

## <sup>234</sup>Th – Comments on Evaluation of Decay Data by A. Luca

*This evaluation was completed in May 2009. The literature available by December 31<sup>st</sup>, 2008 was included.*

### 1. Evaluation Procedures

The Limitation of Relative Statistical Weight (LWM) method was applied for averaging numbers throughout this evaluation; this method was implemented by using the computer code LWEIGHT, ver. 4 (designed for Excel, MS Office). The uncertainty assigned to an average value in this evaluation is never lower than the lowest uncertainty of any of the experimental input values.

### 2. Decay Scheme

<sup>234</sup>Th decays 100 % by beta minus particle emissions, mainly to <sup>234</sup>Pa<sup>m</sup> - the 1.159 min. half-life metastable state of <sup>234</sup>Pa (the first experimentally established case of nuclear isomerism, by O. Hahn, in 1921). The decay scheme was studied by many authors, since early '60s (1961Ge13, 1962Br05, 1963Bj02, 1964Ab04, 1965Fo12 and 1973Go40). The first recommended values for the main <sup>234</sup>Th nuclear decay data were published in the evaluation of Coursol et al., in 1990 (1990Co08); other important evaluation can be found in 1998Ad08. In the present evaluation, the spin, parity, energy and half-life values of the <sup>234</sup>Pa excited levels, and the multiplicities of the  $\gamma$ -ray transitions, have been adopted from the most recent A=234 ENSDF mass-chain evaluation, published by E. Browne and J.K. Tuli (2007Br04). The very important low energy and intensity isomeric transition (maximum energy of less than 10 keV) from <sup>234</sup>Pa<sup>m</sup> to the first excited level of <sup>234</sup>Pa (explaining the 73.92 keV gamma-ray transition to the <sup>234</sup>Pa ground state), was not observed yet, probably because the conversion lines are obscured by intense Auger M and Coster-Kronig electrons (according to Godart and Gizon, 1973); as a consequence, the energies of all the <sup>234</sup>Pa excited levels decaying to <sup>234</sup>Pa<sup>m</sup> are known to be upheld 10 keV at most with a systematic uncertainty (usually considered as "x" keV, in 2007Br04 and other evaluations; in the present evaluation, this quantity is not written in the decay scheme, but it should be added to the energy of the excited levels, respectively subtracted from the reported beta transitions energies). A more detailed decay scheme of <sup>234</sup>Th can be found in 2007Br04. The decay of <sup>234</sup>Pa<sup>m</sup> (by alpha-particle emission and isomeric transition) is not studied in this evaluation.

### 3. Nuclear Data

The adopted beta decay energy value  $Q(\beta^-)=272(10)$  keV, is based on the energy measurements of Godart and Gizon (1973Go40): 198.5 (15) keV for the maximum energy of the beta minus particle emissions and 73.92 (2) keV for the isomeric transition; an uncertainty of 10 keV was assigned to the result, according to the above-mentioned considerations. The adopted value of  $Q(\beta^-)$  is in agreement with the value from 2003Audi03: 273.1 (32) keV (based on some older energy measurements of the beta minus particle emissions). The value adopted by this evaluation is also in good agreement with the effective  $Q(\beta^-)$  value of 273 keV (with an uncertainty of 11 keV), calculated from the decay scheme data, by using the SAISINUC software.

### 3.1. Half-life

In the literature, only a few measured <sup>234</sup>Th half-life ( $T_{1/2}$ ) values are reported; these measurements are very old (the most recent is from 1948), so new half-life measurements are needed to improve the quality of the evaluation. The half-life values and their uncertainties are presented in Table 1; the value recommended by Curie et al. (1931), with an estimated uncertainty added by the evaluator, was also included. The set of data is consistent and the recommended value, 24.10 days, with an uncertainty of 0.03 day, is the weighted average (LWM,  $\chi^2_{\nu}=3.78$ ) of the four input values. The references are expressed as NSR (Nuclear Science References) type keynumbers:

**Table 1 : <sup>234</sup>Th Half-life values**

$T_{1/2}$ (days)	Uncertainty of $T_{1/2}$ (days)	Reference
23.8	0.7	1920Ki01
24.5	0.5	1931Cu01
24.1	0.2	1939Sa11
24.101	0.025	1948Kn23

### 3.2. Beta transitions and emissions

In the literature, the most complete reference reporting measurements of energy and emission intensities for <sup>234</sup>Th beta minus transitions is 1973Go40.

