^{99}Tc - Comments on evaluation of decay data by X. Mougeot

This evaluation was done in 2010, taking into account the available literature by March 2010.

1 Decay Scheme

The decay scheme is complete since all of the levels in ^{99}Ru below the decay energies are populated. The J^{π} of the ground and excited levels are from the evaluation of Muller et al. (1986Mu09).

2 Nuclear Data

The Q value is from 2003AU03: $Q(\beta^-)$ = 293,8 (14) keV. Measurements of the end-point of the main β transition are given in Table 1.

Table 1: Measured end-points of the main β transition.

Reference	E_{max} (keV)	Uncertainty (keV)
1947MO15	320	-
1950KE02	300	10
1951TA05	290	4
1952FE16	290	4
1960BO08	290	10
1966SN02	294	4
1974RE11	293	2

The evaluation of the ^{99}Tc half-life is described in the next section. Table 2 summarizes the measurements and their methodology.

Although the β-decay of ^{99}Tc is practically 100 % from its $9/2^+$ ground state to the $5/2^+$ ground state of ^{99}Ru, 1973LE10 and 1974EN02 observed a β-decay with a very small intensity to the $3/2^+$ first excited state of ^{99}Ru. Thus, its energy, half-life and the multipolarity of the de-exciting γ-ray are evaluated. The β^- branching ratio is evaluated next.

An analysis of the published form factors of the main β^- transition is presented here. With the described limitations, an evaluation was carried out and the mean energy of the spectrum was calculated.

2.1 ^{99}Tc half-life

The measured half-life values of ^{99}Tc are given in Table 2 together with the experimental methods that were used. The value from 1947MO15 was not used in the evaluation because an experimental uncertainty was not reported. The value from 1960BO08 is a more recent one from the same authors.

Table 2: ^{99}Tc half-life measurements.

Reference	$T_{1/2}$ (x10^5 a)	Uncertainty (x10^5 a)	Method	Observations
1947MO15	9,4	-	Aluminium absorption	Not used: no uncertainty
1951FR05	2,12	0,04	Aluminium absorption	
1960BO08	2,15	0,05	Aluminium absorption	Same authors as 1947MO15
1966GO10	2,14	0,05	Liquid Scintillation Counting	
1984CO30	2,111	0,012	Liquid Scintillation Counting	

The statistical processing was done using the LWEIGHT program. A weighted average was adopted here from the resulting consistent data set, with a reduced-χ^2 value of 0,29. The statistical weight is 82 % for 1984CO30, the most recent and precise measurement. Finally, the adopted value, with its internal uncertainty, is:

$$T_{1/2} = 2{,}115\ (11) \times 10^5 \text{ a}.$$

2.2 γ transition : first excited state of ^{99}Ru

2.2.1 Energy

The measured energies of the first excited state of ^{99}Ru are given in Table 3 with the experimental methods used.

Table 3: Measurements of the energy of the first excited state of ^{99}Ru.

Reference	Energy (keV)	Uncertainty (keV)	Method
1967MO20	89,36	0,40	^{99}Rh decay, γ Ge(Li)
1970AN12	89,6	0,5	^{99}Rh, γ Ge(Li)
1971LE20	89,4	1,0	^{98}Mo(α,3n)^{99}Ru, γ Ge(Li)
1973LE10	89,7	0,4	^{99}Tc decay, β Si(Li)
1974EN02	89,5	0,2	^{99}Tc decay, γ Si(Li)

The statistical processing was done using the LWEIGHT program. A weighted average was adopted from the resulting consistent data set, with a reduced-χ^2 value of 0,10. The statistical weight is 59 % for 1974EN02, the most precise measurement. Finally, this evaluation gives:

$$E_\gamma(^{99}\text{Ru}) = 89{,}52\ (15)\ \text{keV}.$$

2.2.2 $T_{1/2}$(^{99}Ru, 89 keV)

The measured half-life values of the first excited state of ^{99}Ru are given in Table 4 together with the experimental methods used. The original uncertainty of 1972GU01, not explained in detail in the article, seems to be underestimated. 1973BE72 used the same method, with nearly the same statistics, and reported an uncertainty of 0,6. The uncertainty of 0,1 given by 1972GU01 seems to be only that from the data fitting. Thus, the evaluator decided to increase the uncertainty of 1972GU01 from 0,1 to 0,6.

Table 4: Measurements of the half-life of the first excited state of ^{99}Ru.

