Evaluations of the $^{235}\text{U}/^{238}\text{U}$ Isotope Ratio through Thick-Walled Containers using the 185.7 keV and 1001.0 keV Gamma rays

R. G. Lanier, C. F. Hayden, D. Clark, W. E. Parker

April 13, 2001

U.S. Department of Energy

Lawrence
Livermore
National
Laboratory

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This work was performed under the auspices of the U. S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

This report has been reproduced directly from the best available copy.

Available electronically at <http://www.doc.gov/bridge>

Available for a processing fee to U.S. Department of Energy
And its contractors in paper from
U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
Telephone: (865) 576-8401
Facsimile: (865) 576-5728
E-mail: reports@adonis.osti.gov

Available for the sale to the public from
U.S. Department of Commerce
National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone: (800) 553-6847
Facsimile: (703) 605-6900
E-mail: orders@ntis.fedworld.gov
Online ordering: <http://www.ntis.gov/ordering.htm>

OR

Lawrence Livermore National Laboratory
Technical Information Department's Digital Library
<http://www.llnl.gov/tid/Library.html>

Evaluations of the $^{235}\text{U}/^{238}\text{U}$ Isotope Ratio through Thick-Walled Containers using the 185.7 keV and 1001.0 keV Gamma rays

Robert G. Lanier, Catherine F. Hayden, DeLynn Clark and Winifred E. Parker

*Analytical and Nuclear Chemistry Division
Lawrence Livermore National Laboratory, Livermore, CA 94550*

Introduction

The non-destructive determination of isotope ratios (NDA)¹ present in plutonium- and uranium-bearing materials is best accomplished with a MultiGroup Analysis² methodology on spectral data obtained with high-resolution germanium detector systems. This methodology is currently employed in codes such as MGA, MGAU or U235. The MGA code, for example, works well on Pu samples and employs a number of isolated spectral lines in the analysis. The algorithm uses Gaussian and Lorentzian peak shapes that are empirically modified to account for various physical and instrumental effects. Additional effects due to external absorbers, sample self-attenuation and detector relative efficiency are also included. Uranium samples are analyzed with MGAU and U235 using a simpler but similar algorithm in the narrow energy range between 85- and 100-keV.

Although the details can be complex, the problem described above is ultimately reduced to evaluating a simple ratio. Applied to uranium, for example, one evaluates

¹ NDA: Non-destructive aalysis.

² See for example: *MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances*, R. Gunnink, UCRL-LR-103220, Vol.1, Lawrence Livermore National Laboratory, Livermore, CA 94550, April 3, 1990 and *MGAU: A New Analysis Code for Measuring U-235 Enrichments in Arbitrary Samples*, R. Gunnink, W.D. Ruhter, P. Miller, *et al.*, UCRL-JC-114713, Livermore, CA 94550 prepared for the IAEA Symposium on International Safeguards, Vienna, Austria, March 14-19, 1994.

$$\frac{N_{235U}}{N_{238U}} = \frac{I_A \epsilon_B}{I_B \epsilon_A} \frac{BR_B}{BR_A} \frac{t_A}{t_B} \frac{\alpha_B^{Ab}}{\alpha_A^{Ab}} \frac{\beta_B^U}{\beta_A^U}, \quad (1)$$

where the subscripts A and B refer to quantities associated with ^{235}U and ^{238}U , respectively. Specifically, N_{235U}/N_{238U} is the isotope ratio $^{235}\text{U}/^{238}\text{U}$, I is the measured gamma-ray peak intensity, and ϵ is the counting efficiency, BR is the appropriate branching ratio, t is the appropriate half-life, α^{Ab} is an external absorber correction (such as cadmium, iron, etc.) and β^U is a correction for sample self absorption. Indications of other isotopes can, of course, be determined from different ratios evaluated with data from other characteristic gamma rays that may be present in the spectrum.

In making the isotope ratio calculation using eqn. (1), the efficiency, ϵ , is expressed either as a linear or a second-degree polynomial in energy, E . Also, the sample self absorption is estimated by a term which is derived from the assumption of a line source placed along the detector axis and is given by

$$\beta = A/A_0 = \frac{1}{\mu x} \cdot (1 - e^{-\mu x}), \quad (2)$$

where μ is the absorption coefficient of the sample material [cm^{-1}], x is the source thickness [cm], A_0 is the total activity of the source³ and A is the activity measured when one end of the source is situated at the detector face. Implicit in eqn. (1) is that no geometric correction is necessary, i.e. it is assumed that different energy gamma rays originate from identical source volumes. Unfortunately, this approximation cannot be generally applicable because energy-dependent self-attenuation effects alter the effective source-detector distance for each gamma ray recorded. However, because the analysis is generally constrained to small energy regions, the geometric effect and the effects due to other limiting approximations are either small or absorbed in some average way through small variations in the large parameter space. In summary, constraining the analysis to a small energy range is the critical feature which allows MGA (for Pu) and MGAU or U235 (for U) techniques to be "self-calibrating"; i.e. using a suitable detector system, the analysis can be completed with data from a single measurement.