For this evaluation, the beta transitions energies were calculated from  $Q(\beta^-)$  and the energies of the decay scheme levels; the high energy uncertainty (10 keV) is explained by the possible low energy and intensity isomeric transition (as described above, in section 2, Decay Scheme). The intensities of the beta branches were deduced from  $\gamma$ -ray transition intensity balance at each level, with the exception of the main branch; its intensity was deduced from the normalization condition of the beta emissions (the sum of the all the beta transitions intensities must be 100 %). The existence of the weakest beta decay branch (95.8 keV) is questionable (2007Br04). The energy and intensity values of the beta transitions, as well as their Log ft values are shown in Table 2.

**Table 2: <sup>234</sup>Th  $\beta^-$  Energies and Emission Probabilities**

$E_{\beta^-}$ (keV)	Uncertainty $E_{\beta^-}$ (keV)	Transition intensity (%)	Transition intensity (%), from 1973Go40	Log ft
85	10	1.6 (6)	1.3 (7)	7.0
95	10	0.016 (5)	-	9.1
105	10	6.5 (7)	5.4 (10)	6.7
106	10	14.1 (12)	20.7 (10)	6.3
198	10	77.8 (15)	72.5 (20)	6.4

### 3.3. $\gamma$ - transitions: $\gamma$ rays and internal conversion electrons

Many measurements of the  $\gamma$ -ray energies and emission intensities following the <sup>234</sup>Th decay were published by different authors: 1973Go40, 1973Sa33, 1973Ta25, 1978Ch06, 1982Mo30, 1990Sc09, 1993Su37, 2004Ab03 and 2006Al28. The interest for high quality data of photon emission probabilities is justified especially in the field of environmental radioactivity monitoring. Table 3 presents measured values of the 63.30 (2) keV  $\gamma$ -ray emission probability following the decay of <sup>234</sup>Th. The set of data is consistent and the recommended value, 3.75 (8) %, is the weighted average (LWM,  $\chi^2_{\nu}=3.32$ ) of the five input values. The references are expressed as NSR type keynumbers.

**Table 3 : Absolute Emission Intensity Results (in %) for the 63.30-keV  $\gamma$  ray.**

Gamma-ray emission probability	Uncertainty of the gamma-ray emission probability	Reference
3.3	0.3	1973Go40
4.05	0.20	1982Mo30
3.6	0.2	1990Sc09
3.99	0.20	1993Su37
3.73	0.07	2004Ab03
<b>3.75</b>	<b>0.08</b>	<b>Adopted</b>

Using this evaluated value and the relative photon intensity values from the measurements of Chu and Scharff-Goldhaber (1978), the corresponding absolute gamma-ray emission probabilities and their uncertainties were computed for all the  $\gamma$  rays and are given below in Table 4. The relative photon intensities measured by Chu and Scharff-Goldhaber were preferred to those of Godart and Gizon (1973), mainly because in this case the U KX-rays contributions were resolved from the gamma-ray peaks situated in the (90-115) keV energy range of the spectra; no other references reporting relative photon intensities measurements were found in the literature.

The intensity balance for level 3 (103.42 keV) was used to compute the emission probability for the 73.85 keV photons, but the obtained value was negative (-0.011 %); as the placement of this transition in the level scheme is uncertain (2007Br04), this low probability photon emission was not considered in this evaluation. Other possible gamma-ray transitions neither confirmed nor placed in the level scheme (proposed / observed only by some authors) are: 57.75 keV, 87.02 keV, 92.00 keV, 103.71 keV, 108.00 keV, 132.9 keV and 184.8 keV.

