^{244}Am - Comments on evaluation of decay data by A. L. Nichols

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Evaluation Procedure

Limitation of Relative Statistical Weight Method (LWM) was applied to average the decay data when appropriate (but see below).

Decay Scheme

A relatively simple decay scheme was constructed from the gamma-ray studies of 1962Va08, 1963Ha29, 1967Sc34 and 1984Ho02. Only the gamma-ray measurements of Hoff *et al*. provide any estimates of the uncertainties in the gamma-ray probabilities expressed in terms of their relative intensity per 100 neutron captures in a high-flux reactor (1984Ho02). All other studies contained no information with respect to their overall uncertainties. Thus, no weighted mean data could be derived, and the data of 1984Ho02 were adopted wholesale and re-adjusted when deemed necessary (expressed in terms of the 743.977-keV gamma-ray emission probability (100 %)). Further measurements are merited to quantify the gamma-ray emission probabilities and decay scheme with greater certainty.

Nuclear Data

^{244}Am is an important actinide for high burn-up fuel within the reactor core, and needs to be better characterised for improved assessments of accelerator-driven systems (ADS) and ^{244}Cm decay heat contribution.

Half-life

The recommended half-life has been adopted from the single known measurement of Vandenbosch and Day (1962Va08). Further measurements are required to determine this half-life with much greater confidence.

Half-life measurement

Reference	Half-life (hours)
1962Va08	10.1 ± 0.1

Gamma Rays

<u>Energies</u>

All gamma-ray transition energies were calculated from the structural details of the proposed decay scheme. The nuclear level energies of Akovali were adopted (2003Ak04), and used to determine the energies and associated uncertainties of the gamma-ray transitions between the various populated-depopulated levels. However, Akovali recommended the gamma-ray energies determined by Hoff *et al.* (1984Ho02) by means of two curved-crystal spectrometers – minor differences do occur between the calculated energies of the higher energy transitions (538.402 (16), 743.977 (5) and 897.840 (7) keV) and those observed by Hoff *et al.*

Emission Probabilities

Relative emission probabilities and their uncertainties were determined from measurements of Hoff *et al*. (1984Ho02). These data were estimated to be in reasonably good agreement with the earlier measurements of Vandenbosch and Day, and Schuman (1962Va08, 1967Sc34), although these latter two sets of data possessed no uncertainties. Under these unsatisfactory circumstances, the data of Hoff *et al.* had to be adopted wholesale as the only suitable starting point in the attempted construction of a consistent decay scheme. Adjustments were made to the relative emission probabilities of the 99.383-, 153.863- and 205.575-keV gamma-rays (adjusted from 7.0 (12) to 7.5 (13), 25 (5) to 28.6 (60), and 0.52 (12) to 0.53 (12), respectively) to conform with respect to the expected population-depopulation balance for the 501.79-, 296.21- and 142.35-keV nuclear levels of ^{244}Cm. Furthermore, a relative emission probability had to be calculated for the 42.96-keV gamma-ray for which there were no data at all (from a population-depopulation balance of the 42.96-keV nuclear level of ^{244}Cm (populated by the 99.38-keV gamma ray and depopulated by the 42.96-keV gamma ray)). Downward adjustments were made to the uncertainties of specific gamma-ray transitions and emissions through consideration of these and other data that are judged to be heavily correlated (99.383- and 153.863-keV gamma rays compared with 743.977-keV gamma-ray and each other).

Measured relative gamma-ray emission probabilities

	E_γ (keV)	P_γ^{rel}		
		1962Va08	**1967Sc34**	**1984Ho02**
$\gamma_{1,0}$ (Cm)	42.965 (10)	-	-	-
$\gamma_{2,1}$ (Cm)	99.383 (4)	-	-	0.23 (4) → 7.0 (12)
$\gamma_{3,2}$ (Cm)	153.863 (2)	72 → 100	-	0.82 (16) → 25 (5)
$\gamma_{4,3}$ (Cm)	205.575 (4)	0.4 → 0.6	-	0.017 (4) → 0.52 (12)
$\gamma_{9,4}$ (Cm)	538.402 (16)	0.4 → 0.6	-	0.033 (7) → 1.0 (2)
$\gamma_{9,3}$ (Cm)	743.977 (5)	72 → 100	66.2 → 100	3.3 (9) → 100 (27)
$\gamma_{9,2}$ (Cm)	897.840 (7)	28 → 39	27.6 → 42	1.4 (4) → 42 (12)

Gamma-ray emissions: recommended energies, relative emission probabilities, multipolarities and theoretical internal conversion coefficients (frozen orbital approximation)

E_γ (keV)	P_γ^{rel}	Multipolarity	α_K	α_L	α_{M+}	α_{tot}	
42.965 (10)	0.145 (12)*	E2	-	760 (11)	290 (4)	1050 (15)	β^-
99.383 (4)	7.5 (13)§	E2	-	13.9 (2)	5.4 (1)	19.3 (3)	β^-
153.863 (2)	28.6 (60)§	E2	0.174 (3)	1.90 (3)	0.74 (1)	2.81 (4)	β^-
205.575 (4)	0.53 (12)§	E2	0.141 (2)	0.541 (8)	0.205 (3)	0.887 (13)	β^-
538.402 (16)	1.0 (2)	E2	0.0292 (4)	0.0149 (2)	0.0054 (1)	0.0495 (7)	β^-
743.977 (5)	100 (27)	M1 + E2 $\delta = -0.92$ (8)	0.059 (4)	0.0130 (7)	0.0050 (3)	0.077 (5)	β^-
897.840 (7)	42 (12)	E2	0.0122 (2)	0.00358 (5)	0.00124 (2)	0.0170 (3)	β^-

* Determined from the calculated theoretical internal conversion coefficients and the transition probability of the 99.383-keV gamma ray feeding the 42.965-keV nuclear level of ^{244}Cm.