³ $A_0 = m \cdot a_0$, where m is the mass [gm] of the source and a_0 is its specific activity [number of gamma rays/sec/gm]. To evaluate the attenuation correction, it is assumed that A_0 is a mono-energetic source so that μ is single valued at energy, E .

While the uranium analysis algorithm described above has been very successful for a number of applications, it is limited to that class of containers that are defined as "thin", i.e. containers that allow for gamma-ray analysis in the ~100 keV region. If for example, a $^{235}\text{U}/^{238}\text{U}$ ratio is desired from a sample enclosed in a "thick-walled" container, the measurement technique described above is not possible because the 85 - 100-keV region will be either very weak or not measurable because of absorption by the container wall. With reasonable count times of thirty minutes or less, only two gamma-ray lines (from a thick-walled container) offer the potential for providing isotope ratio evaluations over the full range of Uranium enrichments encountered in practical MC&A problems. These lines are the 185-keV gamma ray from the decay of ^{235}U and the 1001-keV gamma ray⁴ from the decay of a ^{238}U daughter product. For these lines, the simple assumptions described above are not applicable.

In the following discussion we formally analyze the problem of determining isotope ratios from measured spectra. The analysis provides a derivation of a more complete and strictly accurate form of eqn. (1), isolates the important parameters in isotope-ratio calculations, and finally, yields a better sense of what is possible and what is not possible in these sorts of analyses. We conclude our discussion with the analysis of some idealized uranium measurements and a commentary on the problem of determining the $^{235}\text{U}/^{238}\text{U}$ ratio from the 185 keV and 1001 keV gamma rays.

The Formal Problem

The $^{235}\text{U}/^{238}\text{U}$ isotope ratio in a sample can be determined by measuring the gamma-ray intensities emitted from individual isotopes that escape the material. For uranium sources confined in "thin-walled" containers (i.e. < ~8 mm/ 0.3"), the $^{235}\text{U}/^{238}\text{U}$ ratio can be obtained by analyzing the 85 - 100 keV region of a HPGe⁵ gamma/x-ray spectrum. For thicker containers, this method fails because the 85 - 100 keV region is significantly attenuated and the analysis has unacceptably large errors.

⁴ In fact, an additional line at 766-keV from the daughter ^{238}U product might also be measured reasonably well when the 1001-keV line is prominent; however, this additional datum would not provide sufficiently more new information to alter the conclusions of this paper.

⁵ HPGe: "High-purity Germanium". Refers to a high-resolution germanium detector system.

In principle, when sufficient source material is present and when the ^{238}U decay chain is in equilibrium, an alternate analysis can be done using gamma rays at 185.715 keV from ^{235}U and at 1001.03 keV from $^{234\text{m}}\text{Pa}$, where the latter isotope is a radioactive daughter of ^{238}U . In this case, the range of container thickness can be extended to a nominal⁶ value of $\sim 13.2 \text{ mm} / 0.52 \text{ in.}$

Unfortunately, the energy between the two gamma rays for the alternate analysis (815 keV) is sufficiently large that special care must be exercised in attempting to make a realistic isotope ratio-determination. In order to illuminate the issues associated with such an analysis, we first solve the formal NDA problem in the general case. We then discuss the metrology problems for the particular case of $^{235}\text{U}/^{238}\text{U}$ ratios in thick-walled containers.

Assume that a radioactive isotope is homogeneously distributed through an arbitrary sample matrix, and that it emits a gamma ray of energy E as part of its normal decay chain. Then, a radiation detector (placed at an arbitrary distance from the sample) measures $\eta(E)$, the number of gamma rays of energy E in time Δt . The experimental result⁷, $\eta(E)$, may be expressed in the following way:

$$\eta(E) = N \cdot f_B(E) \cdot P_N(\lambda_i) \cdot P_{Det}(E) \cdot P_A(E) \cdot P_\tau(E) \cdot P_\Omega(E) \cdot \Delta t \quad (3)$$

where

N is the number of atoms of the radioactive isotope of interest present in the sample,

$f_B(E)$ is the branching ratio of the measured gamma ray (i.e. that fraction of the atoms at the end of the decay chain that decay with characteristic energy E),

$P_N(\lambda_i)$ is the probability per unit time that an atom decays. This quantity depends on where in the decay chain the gamma ray of characteristic energy

⁶ This value represents approximately 1.5 mean-free paths in elemental iron for the 185.7 keV gamma ray.

⁷ $\eta(E)$: i.e. the area or number of counts under a gamma-ray peak in the spectral distribution.

E is emitted. For a single-step decay, (i.e. a one step parent – daughter decay, $i = 1$), one is left with the simple form

$$P_M(\lambda_i) \equiv P_M(\lambda) = \lambda$$

where the decay constant $\lambda = 0.693/t_{1/2}$, with $t_{1/2}$ being the half-life of the parent. For multi-component decay chains, $P_M(\lambda_i)$ must be evaluated using the Bateman equations⁸,

$P_{Det}(E)$ is the probability that the detector will record an energy E when the gamma ray of energy E reaches the face of the detector (i.e. the intrinsic detector efficiency),

$P_A(E)$ is the probability that the gamma ray will penetrate any intervening absorber and is given by $e^{-\mu x}$, where μ is the mass attenuation coefficient for the absorber material and x is the absorber thickness,

$P_\epsilon(E)$ is the gamma ray "penetrability" and expresses the probability that a gamma ray, once emitted from an atom interior to the source, will reach the source boundary. That is, the penetrability measures the probability that the gamma ray will exit the source and become available for detection with its full energy. And finally,

$P_\Omega(E)$ is a solid angle term and measures the probability that a given gamma ray will emerge from a sample atom with an orientation that will allow it to impact the detector face.

