^{236}Np – Comments on evaluation of decay data by V.P. Chechev and N.K. Kuzmenko

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1. DECAY SCHEME

From the systematics of the isomer levels it has been assumed in 1981Li30 (see also the analysis carried out in 1991Sc08) that the short-lived state of ^{236}Np (22,5 h) lies higher in energy than the long-lived state of ^{236}Np (1,55 × 10^5 a). In line with this assumption we consider the long-lived state of ^{236}Np as the ground state. Using Q values for electron capture decay of the isomer and ground state and a close energy cycle we can estimate the energy level spacing between these states as 60 (50) keV.

The decay scheme of the long-lived ^{236}Np includes three decay modes: β^- decay to ^{236}Pu, electron capture decay (EC) to ^{236}U and α decay to ^{232}Pa (see 2006Br20). A favored α-particle branch to the (6^-) level at ≈ 400 keV is expected in ^{232}Pa from α systematics (1972El21, 1980Sc26, 2006Br20). However, this decay was not observed experimentally.

The β^- -decay branching, $\Sigma P(\beta^-)$, and alpha-decay branching, $\Sigma P(\alpha)$, have been deduced by the evaluators from the partial half-lives $T_{1/2}(\beta^-)$ and $T_{1/2}(\alpha)$, respectively, measured in 1981Li30. The EC-decay branching, $\Sigma P(EC)$, has been obtained as the difference of $1 - \Sigma P(\beta^-) - \Sigma P(\alpha)$.

2. NUCLEAR DATA

Q^-, Q_{EC}, $Q(\alpha)$ values are from 2003Au03.

The total half-life of ^{236}Np is based on the evaluated partial half-lives $T_{1/2}(\alpha)$, $T_{1/2}(\beta^-)$, $T_{1/2}(EC)$ measured in 1981Li30.

The evaluated $T_{1/2}(\alpha)$ = 9,5 (35) × 10^7 years has been obtained as an average of the two measurements of 1981Li30 (specific activity, ^{232}U gamma-ray of 894 keV was measured): 9,4 (35) × 10^7 and 9,6 (35) × 10^7 years. A standard deviation of the individual measurement has been adopted for the uncertainty of the evaluated alpha-decay half-life using a rule that the uncertainty assigned to the recommended value should be greater than or equal to the smallest uncertainty in any experimental value.

$T_{1/2}(\beta^-)$ = 1,29 (3) × 10^6 years has been adopted here from the ^{236}Pu growth measurement of 1981Li30. The result of this measurement is independent of the decay scheme, and it is equal to the weighted average of 1,34 (15), 1,29 (3), 1,32 (9), 1,69 (30), 1,29 (3), 1,31 (8) (in 10^6 years) given in 1981Li30. The uncertainties of these measurements do not include any estimation of uncertainties from the decay scheme parameters. It agrees well with an earlier measurement in 1972En06 (1,29 (+ 0,07, - 0,05) × 10^5 a).

The evaluated $T_{1/2}(EC)$ = 1,77 (10) × 10^5 years has been obtained as an average of the two ^{236}U/^{235}U mass ratio measurements in 1981Li30: 1,75 (10) × 10^5 and 1,79 (10) × 10^5 years. These ^{236}U growth measurement results are independent of the decay scheme. A standard deviation of the individual measurement has been adopted for the uncertainty of the evaluated partial EC-decay half-life. The specific gamma-ray activity method (^{236}U 160,3-keV gamma-ray was measured) was used in other measurements presented in 1981Li30 (in 10^5 years): 1,60 (4), 1,73 (2), 1,77 (11), 1,75 (10), 1,79 (10), 1,74 (1), 1,78 (10). The uncertainties of these measurements do not include an estimation of uncertainties from the decay scheme parameters.

Thus, the recommended value of the total ^{236}Np half-life obtained from the relation $T_{1/2} = [(T_{1/2}(\alpha))^{-1} + (T_{1/2}(\beta^-))^{-1} + (T_{1/2}(EC))^{-1}]^{-1}$ is 1,55 (8) × 10^5 years.

