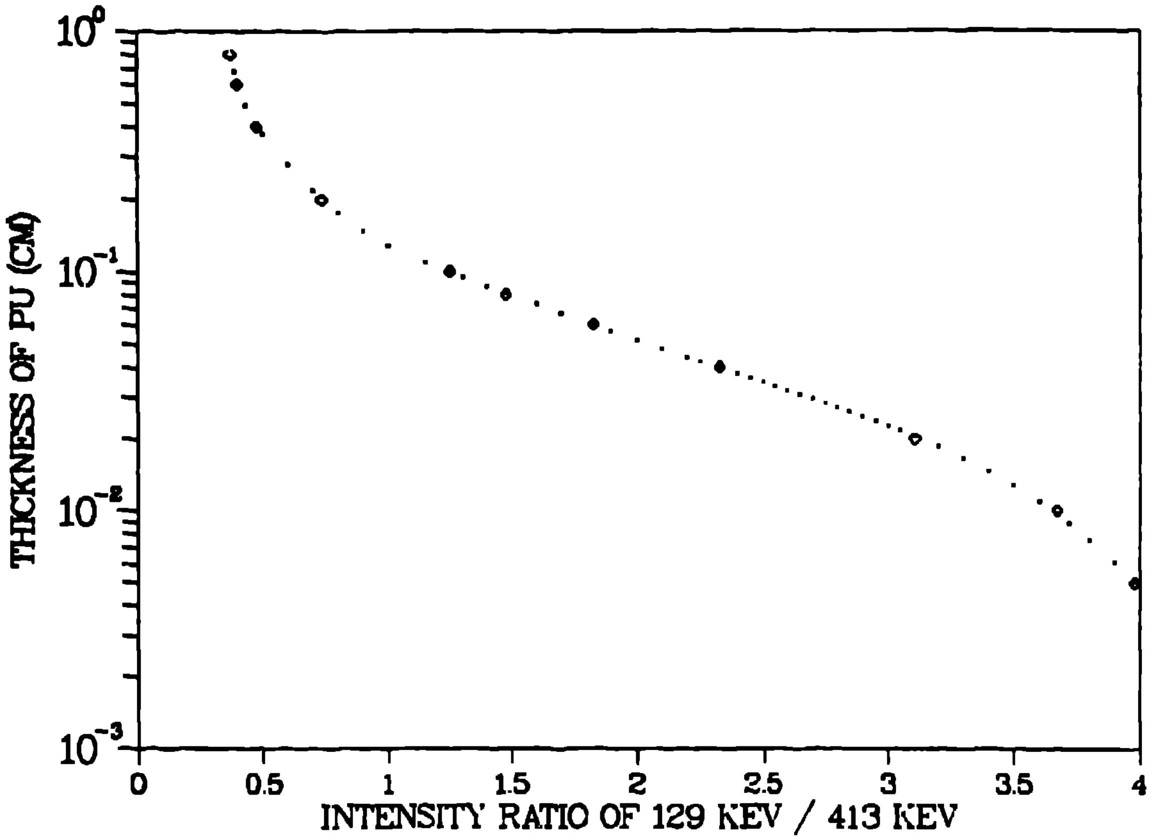


Figure 5 sample thickness vs intensity ratio 129/413

the source ratio = 4.304 ± 0.052 [2]