If a sample described above contains two different radioactive isotopes and if each is identified with a characteristic gamma ray, E and E' , then applying eqn. (3) for each gamma-ray and taking the ratio we have:

$$\frac{\eta(E)}{\eta(E')} = \left(\frac{N}{N'} \right) \cdot \left(\frac{f_B(E)}{f_B(E')} \right) \cdot \left(\frac{P_M(\lambda_i)}{P_M(\lambda_i')} \right) \cdot \left(\frac{P_{Det}(E)}{P_{Det}(E')} \right) \cdot \left(\frac{P_A(E)}{P_A(E')} \right) \cdot \left(\frac{P_\epsilon(E)}{P_\epsilon(E')} \right) \cdot \left(\frac{P_\Omega(E)}{P_\Omega(E')} \right) \quad (4)$$

⁸ See for example *Introduction to Nuclear Physics and Chemistry*, Bernard Harvey, Prentice-Hall Inc., Englewood Cliffs, NJ, pp 34-39 (1962).

Eqn. (4) formally solves the general problem of determining the isotope ratio, N/N' , from a measurement of $\eta(E)/\eta(E')$ and is identical to eqn. (1), in the limit when the two gamma rays emitted are essentially of the same energy or when one has a source with a small spatial extension.

The Specific Case of $^{235}\text{U}/^{238}\text{U}$

Consider the case of ^{235}U and ^{238}U where $E = 185.7$ keV and $E' = 1001$ keV. Then one needs to analyze the following decay chains:

where the individual λ_i are the characteristic decay constants calculated from the half-lives and $f_B(E)$ and $f_B(E')$ are the branching ratios for the 185.7- and 1001-keV lines, respectively. Thus, for ^{235}U

$$P_N(\lambda) = \lambda_1,$$

and using the Bateman equation for ^{238}U evaluated at equilibrium, we have

$$P_N(\lambda'_i) = \lambda'_1 \lambda'_2 \lambda'_3 / ((\lambda'_2 - \lambda'_1) \cdot (\lambda'_3 - \lambda'_1)) \cong \lambda'_1$$

because λ'_2 and $\lambda'_3 \gg \lambda'_1$. Therefore⁹

$$(P_N(\lambda_i)/P_N(\lambda'_i)) \cong \lambda_i/\lambda'_i \tag{5}$$

where¹⁰

⁹ In these and subsequent calculations, the nuclear data are subject to adjustment if better or more consistent measurements of these parameters are available.

^{235}U	$\lambda_1 = 9.848 \times 10^{-10} \text{ y}^{-1}$
^{238}U	$\lambda'_1 = 1.551 \times 10^{-10} \text{ y}^{-1}$
^{234}Th	$\lambda'_2 = 1.050 \times 10^{-4} \text{ y}^{-1}$
^{234}Pa	$\lambda'_3 = 3.115 \times 10^{-5} \text{ y}^{-1}$

For the gamma-ray branching ratios¹¹

$$f_B(E) = 5.739 \times 10^{-1}$$

$$f_B(E') = 8.371 \times 10^{-3}$$

Therefore, from eqn. (4), solving for N/N' and using (5) we obtain

$$\left(\frac{N}{N'} \right) = \left(\frac{\eta(E)}{\eta(E')} \right) \cdot \left(\frac{f_B(E')}{f_B(E)} \right) \cdot \frac{\lambda'_1}{\lambda_1} \cdot \left(\frac{P_{\text{Det}}(E')}{P_{\text{Det}}(E)} \right) \cdot \left(\frac{P_A(E')}{P_A(E)} \right) \cdot \left(\frac{P_r(E')}{P_r(E)} \right) \cdot \left(\frac{P_\Omega(E')}{P_\Omega(E)} \right) \quad (6)$$

or, using the less-cluttered notation of eqn (1), and recalling that $\lambda = 0.693/t_{1/2}$,

$$\frac{N}{N'} = \frac{I}{I'} \cdot \frac{BR'}{BR} \cdot \frac{t_{1/2}}{t'_{1/2}} \cdot \frac{\epsilon' \alpha'}{\epsilon \alpha} \cdot \frac{\beta'}{\beta} \cdot \frac{\gamma'}{\gamma}, \quad (6a)$$

Replacing primed and unprimed quantities with A and B, respectively, and using other obvious identifying notations, we have

$$\frac{N_{^{235}\text{U}}}{N_{^{238}\text{U}}} = \frac{I_A}{I_B} \cdot \frac{BR_B}{BR_A} \cdot \frac{t_A}{t_B} \cdot \frac{\epsilon_B}{\epsilon_A} \cdot \frac{\alpha_B^{Ab}}{\alpha_A^{Ab}} \cdot \frac{\beta_B^U}{\beta_A^U} \cdot \frac{\gamma_B^U}{\gamma_A^U}. \quad (7)$$

¹⁰ *Table of Radioactive Isotopes*, Edgardo Browne and Richard B. Firestone, with Virginia S. Shirley, Editor, John Wiley and Sons, New York, NY (1986).