2.1.1. Electron Capture Transitions

The energies of the electron capture transitions have been deduced from the Q_{EC} value and the level energies given in Table 1 from 2006Br20 where they were deduced from a least squares fit to gamma-ray energies.

Table 1. ^{236}U levels populated in ^{236}Np electron capture decay

Level number	Energy (keV)	Spin and parity	Half-life	Probability of ε - transition (x 100)
0	0,0	0^+	$2{,}342 \cdot 10^7$ a	-
1	45,2440 (20)	2^+	234 (6) ps	-
2	149,477 (6)	4^+	124 (7) ps	0,0 (44)
3	309,785 (7)	6^+	58 (3) ps	87,8 (43)
4	687,59 (4)	1^-	3,78 (9) ns	-
5	744,18 (7)	3^-	< 0,1 ns	-
6	848,1 (8)	5^-		~ 0,09

The probabilities of the electron capture transitions $P(EC_{0,2})$ and $P(EC_{0,3})$ have been deduced from the correlations of:
$P(EC_{0,2}) + P(EC_{0,3}) = 100\ \% - \sum P(\beta^-) - \sum P(\alpha) = 87{,}8\ (6)\ \%$ and $P(EC_{0,3}) = P(\gamma_{3,2} + ce)$(160-keV).
The upper limit of $P(EC_{0,2}) < 4{,}4$ % has been obtained from the level intensity balance: $P(EC_{0,2}) = 0{,}0\ (44)$ %. The estimate of $P(EC_{0,6}) \sim 0{,}1$ % is given in 1996FiZX.

2.1.2. Beta Transitions

The energies of the β^-- transitions have been deduced from the Q^- value and the level energies given in Table 2 from 2006Br20 where they were deduced from a least squares fit to gamma-ray energies.

Table 2. ^{236}Pu levels populated in ^{236}Np β^--decay

Level number	Energy (keV)	Spin and parity	Half-life	Probability of β^-- transition (x100)
0	0,0	0^+	2,858 a	-
1	44,63 (10)	2^+		-
2	147,45 (10)	4^+		0,2 (14)
3	305,80 (11)	6^+		11,8 (12)

The β^- transition probability $P(\beta_{0,3}) = P(\gamma_{3,2}+ce)$(158-keV) and $P(\beta_{0,2}) = 12{,}0\ (6)\ \% - P(\beta_{0,3}) = 0{,}2\ (14)$ %. An upper limit of $P(\beta_{0,2}) < 1{,}6$ % follows this result.

2.2. Gamma Transitions and Internal Conversion Coefficients

The evaluated energies of gamma-ray transitions are virtually the same as the photon energies because nuclear recoil is negligible.

The gamma-ray transition probabilities have been obtained from the gamma-ray emission probabilities and the total internal conversion coefficients (ICCs).

Multipolarities of gamma-ray transitions have been taken from 2006Br20. The ICCs have been interpolated using the BrIcc package with the so called "*Frozen Orbital*" approximation (2008Ki07) except for $\gamma_{4,1}$ (642,3-keV) and $\gamma_{4,0}$ (687,6-keV). The relative uncertainties of the ICCs for pure multipolarities have been taken as 2 %.

For $\gamma_{4,1}$(642,3-keV) and $\gamma_{4,0}$(687,6-keV) the ICC values of α_K and α_L are experimental results from ^{240}Pu α-decay study (1969Le05, 1977Po05). The ICC values of α_M and α_T for these transitions have been deduced using α_M/α_L and α_{NO}/α_M from 1971Dr11. More accurate ICC measurements for these E1 anomalously converted gamma-ray-transitions are required.

3. ATOMIC DATA

The atomic data are from Schönfeld and Janβen (1996Sc06).