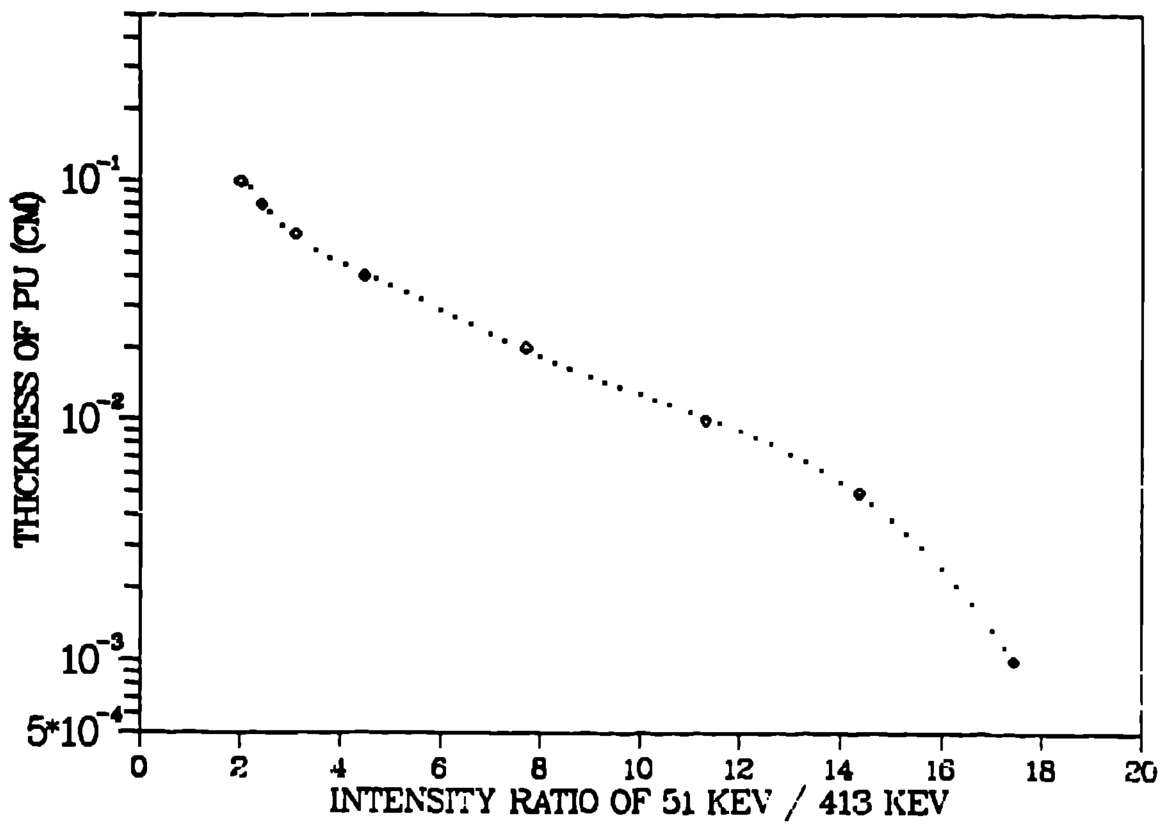


Figure 6 Sample thickness vs intensity ratio 51/413

thin source ratio = 18.48 ± 0.35