¹¹ See Table I in *Uranium Isotopic Analysis with the FRAM Isotopic Analysis Code*, Duc T. Vo and Thomas E. Sampson, LA-13580, Los Alamos National Laboratory Technical Report, Los Alamos, NM 87544 (May 1999).

While eqns. (1) and (7) are virtually identical, they differ because of an added term

$$\frac{\gamma_B^U}{\gamma_A^U}$$

which identifies a geometric correction that becomes progressively more important when the source has a significant spatial extension and when the energy separating the two gamma rays becomes significantly different. For the $^{235}\text{U}/^{238}\text{U}$ ratio, we can insert the appropriate constants noted above and arrive at the final form:

$$\frac{N_{235\text{U}}}{N_{238\text{U}}} = 2.296 \times 10^{-3} \frac{I_A}{I_B} \frac{\epsilon_B}{\epsilon_A} \frac{\alpha_B^{Ab}}{\alpha_A^{Ab}} \frac{\beta_B^U}{\beta_A^U} \frac{\gamma_B^U}{\gamma_A^U} \quad (8)$$

where A and B refer to ^{235}U and ^{238}U , respectively. For large sources, the source geometry is intimately connected with sample self-absorption so, in practice, the term represented by the ratios

$$\frac{\beta_B^U}{\beta_A^U} \frac{\gamma_B^U}{\gamma_A^U} \quad (9)$$

must be obtained by considering these effects together. A reasonable evaluation of eqn. (9) is a complicated endeavor and requires a multiple integral over the surface coordinates¹² of the detector and all the volume elements of the source. For the discussion below we assume a simple form for eqn. (9) which, nevertheless, allows us to explore how far one can go in determining isotope ratios using gamma rays separated by a "large" energy difference.

¹² Even this is an approximation because each surface element along the detector face does not detect gamma rays with the same efficiency. However, in the discussion that follows, we have assumed that we could experimentally determine the detector efficiency in some average way which would then allow us to say that we "know the detection efficiency of a gamma ray" once it encounters the detector at any point.

Discussion

It is instructive to apply the correlation equation

$$\left(\frac{N_{235U}}{N_{238U}} \right)_{\text{Measured}} = \beta \left(\frac{N_{235U}}{N_{238U}} \right)_{\text{Standard}} \quad (10)$$

to a series of measurements employing uranium standards. In eqn. (10) the left-hand side is the evaluation of the isotope ratio using the experimental data with eqn. (8) (the experiments measure the ratio $I_A/I_B = I_{235}/I_{238}$) and the right hand side is the "book value", i.e. the isotope ratio quoted for the standard. If all the terms shown in eqn. (8) are determined properly, then a plot of eqn. (10) for a series of known standards will yield a straight line with unit slope ($\beta = 1$) and zero intercept.

A series of measurements was done on uranium standards placed at various distances from a high-resolution germanium detector¹³. The emitted radiation was collimated through a 5 cm-thick lead collimator¹⁴ with a 3-cm diameter opening. Counting times ranged from one to several hours but were selected so that the statistical errors on the extracted peak areas were of the order of 1% or less. For an estimate of the combined sample-self-absorption- and geometric-correction terms, we also evaluated¹⁵

¹³ The "detector face" mentioned here is the end-cap of the aluminum vacuum housing that contains the germanium detector. An additional ~3mm separates the actual detector face from the aluminum housing. The detector is a right circular cylinder with a height of 53.2 mm and a diameter of 34.3 mm.

¹⁴ Even though a fair fraction of the 1001 keV gamma rays penetrate the 5 cm-thick collimator used, it was thought that this arrangement would better define the relative efficiency curve comparing both the 185 and 1001 keV gamma rays. That is, detector edge effects would be reduced so that the efficiency curve would be more or less constant along the collimator opening. Figure 1 shows that the actual "collimator profile" for the two gamma rays studied is quite variable when the source is much larger than the collimator opening. This information was later used to make an additional "Relative Slit Penetration Correction" noted in Tables I – VI.

¹⁵ See, for example, Robley D. Evans and Richard O. Evans, *Revs. Mod. Phys.* **26**, 305 (1948) or "The Use of Calibration Standards and the Correction for Sample Self-Attenuation in Gamma-Ray Nondestructive Assay", J.L. Parker, LA-10045, Los Alamos National Laboratory, Los Alamos, NM 87545, August (1984).

$$\beta_U \gamma_U \equiv A/A_0 = \frac{b^2}{2\ell} \int_{-\ell}^{\ell} \frac{e^{-\mu x}}{(b-x)^2} dx \quad (11)$$

where ℓ is the length of an idealized linear source, b is the distance between the source center and the detector face, μ is the self-absorption term of the source material and x is a coordinate measured with respect to the center of the source. Eqn. (11) is the correct form¹⁶ for a point detector and a line source. Applied to the measurements described here the "Integral Correction" represented by eqn. (11) appears to be only a small improvement over the "Bulk Correction" of eqn. (2) when ratios are determined. However as the source-detector separation closes, eqn. (11) would be expected to be a better approximation to the physical situation because the count rate has an inverse-square dependence.

