



An Assessment of γ -ray Emission Probabilities in the Decay of ^{238}U

M. Shamsuzzoha Basunia

*Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California
94720, USA. e-mail: SBasunia@lbl.gov*

Aaron M. Hurst

*Department of Nuclear Engineering, University of California Berkeley, Berkeley,
California 94720, USA. e-mail: AMHurst@berkeley.edu*

ABSTRACT

We have deduced the 49.55- and 113.6-keV γ -ray emission probabilities in the decay of ^{238}U from the intensity balance of α and γ transitions feeding and depopulating the levels of ^{234}Th . The deduced value of the 49.55-keV γ -ray emission probability is in good agreement with most of the data available in the literature. A precision of 1.4% has been obtained, which is significantly better compared to the earlier values. The 113.6 γ -ray emission probability of this work is in excellent agreement with one of the literature values, however, the precision of 14% remains comparable to the earlier one. This work highlights the need to remeasure the emission probability of the 49.55-keV γ ray to obtain a high-confidence measurement, given that there is significant spread in the current data by as much as 10%. From this work, we also recommend to remeasure the energy of the 49.55-keV γ ray for improved reconciliation between the deduced and directly measured data.

Keywords: ^{238}U decay; Gamma-ray emission probability; Alpha emission probability

1. Introduction

Gamma-ray emission probability, P_γ , is widely used in nuclear research, nuclear engineering, medical, and metrology applications. Different measurement techniques and methods have been used to obtain P_γ data, i.e. directly or from other types of measured quantities such as X-ray, annihilation radiation, β , α , etc. [1-7]. In recent decades, the development and use of microcalorimeter detectors [8,9] opens up the possibility of faster analysis of actinide isotopes for environmental, nuclear reactor, and nuclear safeguards applications. To implement this technique, better P_γ data are needed in the 50-208 keV energy range, ideally to a precision of about 1%. Most of these data were measured or deduced using a variety of techniques and methods. However, P_γ data for low-energy γ rays are often poorly known due to the higher background, and the presence of nearby X-rays, doublets, etc. in the spectrum.

The microcalorimeter detectors offer better energy resolution and detection efficiency compared to the HPGe detectors in the low-energy regime. A new measurement-campaign is needed to remeasure some of these γ -ray emission probabilities and for that an in-depth analysis and assessment of the available data are required to guide future measurement campaigns.

In this work, we have analyzed available α -particle and γ -ray data associated with the decay of ^{238}U . These data are reviewed and the most recent α -particle data [10] have been used to deduce new γ -ray emission probabilities. The status of these data are presented along with the priority of measurements to resolve the differences.

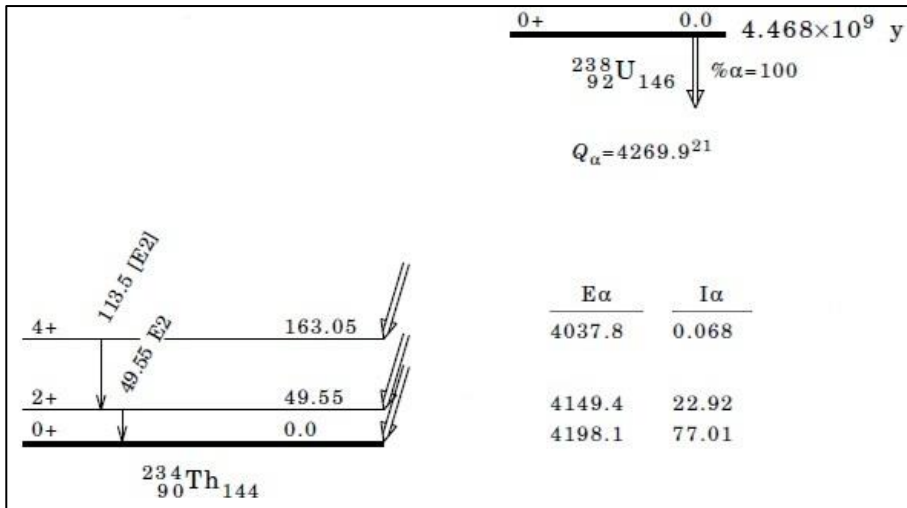


Figure 1. ^{234}Th levels populated in the decay of ^{238}U [11]. Alpha particle energy (E_α) in keV and emission probability (I_α , in the text P_α) per 100 decay of ^{238}U feeding the ground state, first, and second excited states at 0.0-, 49.55- and 163.05-keV, respectively, in ^{234}Th are shown.