The internal conversion coefficients were computed with the program BrIcc, version 2.2/2008, using the "Frozen Orbitals" approximation. A difficult case is the computation of the ICC for the 112.81 keV gamma-ray transition, because this energy is too close to the K-shell binding energy for protactinium (112.6 keV) and the software can not be used directly for this purpose. Following Browne and Tuli (2007), a limit on  $\alpha(K)$  ( $\leq 0.29$ ) has been obtained from extrapolation of  $\alpha(K)$ 's for energies higher than 113.6 keV; however, this procedure introduced a large uncertainty of the total ICC value (see Table 4).

**Table 4: <sup>234</sup>Th  $\gamma$ -ray Energies and Absolute Emission Probabilities**

$E_\gamma$ (keV)	Uncertainty $E_\gamma$ (keV)	Absolute Emission Probability (%)	Uncertainty of absolute emission probability (%)	Total ICC ( $\alpha_T$ )
20.01	0.02	0.005 1	0.002 1	240 (70)
29.50	0.02	0.001 23	0.000 14	4390 (70)
62.88	0.02	0.016 4	0.002 8	25 (5)
63.30	0.02	3.75	0.08	0.405 (6)
73.92	0.02	0.013 3	0.001 4	10.6 (4)
83.31	0.05	0.061	0.005	0.196 (3)
92.38	0.01	2.18	0.19	5.27 (8)
92.80	0.02	2.15	0.19	0.1472 (21)
103.35	0.10	0.003 2	0.001 0	3.81 (6)
112.81	0.05	0.215	0.022	0.23 (14)

#### 4. Atomic data

The K-shell fluorescence yield ( $\omega_K$ ), the mean L-shell fluorescence yield ( $\omega_L$ ) and the mean number of vacancies in the L-shell produced by one vacancy in the K-shell ( $\eta_{KL}$ ) were determined using the computer program EMISSION v3.10, 28-Jan-2003: 0.970 (4), 0.488 (18) and 0.795 (5) respectively.

#### 4.1. Auger electrons and X-rays

The relative probability values of the K Auger electron emissions (KLL, KLX, KXY) normalized to the KLL value, were computed using the same EMISSION computer program. The total K Auger electron emission probability (absolute) and the emission probability of the L Auger electrons were also calculated. The energy ranges for K and L Auger electrons were filled-in by the SAISINUC program, version 2008 April.

The relative probability (normalized to  $K_{\alpha 1}$  X-rays emission) and the absolute emission probability values of the different groups of K and L X-rays were determined using the same EMISSION program. The energy range values of the K and L X-rays are from the tables linked to SAISINUC. Neither measurements of X-ray energies nor of emission probabilities were found in the literature, in order to compare them with the results of this evaluation.

#### 5. Main production mode

The main production mode of <sup>234</sup>Th is by alpha-particle decay of the <sup>238</sup>U nuclei (<sup>234</sup>Th is the daughter of <sup>238</sup>U), present in important quantities in many natural ores.