The efficiency correction term, ε , was experimentally determined using a thin, small-diameter ¹⁵²Eu source.¹⁷ This source has a number of strong reference lines nicely spanning the range between 185 and 1001 keV. Using this source, an additional correction was also made for the relative size of the collimator opening.

Figure 1 displays the experimental data used to determine the correction for collimator penetration effects, which were later used in the isotope ratio determinations. The correction is necessary because this effect causes the collimator opening to appear larger for higher energy gamma rays relative to those of lower energy. The correction was experimentally determined by taking a series of Eu standard spectra with the source placed at various vertical distances relative to the center-line of the collimator-detector system. The data were then energy-renormalized to represent corrections between the 185- and 1001-keV gamma rays. The correction applied to the

¹⁶ For $\mu = 0$, eqn. (11) reduces to $I/I_0 = b^2 / (b+\ell)(b-\ell)$, which shows that effective source-detector distance for an extended uniformly distributed source is the geometric mean of the distances measured between the two extreme ends of the source: $(b+\ell)$ is the end closest to the detector while $(b-\ell)$ is the end of the source furthest from the detector.

¹⁷ Efficiencies were established through measurements of a Eu source set a distance of 31 cm from the detector face. The source is virtually a "point source" and was also used to "map" the collimator opening.

present data is simply the ratio of the appropriate relative areas¹⁸ under each curve in Figure 1.

Figure (2) through (7), display the correlation plot [eqn (10)] established from the measurements taken at selected source-detector separations. Statistical error bars are not shown but are less than the size of the data points. The data show reasonable isotope ratio determinations up until the source comes in contact with the collimator. That is, the data suggest that by properly treating the source as finite and by reasonably characterizing the experimental arrangement, it is possible to get a credible measure of the $^{235}\text{U}/^{238}\text{U}$ isotope ratio using the 185 and 1001 keV lines at source-detector separations greater than ~ 10 cm. However, additional measurements need to be made on larger diameter sources to determine better limits on where the approximation of a line source fails and perhaps uncover other limitations in the analyses.

A question could be raised about whether or not the present analysis might have been more transparent if a collimator were not used. In effect, the collimator causes the detector to see an energy-degraded gamma-radiation flux which is corrected by the transmission curve shown in Figure 1. Superficially, the addition of a collimator appears as an unnecessary complication. However, a bare detector would seem to be more difficult to characterize for larger-sized sources. This is because, absent a collimator, one would still need to define detector efficiency response as a function of displacement from the detector center-line to accommodate sources with relative differences in the area that each presents to the detector.

The failure of the analysis at small source-detector distances is probably due to a combination of factors. Among the more important is penetration of the collimator by the 1001-keV gamma ray. While the correction derived from the data in Figure 1 would be expected to suitably account for this effect, its proper application depends intimately on the geometry of the source. Simply applying it as a multiplicative correction to the measured intensity cannot be expected to represent the true physical situation. An additional problem involves the finite-source model used – the "Integral Correction". Approximating the experimental arrangement as a line source viewed by a point detector - as eqn. (11) implies - is clearly an inadequate approximation

¹⁸ The integration limits depend on the source-detector separation because the source extends a finite distance in the vertical direction. As the source is placed closer to the detector, the integration limits are extended vertically until the vertical distance is larger than the diameter of the detector.

when the source-detector separation is small. The correct procedure for such a situation is to properly integrate over the volume elements of the source, the detector surface, and to include collimator penetration as well.

Finally, it is interesting to remark that the "Bulk Correction" – represented by eqn. (2) – did a good job reproducing the expected isotope ratios. While the physical situation does not justify the use of eqn. (2), it nevertheless seems to provide a credible estimate of self-attenuation effects if one can tolerate errors in the final results of the order of $\pm 10\%$. This is true even when the detector is in contact with the collimator. Moreover, because the eqn. (2) correction is easily applied to the experimental results, a technique might be developed for obtaining 10% isotope ratio numbers from one measurement by using an additional ^{238}U gamma-ray at 766 keV. A further study would need to determine whether the required *a priori* information to make such an analysis is more relaxed than what was necessary for the present exercise.

Conclusions

The details presented here for determining isotopic ratio of $^{235}\text{U}/^{238}\text{U}$ include an efficiency calibration, as well as a calculation of the ratio of effective solid angle and penetrability for gamma rays of 185 keV and 1001 keV. It is clear that even with the ideal conditions of the measurements and the quality of the data obtained, the techniques employed fail when the source-detector separation is small. The reason for this is likely due to collimator penetration by the 1001 keV gamma ray and because finite-source effects are not properly taken into account. Nevertheless, to be guaranteed of getting the "correct" $^{235}\text{U}/^{238}\text{U}$ ratio from a gamma ray measurement using only the 185 and 1001 keV lines, one would have to know the detailed physical makeup of the source and properly determine all the implications of those details. For "close in geometries", this means that the size of the source has to be correctly taken into account and all the detailed integrations properly performed. Doing this properly would require more information than appears practical.