2. γ -ray emission probability from direct and α measurements

The isotope ^{238}U , half-life $(4.468 \pm 0.006) \times 10^9$ years [11], decays to ^{234}Th by α -particle emission with an α -decay branching of 100%. There are three α -particle groups of energies 4198.1-, 4149.4-, and 4037.8-keV feeding the ground state, first, and second excited states, respectively, in ^{234}Th , as shown in Fig. 1. The first and second excited states are depopulated by 49.55- and 113.5-keV γ -rays, respectively. In the 1980s and 1990s, the P_γ of these γ rays was measured by relative and absolute methods. Alpha particle data of ^{238}U decay were measured earlier in the 1950s. The simple level scheme of ^{234}Th allows for the deduction of γ -ray emission probabilities from the reported α -particle intensities.

So far, only two articles report the directly measured emission probability, P_γ , for 49.55- and 113.5-keV γ -rays [12, 13]. In 1984, Roy *et al.* [12] reported the $P_\gamma(49.55)$ and $P_\gamma(113.5)$ from

a relative measurement, where three known P_γ from ^{234}Th β decay and ^{234}U α decay were used as reference. One was the $P_\gamma(63.29)$ in ^{234}Th β decay and the other two were $P_\gamma(53.20)$ and $P_\gamma(120.90)$ following ^{234}U α decay. From their work, authors report the $P_\gamma(113.5)$ for the first time, which is about 6 times weaker compared to the $P_\gamma(49.55)$. In 1990, Komura *et al.* [13], measured P_γ for the 49.55 γ ray based on an absolute method, where uranium oxide (U_3O_8) of known activity was used and the γ -ray spectrometry was performed using a well-characterized germanium detector – a LEPS (Low Energy Photon Spectrometer).

Over the decades, the P_γ data were also deduced using measured α emission probabilities. For example, the data available in the Table of Radioactive Isotopes [5] and in the Table of Radionuclides [14] were deduced using the α data reported in Refs. [15] and [16], respectively. Recently, Pomme *et al.* [10] performed a high-resolution α -decay measurement and reported P_α with improved precision. In this work, we have used their data to deduce the $P_\gamma(49.55)$ and $P_\gamma(113.5)$ from the intensity balance of P_α feeding the first and second excited states of ^{234}Th and depopulating γ -ray transition intensities as follows:

$$P_{\alpha k} = \sum_{i=1}^n (I_{\gamma i} + I_{CEi})out - \sum_{j=1}^m (I_{\gamma j} + I_{CEj})in,$$

or

$$P_{\alpha k} = \sum_{i=1}^n I_{\gamma i}(1 + CC_i)out - \sum_{j=1}^m I_{\gamma j}(1 + CC_j)in, \quad (1)$$

where $P_{\alpha k}$ is the α intensity feeding the k^{th} level, $k = 1, 2, \dots, p$; $I_{\gamma i}$ and I_{CEi} are the γ -ray branching ratios, and conversion electron (CE) intensities, respectively, of the i^{th} γ ray depopulating the k^{th} level; while $I_{\gamma j}$ and I_{CEj} are the same for the j^{th} γ ray, but populating the k^{th} level. The conversion coefficient (CC) is the ratio of the conversion electron intensity over the γ -ray intensity, i.e. I_{CE}/I_γ . The first and second terms on the right hand side in Equation (1) represent the total depopulation and population intensities, respectively, associated with the k^{th} level. Rearranging the terms of Equation (1), we get

$$NR = \frac{P_{\alpha k}}{\sum_{i=1}^n I_{\gamma i}(1 + CC_i)out - \sum_{j=1}^m I_{\gamma j}(1 + CC_j)in} \quad (2)$$

Here we define ‘NR’ as a multiplication factor to obtain γ -ray emission probability, P_γ , from the following relation

$$P_{\gamma i} = NR \times I_{\gamma i}, \quad (3)$$

The input data used to deduce the P_γ of 49.55- and 113.5-keV γ rays are listed in Table 1. Note that each of the excited levels in ^{234}Th , as can be seen in Fig. 1, depopulated by only one γ ray – so the branching ratio, $\sum_{i=1}^n I_{\gamma i}$, is 1.