#### 6. References

- 1920Ki01 G. Kirsch, Mitt. Ra. Inst. 127, Wien. Ber. 11a, 129, 309 (1920), M-Sch. P. 377.
- 1931Cu01 M. Curie, A. Debierne, A.S. Eve, H. Geiger, O. Hahn, S.C. Lind, St. Meyer, E. Rutherford and E. Schweidler, "The Radioactive constants as of 1930", Rev. Mod. Phys. 3, 427-445 (1931).
- 1939Sa11 B. W. Sargent, Can. J. Research A17, 103 (1939).
- 1948Kn23 G.B. Knight and R.L. Macklin, "Half-Life of UX<sub>1</sub> (Th<sub>234</sub>)", Phys. Rev. 74, 1540-1541 (1948).
- 1961Ge13 J.S. Geiger, R.L. Graham and T.A. Eastwood, "The Decay of Th<sup>234</sup>", AECL-1472, PR-P-52, 26-27 (1961).
- 1962Br05 J.-P. Briand, "Sur la nature de la transition de 29 keV du Protactinium 234 (UX<sub>2</sub>)", Comp. Rend. Acad. Sci. (Paris) 254, 84-86 (1962).
- 1963Bj02 S. Bjornholm and O.B. Nielsen, "The decay of the 1.14 min Isomer of Pa<sup>234</sup> (UX<sub>2</sub>)", Nucl. Phys. 42, 642-659 (1963).
- 1964Ab04 H. Abou-Leila, "Mesures des périodes des niveaux du protactinium 234 désexcités par les transitions  $\gamma$  de 63 et 93 keV", Comp. Rend. Acad. Sci. (Paris) 258, 5632-5635 (1964).
- 1965Fo12 R. Foucher, "New Data on the Level Diagram of the Odd-Odd Nucleus Pa<sup>234</sup>", Izv. Akad. Nauk SSSR, Ser.Fiz. 29, 100 (1965); Bull. Acad. Sci. USSR, Phys. Ser. 29, 99-100 (1966).
- 1973Sa33 T.E. Sampson, "Precision Measurement of Gamma Ray Energies from <sup>238</sup>U Daughters, II", Nucl. Instrum. Methods 111, 209-211 (1973).
- 1973Ta25 Harry W. Taylor, "Gamma Rays Emitted by Uranium and Thorium in the Energy Range 10-120 keV", Int. J. Appl. Radiat. Isotop. 24, 593-597 (1973).
- 1973Go40 J. Godart and A. Gizon, "Niveaux de <sup>234</sup>Pa Atteints par la Désintégration de <sup>234</sup>Th", Nucl. Phys. A217, 159-176 (1973).
- 1978Ch06 Y.Y. Chu and G. Scharff-Goldhaber, "Decay of <sup>234</sup>Th to the <sup>234</sup>Pa isomers", Phys. Rev. C 17, 1507-1509 (1978).
- 1982Mo30 Michael H. Momeni, "Analyses of Uranium and Actinium Gamma Spectra: An Application to Measurements of Environmental Contamination", Nucl. Instrum. Methods 193, 185-190 (1982).
- 1990Sc09 H.L. Scott and K.W. Marlow, "Gamma-Ray Emission Probabilities of the Daughters of <sup>238</sup>U", Nucl. Instrum. Methods A 286, 549-555 (1990).
- 1990Co08 N. Coursol, F. Lagoutine and B. Duchemin, "Evaluation of Non-Neutron Nuclear Data for the Uranium-238 Decay Chain", Nucl. Instrum. Methods A 286, 589-594 (1990).

- 1993Su37 G.A. Sutton, S.T. Napier, M. John and A. Taylor, "Uranium-238 Decay Chain Data", *The Science of the Total Environment* 130/131, 393-401 (1993).
- 1998Ad08 I. Adsley, J.S. Backhouse, A.L. Nichols and J. Toole. "U-238 Decay Chain: Resolution of Observed Anomalies in the Measured Secular Equilibrium Between Th-234 and Daughter Pa-234m", *Appl. Rad. Isotopes* 49, 1337-1344 (1998).
- 2003Au03 G. Audi, A.H. Wapstra and C. Thibault, "The AME2003 atomic mass Evaluation (II). Tables, graphs, and references", *Nucl. Phys. A* 729, 337-676 (2003).
- 2004Ab03 S. Abousahl, P. van Belle, B. Lynch and H. Ottmar, "New measurement of the emission probability of the 63.290 keV  $^{234}\text{Th}$   $\gamma$ -ray from  $^{238}\text{U}$   $\alpha$  decay", *Nucl. Instrum. Methods A* 517, 211-218 (2004).
- 2006Al28 F.S. Al-Saleh, Al-J.H. Al-Mukren and M.A. Farouk, "Precise determination of the absolute intensities of the gamma-ray lines of  $^{235}\text{U}$  and some  $^{238}\text{U}$  daughters", *Nucl. Instrum. Methods A* 568, 734-738 (2006).
- 2007Br04 E. Browne and J.K. Tuli, "Nuclear Data Sheets for A = 234", *Nucl. Data Sheets* 108, 681-772 (2007).