§ Adjusted to conform with respect to the expected population-depopulation balances for the 501.79-, 296.21- and 142.35-keV nuclear levels of ^{244}Cm.

A normalisation factor of 0.66 (14) was calculated from the relative emission probabilities of the three gamma rays that depopulate the 1040.188-keV nuclear level:

$$\sum^{3} P_{\gamma}\,(1+\alpha_{tot})\times F \quad = \quad 100\%$$

$$[P^{rel}(897.84\,keV)(1+\alpha_{tot}) + P^{rel}(743.97keV)(1+\alpha_{tot}) + P^{rel}(538.40keV)(1+\alpha_{tot})]\times F \; = 100$$

$$F = 100/151\ (32) = 0.66 \pm 0.14$$

<u>Multipolarities and Internal Conversion Coefficients</u>
The nuclear level scheme specified by Akovali has been used to define the multipolarities of the gamma transitions on the basis of known spins and parities (2003Ak04). Hansen *et al.* undertook angular correlation measurements to confirm the assignment of the 1040.2-keV nuclear level as the only ^{244}Cm nuclear level populated directly by β^- decay (1963Ha29), in which the depopulating 743.977-keV gamma ray was defined as (46 ± 4) % quadrupole [E2] and (54 ± 4) % dipole [M1] to give a mixing ratio (δ) of – 0.92 (8) for this transition. Recommended internal conversion coefficients have been determined from the frozen orbital approximation of Kibédi *et al*. (2008Ki07), based on the theoretical model of Band *et al*. (2002Ba85, 2002Ra45).

Beta Particle
<u>Energy and emission probability</u>
The single beta-particle energy was calculated from the structural detail of the proposed decay scheme. A nuclear level energy of 1040.188 (12) keV from Akovali (2003Ak04) and a Q_{β^-} value of (1427.3 ± 1.0) keV from Audi *et al.* (2003Au03) were used to determine the energy and uncertainty of the beta-particle transition. By definition, this single beta transition was assigned an emission probability of 100 %.

Beta-particle Emission Probability per 100 Disintegrations of ^{244}Am.

	E_β (keV)	P_β	Transition type	log *ft*
$\beta^-_{0,9}$	387.1 ± 1.0	100	(1st forbidden non-unique)	5.63

Atomic Data

The x-ray and Auger-electron data have been calculated using the evaluated gamma-ray data, and atomic data from 1996Sc06, 1998ScZM and 1999ScZX. Both the x-ray and Auger-electron emission probabilities were determined by means of the EMISSION computer program (version 4.01, 28 January 2003, with the emission.101 database extended to Z = 96 to calculate component L x-ray data of daughter Cm). This program incorporates atomic data from 1996Sc06 and the evaluated gamma-ray data.

K and L X-ray Emission Probabilities per 100 Disintegrations of ^{244}Am.

			Energy keV	Photons per 100 disint.
XL		(Cm)	12.633 – 23.527	100 (10)
	XL_l	(Cm)	12.633	2.36 (24)
	XL_α	(Cm)	14.746 – 14.961	36 (4)
	XL_η	(Cm)	17.314	1.15 (15)
	XL_β	(Cm)	17.286 – 19.688	51 (5)
	XL_γ	(Cm)	22.735 – 23.527	12.5 (13)
XK_α	$XK_{\alpha 2}$	(Cm)	104.590	2.2 (3)
	$XK_{\alpha 1}$	(Cm)	109.271	3.4 (4)
$XK'_{\beta 1}$	$XK_{\beta 3}$	(Cm)	122.304	)
	$XK_{\beta 1}$	(Cm)	123.403	) 1.29 (16)
	$XK''_{\beta 5}$	(Cm)	124.124	)
$XK'_{\beta 2}$	$XK_{\beta 2}$	(Cm)	126.889	)
	$XK_{\beta 4}$	(Cm)	127.352	) 0.45 (6)
	$XKO_{2,3}$	(Cm)	127.970	)

Electron energies were determined from electron binding energies tabulated by Larkins (1977La19) and the evaluated gamma-ray energies. Absolute electron emission probabilities were calculated from the evaluated absolute gamma-ray emission probabilities and associated internal conversion coefficients.

Data Consistency

A Q_β-value of 1427.3 (1) keV has been adopted from the atomic mass evaluation of Audi *et al.* (2003Au03) while in the course of formulating the decay scheme of ^{244}Am. This value has subsequently been compared with the Q-value calculated by summing the contributions of the individual emissions to the ^{244}Am beta-decay process (i.e. β^-, conversion electrons, γ, etc.):

calculated Q-value = $\sum (E_i \times P_i)$ = 1431 (90) keV

Percentage deviation from the Q-value of Audi *et al.* is (0 ± 6) %, which supports the derivation of a highly consistent decay scheme with a large variant.

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