4. ELECTRON EMISSIONS

The energies of the conversion electrons have been obtained from the gamma transition energies and the atomic electron binding energies.

The emission probabilities of the conversion electrons have been obtained using evaluated P(γ) and ICC values.

The absolute emission probabilities of K and L Auger electrons have been obtained using the EMISSION computer program.

β^- average energies have been obtained using the LOGFT computer program.

5. PHOTON EMISSIONS

5.1. X-Ray Emissions

The absolute emission probabilities of U and Pu KX- and LX-rays have been deduced using the EMISSION computer program.
For U LX-ray intensity calculations the theoretical fractional ratios $P_{EC}(L2)/P_{EC}(L1) = 0,115$ and $P_{EC}(L3)/P_{EC}(L1) = 0$ for the EC-transition to the level ''3'' (309 keV) of ^{236}U have been used (1972Dzhelepov). The calculated relative intensities of U KX- rays accompanying the electron capture of ^{236}Np are in a good agreement with the experimental results (Table 3).

Table 3. Intensities of U KX- rays (relatively to P($\gamma_{3,2}$ -160,3 keV)) accompanying ^{236}Np electron capture

	1983Ah03 (experimental)	Adopted (deduced)
X_K		
$K\alpha_2$	0,61 (2)	0,64 (3)
$K\alpha_1$	0,99 (3)	1,02 (5)
$K\beta_1$'	0,38 (2)	0,368 (19)
$K\beta_2$'	0,131 (7)	0,126 (7)

5.2. Gamma Ray Emissions

5.2.1. Gamma Ray Energies (^{236}U)

The energies of gamma-rays in ^{236}Np electron capture have been taken from 2006Br20.

5.2.2. Gamma Ray Energies (^{236}Pu)

The energies of gamma-rays $\gamma_{1,0}$ (44,6 keV), $\gamma_{2,1}$ (102,8 keV), $\gamma_{3,2}$ (158,3 keV) accompanying β^- decay of ^{236}Np have been adopted from measurements given in 1983Ah02 (see also 2006Br20).

5.2.3. Gamma-Ray Emission Probabilities (^{236}U)

The evaluated gamma-ray emission probabilities P(γ) have been deduced using the relative gamma-ray intensities from 1983Ah02 (Table 4), the relation of $\sum P(EC_{0,i})$ = 87,8 (6) % = P($\gamma_{2,1}$ +ce)(104,23 keV) and the intensity balance at ^{236}U each level. We have assumed that the populations to the two lower levels ("0" and "1") in the ^{236}Np electron capture decay are negligible and have taken into account the intensity balance correlation for the gamma-ray transitions to these levels, that is P($\gamma_{1,0}$ +ce)(45,2 keV) = P($\gamma_{2,1}$ +ce) (104,2 keV).

The recommended gamma-ray emission probabilities for γ-rays de-exciting level ''4'' ($\gamma_{4,2}$(538,1 keV), $\gamma_{4,1}$(642,3 keV), and $\gamma_{4,0}$(687,6 keV)) have been deduced from the correlation:
P($\gamma_{5,4}$ +ce)(56,6 keV) = P($\gamma_{4,2}$ +ce)(538,1 keV) + P($\gamma_{4,1}$ +ce)(642,3 keV) + P($\gamma_{4,0}$ +ce)(687,6 keV) using the relative intensities for these γ-rays evaluated from the ^{240}Pu α-decay (Table 5) and assuming $P(EC_{0,4})$ = 0.