To meet the IAEA requirements, a measurement should be complete in thirty minutes or less. For uranium in thick-walled containers, in a count time of thirty minutes, only two lines are clearly seen, the 185- and the 1001-keV lines. These lines are too far apart in energy to allow for their relative ratio determination without significant knowledge about the source.

Therefore, the conclusion based on the present data is that too much information is required to extract a reasonable $^{235}\text{U}/^{238}\text{U}$ ratio from the 185- and 1001-keV lines. Furthermore, this applies to any technique which involves an arbitrary uranium sample and which is based solidly on all the physics of the problem. While future study might uncover ad hoc recipes for isotope-ratio determinations involving these lines, none of these techniques can be generally reliable when applied outside the range of their confirmed applicability.

In general, increasing the measurement time to observe more ^{238}U gamma rays on which to base an analysis is not sufficient to overcome the physical situation presented by the problem. While the development of empirical recipes using more spectral lines is always possible, these techniques are limited both by the number of gamma ray lines required and by their energy distribution in the spectra. In addition, the results from such analyses, when taken outside the range of their known applicability, will always be questionable. Finally and just as important, measurements will only be as good as the user's ability to recognize when measurements are being made outside such limiting boundaries.

In summary, the physics of the $^{235}\text{U}/^{238}\text{U}$ isotope ratio problem demands certain information for a reasonable solution if only the 185- and 1001-keV gamma rays are used. The techniques used here to extract the $^{235}\text{U}/^{238}\text{U}$ isotope ratio are not practical for field use.

Figures & Tables

Figure 1: Relative transmission of a 5cm-thick lead collimator through a 3cm diameter opening for the 185- and 1001- keV gamma rays. The open squares plot the 185 keV transmission and the open circles display the transmission trend for the 1001-keV gamma ray. Note that the abscissa scale is shown in millimeters. The data plotted represent a three-point smooth of the raw data.

Figure 2: Correlation plot for atom ratio determinations at a source-detector separation of 31 cm. The "ideal" line (line with slope = 1) is the expected trace if the all aspects of the analysis are taken properly into account. The broken lines are least-square fits through the measured data with different finite source correction recipes. The "integral correction" is given by eqn. 11 and the "bulk correction" is given in eqn. 2.

Figure 3: Correlation plot for atom ratio determinations at a source-detector separation of 21 cm. See Figure 2 caption.

Figure 4: Correlation plot for atom ratio determinations at a source-detector separation of 16 cm. See Figure 2 caption.

Figure 5: Correlation plot for atom ratio determinations at a source-detector separation of 16 cm. A repeat measurement. See Figure 2 caption.

Figure 6: Correlation plot for atom ratio determinations at a source-detector separation of 11 cm. See Figure 2 caption.

Figure 7: Correlation plot for atom ratio determinations at a source-detector separation of 5.2 cm. These data represent an experimental arrangement where a 5.2 cm thick lead collimator in contact with both the detector face and front surface of the standard source. See Figure 2 caption.

Table I: Data for a source-detector separation of 31 cm. The columns are labeled to correspond to the terms identified in eqn. 7. An additional term, "the collimator transmission correction" is more properly included in the geometric term, γ^U , if a more exact calculation had been done to take account of source extent. In such a calculation, collimator penetration would be an included effect. Eqn. 11 includes no collimator penetration effects so this correction was determined by experimental measurement.

Table: 31 cm Data											
Measured I_A Intensity 185 keV	Δ_A Intensity Error	^{235}U Enrichment [%]	λ_A [yr ⁻¹]	ER_A Branching Ratio	ϵ_A Efficiency Factor	α_A^{Ab} Aluminum Attenuation	Relative Slit Penetration Correction ²	$\beta_A^U \gamma_A^U$ Integral Correction	Cumulative Corrections [Int. Corr.]	$\beta_A^U \gamma_A^U$ Bulk Correction	Cumulative Corrections [Bulk Corr.]
87166	630	0.3206	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1827	2.55E-13	1.555E-01	2.46E-13
191981	717	0.7209	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1827	2.55E-13	1.555E-01	2.46E-13
57039	266	1.9864	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1827	2.55E-13	1.555E-01	2.46E-13
46219	228	2.9557	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1827	2.55E-13	1.555E-01	2.46E-13
93542	266	4.5168	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1854	2.44E-13	1.527E-01	2.40E-13
		Quoted/Atom Fraction Ratio		Calculated $^{235}\text{U}/^{238}\text{U}$ Enrichment Ratio [Int. Corr.]	Percent Difference [Quoted vs Calc]		Calculated $^{235}\text{U}/^{238}\text{U}$ Enrichment Ratio [Bulk Corr.]	Percent Difference [Quoted vs Calc]			
		0.00322		0.00331	2.80%		0.00341	5.97%			
		0.00726		0.00725	0.12%		0.00749	3.09%			
		0.02006		0.01992	3.77%		0.01994	0.61%			
		0.03079		0.03039	1.30%		0.03137	1.87%			
		0.04733		0.04731	0.04%		0.04846	2.39%			
				Avg. Dev	1.8%		Avg. Dev	2.8%			
Measured I_B Intensity 1001 keV	Δ_B Intensity Error	^{235}U Enrichment [%]	λ_B [yr ⁻¹]	ER_B Branching Ratio	ϵ_B Efficiency Factor	α_B^{Ab} Aluminum Attenuation	Relative Slit Penetration Correction ²	$\beta_B^U \gamma_B^U$ Integral Correction	Cumulative Corrections [Int. Corr.]	$\beta_B^U \gamma_B^U$ Bulk Correction	Cumulative Corrections [Bulk Corr.]
73971	294	99.683	1.551E-10	8.371E-03	0.000579	0.9574	1.010	0.8325	7.17E-16	8.370E-01	7.21E-16
74310	236	99.274	1.551E-10	8.371E-03	0.000579	0.9574	1.010	0.8325	7.17E-16	8.370E-01	7.21E-16
8296	98	98.016	1.551E-10	8.371E-03	0.000579	0.9574	1.010	0.8325	7.17E-16	8.370E-01	7.21E-16
4270	70	96.983	1.551E-10	8.371E-03	0.000579	0.9574	1.010	0.8325	7.17E-16	8.370E-01	7.21E-16
3313	61	95.44	1.551E-10	8.371E-03	0.000579	0.9574	1.014	0.8361	7.14E-16	8.316E-01	7.19E-16
Notes: 1. The ratio N_{235}/N_{238} in eqn. 8 is equal to ratio of fractional enrichments listed in column C											
2. In a more proper calculation slit penetration correction would be part of the geometric term $\beta^U \gamma^U$											