Reference	$T_{1/2}$ (ns)	Uncertainty (ns)	Method	Observation
1964BO28	19,7	0,4	^{99}Rh, γ spectro.	
1965KI01	20,0	1,0	^{99}Rh, γ spectro.	
1965MA27	20,7	0,3	^{99}Rh, γ spectro.	
1972GU01	20,5	0,6	^{99}Rh, γ Ge(Li)	Uncertainty increased from 0,1 to 0,6
1973BE72	21,04	0,6	^{99}Rh, γ Ge(Li)	
1974EN02	18,9	1,0	^{99}Tc decay, γ Si(Li)	

The statistical processing was done using the LWEIGHT program. A weighted average was adopted here from the resulting consistent data set, with a reduced-χ^2 value of 1,52. The statistical weight is 45 % for 1965MA27. These authors gave some details on their estimation of the uncertainty, and there is no reason to believe it was underestimated. Finally, this evaluation gives:

$$T_{1/2}(^{99}\text{Ru, 89 keV}) = 20{,}36\ (25)\ \text{ns}.$$

2.2.3 Multipolarity

The γ transition from the $3/2^+$ first excited state to the $5/2^+$ ground state of ^{99}Ru is M1+E2. Measurements were carried out to obtain the mixing ratio δ^2 = E2/M1. They are summarized in Table 5 with the experimental methods used. Only two measurements were used for the evaluation because most of the publications are from the same author. Only the most recent one, which is also the most precise, was included. The value from 1973BE72 was not used because the experimental uncertainty was not reported.

The statistical processing was done using the LWEIGHT program. A weighted average was adopted here from the resulting consistent data set, with a reduced-χ^2 value of 2,69. The statistical weight is 94 % for 1976KI02, the most precise measurement. Finally, this evaluation gives:

$$\delta^2(^{99}\text{Ru, 89 keV}) = 2{,}45\ (6).$$

Then $\delta = -1{,}56\ (2)$, and the multipolarity is:

$$\text{M1} + 71{,}0\ (5)\ \%\ \text{E2}.$$

Table 5: Measurements of the multipolarity mixing ratio of the first excited state of ^{99}Ru. The values from 1973Gibb and 1976KI02 were the only ones that were used for the evaluation.

Reference	δ^2 = E2/M1	Uncertainty	Method	Observation
1964KI01	~ 2	-	Ru-99 Mössbauer γ transition	Not used: no uncertainty
1965KI01	2,4	0,9	Ru-99 Mössbauer γ transition	Same author as 1964KI01
1966KI02	2,7	0,6	Ru-99 Mössbauer γ transition	δ<0, same author as 1964KI01
1972Wagner	2,7	0,6	Ru-99 Mössbauer γ transition	Coming from 1966KI02
1973BE72	2,57	-	^{99}Rh, γ Ge(Li)	Not used: no uncertainty
1973Gibb	2,72	0,17	Ru-99 Mössbauer γ transition	
1976KI02	2,43	0,04	Ru-99 Mössbauer γ transition	δ = -1,56 (2), same author as 1964KI01

2.2.4 Branching ratio

1973LE10 and 1974EN02 inferred a small β^- transition from the $9/2^+$ ground state of ^{99}Tc to the $3/2^+$ 89 keV level of ^{99}Ru, by detecting a de-exciting γ-ray. Thus, this β transition is second unique forbidden, whereas the main transition is second non-unique forbidden.

The authors reported the number of photons detected per decay: 6,5 (1,5) × 10^{-6} for 1973LE10 and 4,9 (1,7) × 10^{-6} for 1974EN02. Next, they used a total internal conversion coefficient α_T = 1,5 calculated from 1968HA52 to determine the corresponding total γ-ray transition probability, and thus the β^- branching.

The absolute γ-ray intensity was evaluated. The statistical processing was done using the LWEIGHT program. A weighted average was adopted from the resulting consistent data set, with a reduced-χ^2 value of 0,50. The statistical weight is 56 % for 1973LE10. Finally, this evaluation gives:

$$I_{abs}(^{99}\text{Ru, 89 keV}) = 5{,}8\ (11) \times 10^{-4}\ \%.$$

The total conversion coefficient α_T was calculated using the BrIcc program (2008KI07): α_T = 1,495 (25). Thus the β^- branching is equal to $I_{abs}(1+\alpha_T)$. Finally, this evaluation gives:

$$P_{\beta 0,1} = 1{,}45\ (30) \times 10^{-3}\ \%.$$

2.3 β^- transition

The branching of the main β^- transition is practically 100 %. A small contribution to the first excited state of ^{99}Ru exists, with a branching of 1,45 (30) × 10^{-3} %, as deduced in Section 2.2.4.

The main β^- transition is from the $9/2^+$ ground state of ^{99}Tc to the $5/2^+$ ground state of ^{99}Ru. This is a second forbidden non-unique transition, thus one can expect a form factor as given below (1976Behrens):

$$C(W) = A(W)q^2 + B(W)\lambda_2 p^2 + D(q^4 + 10/3\lambda_2 q^2 p^2 + \lambda_3 p^4),$$

where q is the linear momentum of the neutrino, p the linear momentum of the electron, and W is the normalised energy of the electron. Measurements show that the following form factor for a first unique forbidden transition gives a good description of the measured energy spectrum:

$$C(W) = q^2 + \lambda p^2.$$

The determination of the form factor is highly dependent on the calculated spectrum used for the comparison with experimental data. Consequently, the form factor depends on the hypothesis made and the data used for the calculation: Coulomb corrections, screening correction due to electron cloud, finite nuclear size correction, radiative corrections, end-point energy, and nature of the transition. The form factor is generally determined by a comparison with a calculated allowed spectrum.