Table 4. Gamma rays in decay of the long-lived ^{236}Np measured in 1983Ah02

	Energy (keV)	Relative intensity
$\gamma_{1,0}$ (^{236}U)	45,23 (3)	0,4 (1)
$\gamma_{2,1}$ (^{236}Pu)	102,82 (2)	2,9 (2)
$\gamma_{2,1}$ (^{236}U)	104,23 (2)	23 (1)
$\gamma_{3,2}$ (^{236}Pu)	158,35 (2)	13,5 (7)
$\gamma_{3,2}$ (^{236}U)	160,33 (2)	100

Table 5. Experimental and evaluated absolute emission probabilities of gamma-rays de-exciting the ^{236}U level with energy of 687,6 keV in decay of ^{240}Pu (per 10^8 α-decays) and the deduced relative intensities of these gamma-rays

	Energy (keV)	1969Le05	1971GuZY	1975OtZX	1975Dr05	1976GuZN	Evaluated	Evaluated relative intensities
$\gamma_{4,2}$	538,1	≈ 0,23 [a]		0,147 (12)			0,147 (12)	1,17 (10)
$\gamma_{4,1}$	642,3	14,5 [a]	14,5 (5) [b]	12,6 (4)	13 (1)	12,45 (30)	12,6 (3) [c]	100 (3)
$\gamma_{4,0}$	687,6	3,77 (11)	3,70 (15) [b]	3,30 (13)		3,55 (9)	3,56 (15) [d]	28,3 (13)

[a] Omitted from averaging as uncertainty is not quoted
[b] Omitted from averaging as the data of 1971GuZY have been revised in 1976GuZN
[c] Weighted average of 3 experimental values; the uncertainty is the smallest quoted uncertainty
[d] Weighted average of 3 experimental values; the uncertainty is external

5.2.4. Gamma-Ray Emission Probabilities (^{236}Pu)

The recommended gamma-ray emission probabilities P(γ) have been deduced using the relative gamma-ray intensities from 1983Ah02 (Table 4), the quantity $\sum P(\beta^-)$ = 12,05 (60) % = P($\gamma_{2,1}$+ce)(102,8 keV) and the intensity balance at each ^{236}Pu level. We have assumed that the populations to the two lower levels ("0" and "1") in the ^{236}Np beta minus decay are negligible and have taken into account the intensity balance of the gamma-ray transitions to these levels, that is P($\gamma_{1,0}$+ce)(44,6 keV) = P($\gamma_{2,1}$+ce)(102,8 keV).

6. CONSISTENCY OF RECOMMENDED DATA

The most accurate Q value, Q(M), is taken from the atomic mass adjustment table of Audi et al. (2003Au03). Comparison of Q(eff) (deduced as the sum of average energies per disintegration ($\sum E_i \times P_i$) for all emissions accompanying ^{236}Np β$^-$-decay or ^{236}Np electron capture) with the tabulated decay energy $Q^-(M) \times P(\beta^-)$ for β$^-$-decay or $Q_{EC}(M) \times P(EC)$ for electron capture allows to check a consistency of the recommended ^{236}Np decay-scheme parameters obtained in this evaluation.

Here E_i and P_i are the evaluated energies and emission probabilities of the i-th beta particle, gamma ray, X-ray, etc. The values of P(β$^-$), P(EC) are β$^-$ -decay and EC branching, respectively. Consistency (percentage deviation) is determined by {[Q(M) – Q(eff))]/ Q(M)} × 100. "Percentage deviations above 5 % would be regarded as high and imply a poorly defined decay scheme; a value of less than 5 % indicates the construction of a reasonably consistent decay scheme" (quoted from the article by A. L. Nichols in Appl. Rad. Isotopes 55(2001)23-70).

For the above ^{236}Np decay data evaluation we have for β$^-$-decay $Q^-(M) \times P(\beta^-)$ = 58 (7) keV and Q(eff, β$^-$) = 57 (7) keV, i.e. consistency is less than 2 % if we do not take into account the uncertainties, and the exact percentage deviation is (1.7 ± 17) % if we consider the uncertainties. Similarly, for ^{236}Np electron capture we have $Q_{EC}(M) \times P(EC)$ = 817 (44) keV and Q(eff, EC) = 817 (50) keV and the percentage deviation is 0 ± 8 %. These values indicate the right evaluation and inaccurate measurements of ^{236}Np decay-scheme parameters.

7. REFERENCES

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