Figure II: Data for a source-detector separation of 21 cm. See Table I caption.

Table II: 21 cm Data										
Measured I_A Intensity 185 keV	Δ_A Intensity Error	^{235}U Enrichment ¹ [%]	λ_A [yr ⁻¹]	BR_A Branching Ratio	ϵ_A Efficiency Factor	α_A^{Ab} Aluminum Attenuation	Relative Slit Penetration Correction ²	$\beta_A^U \gamma_A^U$ Integral Correction	Cumulative Corrections [Int. Corr.]	$\beta_A^U \gamma_A^U$ Bulk Correction
9606	140	0.3206	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1643	2.59E-13	1.566E-01
47043	261	0.7209	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1643	2.59E-13	1.566E-01
83084	312	1.9664	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1643	2.59E-13	1.566E-01
86424	310	2.9657	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1643	2.59E-13	1.566E-01
105520	341	4.5168	9.846E-10	5.739E-01	0.00298	0.9339	1.000	0.1663	2.46E-13	1.527E-01
				Calculated ^{235}U Enrichment Ratio [Int. Corr.]	Percent Difference [Quoted vs Calc]		Calculated ^{235}U Enrichment Ratio [Elk. Corr.]	Percent Difference [Quoted vs Calc]		
		Quoted Atom Fraction Ratio								
		0.00322		0.00332	3.27%		0.00349	8.08%		
		0.00726		0.00739	1.79%		0.00776	6.61%		
		0.02006		0.01971	1.79%		0.02068	3.02%		
		0.03079		0.03078	0.01%		0.03230	4.81%		
		0.04733		0.04698	4.05%		0.05116	7.78%		
				Ave. Dev	2.2%		Ave. Dev	6.1%		
Measured I_B Intensity 1001 keV	Δ_B Intensity Error	^{235}U Enrichment ¹ [%]	λ_B [yr ⁻¹]	BR_B Branching Ratio	ϵ_B Efficiency Factor	α_B^{Ab} Aluminum Attenuation	Relative Slit Penetration Correction ²	$\beta_B^U \gamma_B^U$ Integral Correction	Cumulative Corrections [Int. Corr.]	$\beta_B^U \gamma_B^U$ Bulk Correction
8398	96	99.663	1.551E-10	8.371E-03	0.000579	0.9574	1.033	0.8269	7.50E-16	8.370E-01
16460	141	99.274	1.551E-10	8.371E-03	0.000579	0.9574	1.033	0.8269	7.50E-16	8.370E-01
12252	115	99.016	1.551E-10	8.371E-03	0.000579	0.9574	1.033	0.8269	7.50E-16	8.370E-01
8158	93	96.983	1.551E-10	8.371E-03	0.000579	0.9574	1.033	0.8269	7.50E-16	8.370E-01
6518	85	95.44	1.551E-10	8.371E-03	0.000579	0.9574	1.033	0.8198	7.47E-16	8.316E-01
Notes: 1. The ratio $N_{^{235}\text{U}}/N_{^{238}\text{U}}$ in eqn. 8 is equal to ratio of fractional enrichments listed in column C										
2. In a more proper calculation, slit penetration correction would be part of the geometric term $\beta^U \gamma^U$										

Copyright © 2009 John Wiley & Sons, Ltd.

23

Figure IV: Data for a source-detector separation of 16 cm. A repeat measurement. See Table I caption.