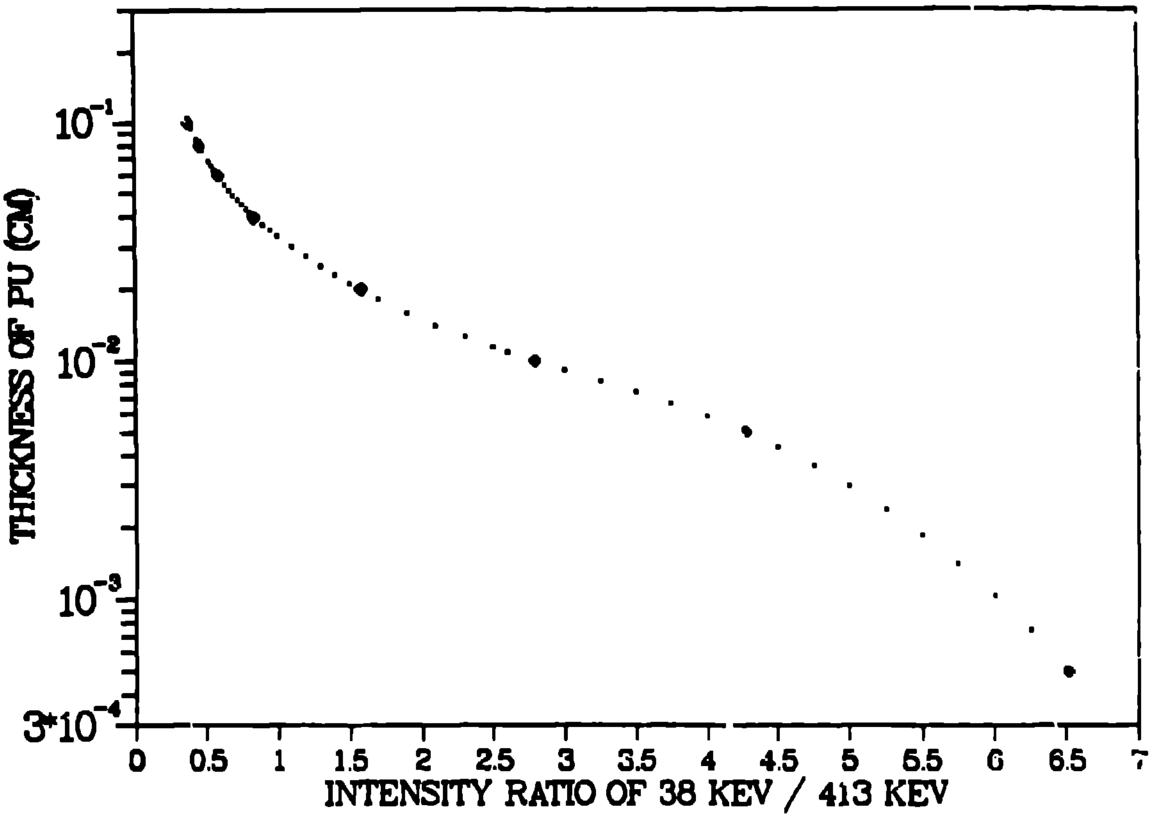


Figure 7 Sample thickness vs intensity ratio 38/413

Thin source ratio - 7.162 = 0.143 (2)