Table 1. P_α , I_γ branching, and conversion coefficient (CC) for E2 multipolarity data are listed in columns 2, 3, and 4, respectively.

Energy (keV) E_α or E_γ	% P_α [10]	I_γ branching	CC [17] for E2
4038	22.92 ± 0.10		
4149	0.068 ± 0.010		
49.55 ± 0.06		1	322 ± 5
113.5 ± 0.1		1	6.49 ± 0.10

Substituting the data of Table 1 into Equations (2) and (3), we arrive at the following deduced γ -ray emission probabilities for the 49.55- and 113.4-keV γ rays: $P_\gamma(49.55)=0.071 \pm 0.001$ and $P_\gamma(113.5)=0.0091 \pm 0.0013$. Note that the second term in the denominator of Equation (2) is zero, since there are no γ rays populating the second excited state of ^{234}Th . We present all pertinent data from this work and the literature in Table 2.

Table 2. γ -ray emission probability, P_γ , of the 49.55 and 113.5 keV γ rays in ^{238}U α decay. Relative uncertainties are listed in columns 4 and 6.

Ref. (year) (type)	Method	% P_γ (49.55)	% ΔP_γ (49.55)	P_γ (113.5)	% ΔP_γ (113.5)
[5] (1986) (deduced)	using P_α [15]	0.070 ± 0.012	17	0.024 ± 0.008	33
[12] (1984) (experiment)	relative measurement	0.064 ± 0.008	12.5	0.0102 ± 0.0015	15
[13] (1990) (experiment)	absolute measurement	0.059 ± 0.002	3.4		
[14] (2006) (deduced)	using P_α [16]	0.0697 ± 0.0026	3.7	0.0174 ± 0.0047	27
This work (deduced)	using P_α [10]	0.071 ± 0.001	1.4	0.0091 ± 0.0013	14

3. Discussions

a) P_γ of 49.55 keV

As can be seen from Table 2, the deduced $P_\gamma(49.55)$ values from the P_α of Refs. [10, 15, 16] are about 0.07. From the relative measurement, a value of 0.064 ± 0.008 was obtained [12], and 0.059 ± 0.002 was reported from the absolute measurement [13]. As mentioned earlier, in the relative method, reference P_γ data of ^{234}U and ^{234}Th were used. These reference P_γ data of ^{234}U and ^{234}Th were checked for any variations compared to the current values. Minor changes were found but the influence of those changes was insignificant. So, the data from the relative measurement remain the same. The precision of $P_\gamma(49.55)$ has improved over the decades as can be seen from the data in column 4 of Table 2, where the most precise result with a 1.4% level of uncertainty is obtained from this work due to the better P_α measurements reported in Ref. [10].

The $P_\gamma(49.55)$ data in Table 2 are inconsistent, $\chi^2=7.3 > \chi^2_{\text{crit}}=2.37$, where 0.059 ± 0.002 can be identified as an outlier by Peirce's criterion [18]. Without this value the data set becomes consistent and the weighted average yields 0.071 ± 0.001 , which is the same as that of the current work. The unweighted average of all data is 0.067 ± 0.002 . Either of these two values can be accepted as a reasonable estimation of accuracy based on the available data, while $P_\gamma(49.55)=0.064 \pm 0.006$, from the relative measurement [12], is recommended in the latest evaluation [11]. This latter value closely represents the arithmetic mean and standard deviation of three data points, these are the data from relative measurement, absolute measurement, and one from the deduced values using P_α .