Table IV. 16cm Data (repeat)										
Measured I_A Intensity 165 keV	Δ_A Intensity Error	^{235}U Enrichment [%]	λ_A [yr ⁻¹]	EP_A Branching Ratio	ϵ_A Efficiency Factor	α_A^{Ab} Aluminum Attenuation	Relative Self Penetration Correction ²	β_{AY}^U Integral Correction	Cumulative Corrections [Int. Corr.]	β_{AY}^U Bulk Correction
92484	123	0326	984E+10	573E01	00028	0989	1.00	0162	25E+13	158E01
10944	36	0729	984E+10	573E01	00028	0989	1.00	0162	25E+13	158E01
18258	47	1964	984E+10	573E01	00028	0989	1.00	0162	25E+13	158E01
27231	54	2957	984E+10	573E01	00028	0989	1.00	0162	25E+13	158E01
29125	30	4518	984E+10	573E01	00028	0989	1.00	0166	24E+13	152E01
		Quoted Atom Fraction Ratio		Calculated ^{235}U Enrichment Ratio [Int. Corr.]	Percent Difference [Quoted vs Calc]		Calculated ^{235}U Enrichment Ratio [Bulk Corr.]	Percent Difference [Quoted vs Calc]		
		00022		00029	234%		00031	864%		
		00026		00039	178%		00087	807%		
		00006		00008	011%		002139	642%		
		00009		00057	070%		00325	561%		
		00433		00497	291%		00481	203%		
				As Div	18%		As Div	62%		
Measured I_B Intensity 1001 keV	Δ_B Intensity Error	^{235}U Enrichment [%]	λ_B [yr ⁻¹]	EP_B Branching Ratio	ϵ_B Efficiency Factor	α_B^{Ab} Aluminum Attenuation	Relative Self Penetration Correction ²	β_{BY}^U Integral Correction	Cumulative Corrections [Int. Corr.]	β_{BY}^U Bulk Correction
83482	93	9863	155E+10	837E03	00059	0954	1.1157	0819	78E+16	830E01
44619	219	9274	155E+10	837E03	00059	0954	1.1157	0819	78E+16	830E01
27339	171	9806	155E+10	837E03	00059	0954	1.1157	0819	78E+16	830E01
26847	171	9693	155E+10	837E03	00059	0954	1.1157	0819	78E+16	830E01
20381	145	9544	155E+10	837E03	00059	0954	1.1251	08115	78E+16	831E01

Notes: 1. The ratio N_{235}/N_{238} in eqn 8 is equal to ratio of fractional enrichments listed in column C.

2. In an improved calculation self penetration correction would be part of the geometric term β_{BY}^U .

Figure V: Data for a source-detector separation of 11 cm. See Table I caption.

Table V. 11cm Data										
Measured I_A Intensity 165keV	Δ_A Intensity Error	^{235}U Enrichment [%]	λ_A [yr ⁻¹]	EP_A Barthling Ratio	ϵ_A Efficiency Factor	α_A^{Ab} Aluminum Attenuation	Relative Self Penetration Correction ²	β_{AY}^U Integral Correction	Cumulative Corrections [Int. Corr.]	β_{AY}^U Bulk Correction
36669	811	03206	984E+10	573E-01	000288	09339	1.000	01682	259E+13	1588E-01
16740	484	07209	984E+10	573E-01	000288	09339	1.000	01682	259E+13	1588E-01
38665	674	19664	984E+10	573E-01	000288	09339	1.000	01682	259E+13	1588E-01
30462	588	29657	984E+10	573E-01	000288	09339	1.000	01682	259E+13	1588E-01
25451	557	45168	984E+10	573E-01	000288	09339	1.000	01688	246E+13	1527E-01
				Calculated ^{235}U Enrichment Ratio [Int. Corr.]	Percent Difference [Quoted vs Calc]		Calculated ^{235}U Enrichment Ratio [Bulk Corr.]	Percent Difference [Quoted vs Calc]		
		Quoted Atom Fraction Ratio								
		000822		000886	134%		000855	998%		
		000726		000728	024%		000794	888%		
		000806		000823	081%		000805	948%		
		000879		000842	203%		000846	1063%		
		000733		000637	422%		000827	1128%		
				Ave Dev	17%		Ave Dev	101%		
Measured I_B Intensity 101keV	Δ_B Intensity Error	^{235}U Enrichment [%]	λ_B [yr ⁻¹]	EP_B Barthling Ratio	ϵ_B Efficiency Factor	α_B^{Ab} Aluminum Attenuation	Relative Self Penetration Correction ²	β_{BY}^U Integral Correction	Cumulative Corrections [Int. Corr.]	β_{BY}^U Bulk Correction
36662	640	99668	155E+10	837E-03	000659	09574	12316	08008	841E+16	8370E-01
7450	283	99274	155E+10	837E-03	000659	09574	12316	08008	841E+16	8370E-01
6080	255	98016	155E+10	837E-03	000659	09574	12316	08008	841E+16	8370E-01
3144	184	96988	155E+10	837E-03	000659	09574	12316	08008	841E+16	8370E-01
17609	137	9544	155E+10	837E-03	000659	09574	12412	07905	837E+16	8316E-01
Notes: 1. The ratio N_{235}/N_{238} in eq 8 is equal to ratio of fractional enrichments listed in column C										
2. In our proper calculation self penetration correction would be part of the geometric term β^{UY}										