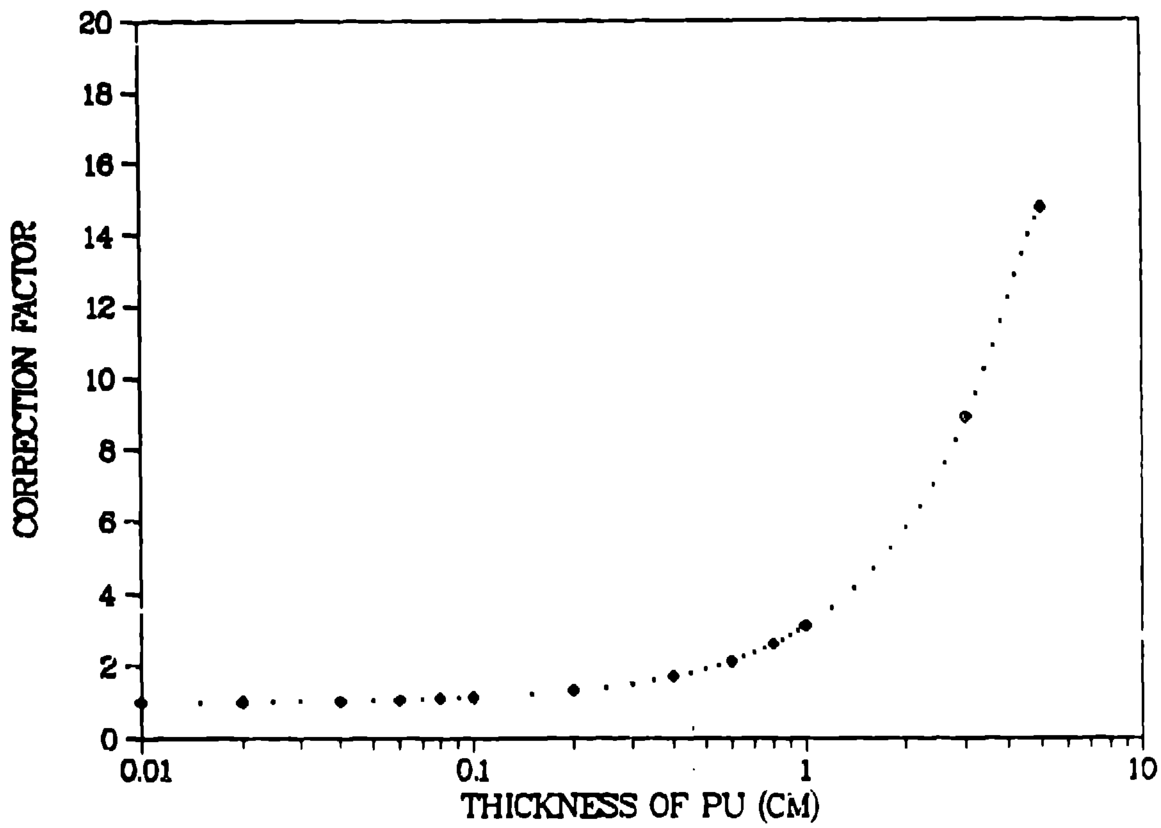


Figure 8   413.7 keV gamma ray intensity
correction factor vs sample thickness

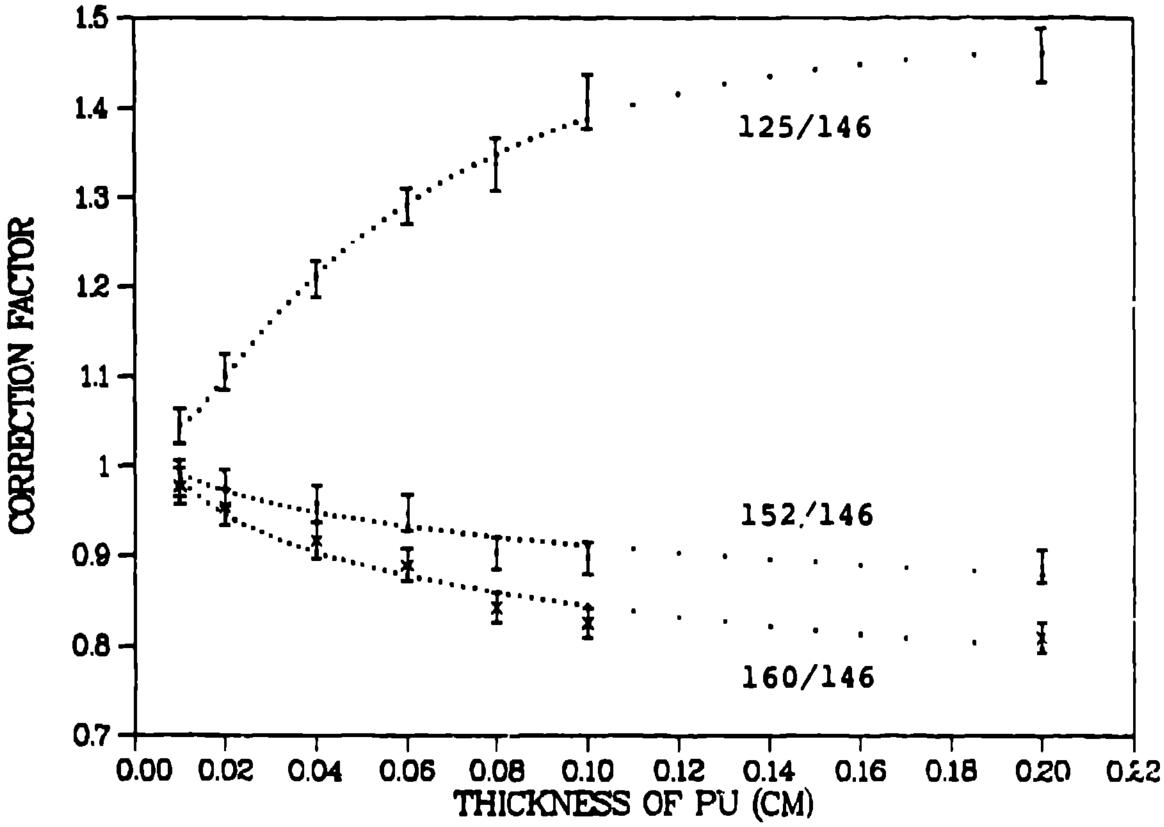


Figure 7. Intensity ratio correction factor vs sample thickness