Table 6: Measurements of the form factor of the main β transition.

Reference	λ	Uncertainty	E_{max} (keV)	Energy range (keV)	Method	Observation
1951TA05	~ 1	-	290 (4)	150 - end-point	Mag. spectro.	Not used: no uncertainty
1952FE16	0,50	0,13	292 (3)	60 - end-point	Mag. spectro	Recalculated by 1966Lipnik
1966SN02	0,49	0,04	294 (4)	50 - 280	Plastic scint.	Recalculated by 1976Behrens
1974RE11	0,54	0,02	293 (2)	55 - 250	Si(Li)	

Experimental data are summarized in Table 6 with the energy range of their validity, and the experimental methods used. The value from 1951TA05 was not used because no experimental uncertainty was reported. The value from 1952FE16 was calculated by 1966Lipnik in the correct form. The value from 1966SN02 was recalculated by 1976Behrens using more recent tables for the Fermi function, leading to an increase of the uncertainty from 0,011 to 0,04. The statistical processing was done using the LWEIGHT program. A weighted average was adopted from the resulting consistent data set, with a reduced-χ^2 value of 0,65. The statistical weight is 78 % for 1974RE11, the most recent and precise measurement. Finally, this evaluation gives:

$$C(W) = q^2 + 0{,}529\ (18)\ p^2.$$

It should be underline the main difficulty of the evaluation of form factors: the authors of the published data did not describe in detail all the possible sources of distortion of the measured spectra and their contributions. Obviously, this is a difficult task. In some articles, it is clear that all these problems were not taken into account. The temptation could be great to adjust some known parameters within the uncertainty range to obtain a result close to the previous published ones.

Resulting from the violation parity, electrons emitted in nuclear β-decay are longitudinally spin-polarized. If the decaying neutron is influenced by the nuclear structure in which it is embedded, the value of the polarization may be altered. The authors of 1990GA13 measured the longitudinal electron polarization, and they suggested that the decaying neutron is not influenced more than 3,3 % by the nuclear structure. This could explain why a form factor of a first unique forbidden transition is sufficient to describe a second non-unique forbidden transition. One can note the usual approximation in the theoretical calculation of β spectra: a non-unique forbidden transition is treated as a unique forbidden transition with the same variation of the total angular momentum. It means that a second non-unique forbidden transition is treated as a first unique forbidden transition.

The mean energy of the β spectrum was calculated with the Q value and the form factor given previously. The calculation is based on the analytical approach developed by N.B. Gove and M.J. Martin (1971GO40) and it includes the following correction terms: Coulomb corrections (1961RO33), screening correction due to electron cloud (1954GO69), finite nuclear size correction (1980Dillman), and radiative corrections (1982Behrens). The uncertainty is estimated by the product of E_{mean} with the uncertainty on λ. The result is:

$$E_{mean} = 94{,}6\ (17)\ \text{keV}.$$

The log *ft* value for the main transition (second non-unique forbidden) has been calculated with the LOGFT program: log *ft* (^{99}Tc→^{99}Ru^{gs}) = 12,323 (7). In the same way, for the second unique forbidden transition to the first excited state of ^{99}Ru: log *ft* (^{99}Tc→^{99}Ru*) = 15,82 (9).

For the sake of completeness, we mention some publications on K-shell auto-ionization probabilities accompanying the β decay of ^{99}Tc: 1967ST36, 1972WA32, 1974HA12, and 1980LA02, for experimental studies, and 1977IS05, for theoretical studies. The emitted β particle can ionize the electron cloud of the daughter nucleus, ^{99}Ru, distorting the β spectrum. This phenomenon is negligible in almost all applications, since its probability is about 0,05 % per emitted β.

3 Atomic Data (Ru, Z=44)

3.1 X Radiations and Auger electrons

The X-ray and Auger electron data were computed using the EMISSION program with the atomic data of Schönfeld and Janßen (1996SC06).

4 Radiation Emissions

4.1 Electron Emission

The β^- intensities were evaluated as described above in Section 2.

4.2 Photon Emission

The details of the photon emission evaluation are in Section 2. ^{99}Ru decays from its first excited state at 89,52 (15) keV, with a half-life of 20,36 (25) ns, and a γ-ray multipolarity of M1 + 71,0 (5) % E2. The absolute γ-ray emission intensity is evaluated as 5,8 (11) x10^{-4} %, leading to a β^- branching to ^{99}Ru*(89 keV) of 1,45 (30) × 10^{-3} %.

5. References

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