While the 1.4% precision of $P_\gamma(49.55)$ can be considered within the desired level of accuracy, the estimated accuracy from the weighted, unweighted, or other methods differ by as much as 10%. Note that to deduce the $P_\gamma(49.55)$ using the P_α , conversion coefficient data are needed. The calculation of CC involves many parameters including the γ -ray energy. The γ -ray energy of 49.55 ± 0.06 keV was measured in 1973 [19] using a Si(Li) detector with a 1-mm Be window. Earlier this energy was estimated to be ~ 45 keV from conversion-electron energy measurements [20], where an uncertainty of 5 keV was attributed. For the 113.6 keV γ ray, we find that $P_\gamma(113.6) = 0.0102 \pm 0.0015$ [12] is in excellent agreement with $P_\gamma(113.6) = 0.0091 \pm 0.0013$ of this work. However, our result $P_\gamma(49.55) = 0.071 \pm 0.001$ compares less favorably with $P_\gamma(49.55) = 0.064 \pm 0.008$ [12] and in disagreement with $P_\gamma(49.55) = 0.059 \pm 0.002$ [13]. Furthermore, if the true γ -ray energy is slightly different compared to the reported value of 49.55 keV, this may have a significant impact on the corresponding CC value that may lead to different overall emission probabilities. For example, one would get a CC value of 350 for a 48.70 keV electric quadrupole (E2) γ transition, which would yield $P_\gamma(48.70)=0.065$. In this scenario, the estimated accuracy would have been in excellent agreement with all the measured and deduced $P_\gamma(49.55)$ values. It would thus be very useful to also remeasure the γ -ray energy, in addition to a new measurement of the γ -emission probability for this transition.

b) P_γ of 113.5 keV

The $P_\gamma=0.0091 \pm 0.0013$ for the 113.5-keV γ ray from this work is almost the same with the value measured by relative method [10] and all together the four data points in column 5 of Table 2 represent a consistent set, mainly due to poor precision. The weighted average of all data is 0.0101 ± 0.0014 . The precision remains 14% at best because it is determined from a weak α -decay branch with higher associated uncertainty. For ^{238}U isotope analysis in the sample, it would be a less useful γ ray due to its weak intensity.

4. Conclusions

In this work, we deduced γ -ray emission probabilities of $P_\gamma(49.55)=0.071 \pm 0.001$ and $P_\gamma(113.5)=0.0091 \pm 0.0013$ for the 49.55- and 113.5-keV γ rays, respectively. The deduced value of $P_\gamma(49.55)$ in this work is in good agreement with all the published data other than Ref. [12]. A precision of 1.4% was obtained for the 49.55 keV γ -ray emission probability, which is within the desired level of accuracy for actinide isotope analysis in the sample. The

$P_\gamma(113.5)$ value deduced in this work is also in good agreement with all of the literature data. However, the precision of $P_\gamma(113.5)$ is poor and remains around 14% at best, owing to the weaker γ -ray intensity resulting from the weaker α -decay branch feeding the level.

Among the $P_\gamma(49.55)$ values, 0.059 ± 0.002 was identified as an outlier by Peirce's criterion. Without this value, the weighted average of all other data is 0.071 ± 0.001 . The unweighted average of all data is 0.067 ± 0.002 . Any of these two values can be considered as a good estimated accuracy, while the measured value of 0.064 ± 0.006 appears to be another option which has been recommended in Ref. [11]. This latter value closely represents the arithmetic mean and standard deviation of three data points: from relative method, absolute method, and one of the deduced values. For $P_\gamma(113.5)$, all the four data points are consistent and the weighted average is 0.0101 ± 0.0014 . While the estimated accuracy is robust, the poor precision would not be useful to use this datum for ^{238}U analysis in the sample.

Based on the investigation and new data from this work, we recommend to remeasure the $P_\gamma(49.55)$ emission probability and its transition energy to provide a high-confidence measurement with improved accuracy, especially considering the up to 10% spread in the estimated accuracy from the available data.

Acknowledgement

This work was supported by the Office of International Nuclear Safeguards, U.S. Department of Energy.

References

- [1] Marnada, N., *et al.* (2002). γ -ray emission probabilities of ^{193}Os , *Nucl. Instrum. Methods Phys. Res.: A*, 480, 591-597. [https://doi.org/10.1016/S0168-9002\(01\)01107-X](https://doi.org/10.1016/S0168-9002(01)01107-X)
- [2] Schötzig, U. (1990). Photon emission probabilities of ^{44}Ti , ^{65}Zn , ^{88}Y , ^{89}Sr , ^{147}Pm , ^{204}Ti and ^{210}Pb , *Nucl. Instrum. Methods Phys. Res. A*: 286, 523-528. [https://doi.org/10.1016/0168-9002\(90\)90910-X](https://doi.org/10.1016/0168-9002(90)90910-X)
- [3] D. Pérez-Loureiro, *et al.* (2016). β -delayed γ decay of ^{26}P : Possible evidence of a proton halo, *Phys. Rev.: C* 93, 064320. <https://doi.org/10.1103/PhysRevC.93.064320>
- [4] Thomas, J.C., *et al.* (2004). Beta-decay properties of ^{25}Si and ^{26}P , *Eur. Phys. J. A*: 21, 419-435.
- [5] *Table of Radioactive Isotopes*, edited by V. S. Shirley (1986), New York: Wiley.
- [6] *Evaluated Nuclear Structure Data File* (<https://www.nndc.bnl.gov/ensdf/>)
- [7] *Decay Data Evaluation Project* (http://www.lnhb.fr/home/conferences-publications/ddep_wg/)
- [8] Wollman, D.A., *et al.* (1997). High-resolution, energy-dispersive microcalorimeter spectrometer for X-ray microanalysis, *Journal of Microscopy*: 188, 196-223. <https://doi.org/10.1046%2Fj.1365-2818.1997.2670824.x>

- [9] Beck, B.R., *et al.* (2007). Energy splitting of the ground-state doublet in the nucleus ^{229}Th , *Phys. Rev. Lett.* 98, 142501.
<https://dx.doi.org/10.1103/PhysRevLett.98.142501>
- [10] Pomme, S., *et al.* (2014). High-resolution alpha-particle spectrometry of ^{238}U ; *Appl. Radiat. Isot.*: 87, 315-319. doi: 10.1016/j.apradiso.2013.11.075
- [11] Ota, S. (2026). Nuclear Data Sheets for A=234, Nuclear Data Sheets: 207, 351-506.
<https://doi.org/10.1016/j.nds..12.002>
- [12] Roy, J.C., *et al.* (1984). A study of γ -rays from the ^{238}U decay, *Int. J. Appl. Radiat. Isotop.*: 35, 899. doi: 10.1016/0020-708X(84)900280
- [13] Komura, K., *et al.* (1990). Abundance of low-energy gamma rays in the decay of ^{238}U , ^{234}U , ^{230}Th , ^{227}Ac , ^{226}Ra and ^{214}Pb , *Instrum. Methods Phys. Res. A.*: 295, 461. doi: 10.1016/0168-9002(90)90727-N.
- [14] *Table of Radionuclides*; Édité par le BIPM, Pavillon de Breteuil (2006). ISBN 92-822-2218-7: Vol. 3. <http://www.lnhb.fr/home/nuclear-data/nuclear-data-table/>
- [15] Kocharov, *et al.* (1959). Energy spectra of alpha particles from the long-lived isotopes Th^{232} and U^{238} , *Soviet Phys. JETP*: 9, 48-53.
- [16] Garcia-Torano, E. (2000). Alpha-particle emission probabilities in the decay of ^{238}U , *Appl. Radiat. Isot.*: 52, 591-594. [https://doi.org/10.1016/S0969-8043\(99\)00215-8](https://doi.org/10.1016/S0969-8043(99)00215-8)
- [17] Kibedi, T. *et al.* (2008). *Nucl. Instrum. Methods Phys. Res. A*: 589, 202-229.
10.1016/j.apradiso.2013.11.075 and <https://bricc.anu.edu.au/>
- [18] Peirce, B. (1852). Criterion for the rejection of doubtful observations, *Astronomical Journal*: 2, 45, 161-163.
- [19] Taylor, H.W. (1973). Gamma rays emitted by uranium and thorium in the energy range 10-120 keV, *Int. J. Appl. Radiat. Isotop.*: 24, 593-597. doi: 10.1016/0020-708X(73)90128-2
- [20] Dunlavey, D.C. and Seaborg, G.T. (1952). Investigations of complex structure in alpha-emission with nuclear emulsions, *Phys. Rev.*: 87, 165.
doi: 10.1103/PhysRev.87.165