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The Decay of ^{99}Mo

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NOTES
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ABSTRACT

Relative intensities for K x-rays and gamma rays emanating from ^{99}Mo in equilibrium with its $^{99}\text{Tc}^*$ daughter have been measured using several Ge photon detectors. Combining these intensities with an evaluated set of electron-conversion coefficients has provided a set of absolute intensities for the observed gamma rays. The absolute intensity for the dominant 140.5-keV gamma ray in ^{99}Tc was determined to be $90.7 \pm 0.6/100$ ^{99}Mo disintegrations for ^{99}Mo decay in equilibrium with decay of the $^{99}\text{Tc}^*$ daughter.

INTRODUCTION

The decay properties of 66-hr ^{99}Mo and its 6.0-hr daughter $^{99}\text{Tc}^*$ have been extensively studied,¹⁻⁵ and these results have been summarized recently in the Nuclear Data Sheets.⁶ However, despite the comparative ease in obtaining high-precision data, there remain rather large discrepancies among reported experimental values for branching ratios in the decay of $^{99}\text{Mo} - ^{99}\text{Tc}^*$. Very recently, two evaluations (of the existing data) have been published;^{7,8} the discrepancies were not resolved. It is important to resolve them, if possible, for the practical reason that the cumulative yield for ^{99}Mo is close to the maximum for fission of U and Pu isotopes; hence radioassay via its emissions is important in total fission determinations. Furthermore, the 140-keV gamma ray is the strongest observed line in the decay of $^{99}\text{Mo} - ^{99}\text{Tc}^*$, and is near the maximum of photon detection efficiency for standard large-volume Ge(Li) detectors. Measuring the yield of the 140-keV photon in a gross fission-product spectrum 60 hours or so after thermal-neutron fission of ^{235}U is quite straight forward and should provide an accurate determination of the amount of ^{99}Mo in the sample. This method should be as accurate as the present technique of counting ^{99}Mo decay betas, and has the advantage of circumventing chemical separation of molybdenum, provided the intensity of the 140-keV gamma-ray decay can be established to the required precision. Because we are involved in a program very dependent upon fission determination,⁹ we have remeasured the intensities of the dominant x- and y-rays, and have compared our values to existing experimental and evaluated data.

EXPERIMENTAL DETAILS

Three different germanium detectors were used: (a) a 90 cm³ coaxial Ge(Li) detector capable of 2.1-keV resolution at $E_\gamma = 1.33$ MeV, (b) a 10 cm³ intrinsic planar Ge detector having about 2.4 keV resolution to 0.66-MeV photons, and (c) a small-volume intrinsic planar Ge detector (LEPS) capable of 0.51-keV resolution at $E_\gamma = 0.122$ MeV. Using different detectors permitted high-resolution study for the lower-energy photons, good efficiency for the higher-energy photons, and a variety of efficiency calibration curves. It should be pointed out, however, that for all three detectors the efficiencies at $E_\gamma = 122$ and 136 keV were obtained using the same ⁵⁷Co source which has an uncertainty[†] in source strength of 1.1% at the 1σ confidence level. Of some concern is the branching ratio of the 136-keV photon in this source. We used 10.61% given in the evaluation of Ref. 7. Efficiencies at other photon energies were determined using standard well-calibrated sources (uncertainties[†] in source strength between 0.7% and 1.7% at the 1σ confidence level) supplemented by several multi-gamma-ray sources for which precise absolute source strengths had to be determined by adjusting the relative data to the absolute values in the efficiency curve. For the 90 cm³ detector, the two efficiency values obtained for $E_\gamma = 0.2$ and 1.8 MeV were used to determine the parameters, C_0 and x of the approximate efficiency function

$$\epsilon(E_\gamma) = C_0 E_\gamma^{-x}$$

Then the deviations of the measured efficiency values for $E_\gamma > 0.2$ MeV were plotted, and a smooth curve was drawn through the plotted points. The technique is the same as Debertin *et al.*,¹⁰ and appears to us to

[†] Uncertainties according to information from the supplier of the source(s).

provide absolute $\epsilon(E_\gamma)$ to 0.9% at the 1 σ confidence level for $E_\gamma > 0.2$ MeV. For the three detectors and for $E_\gamma < 0.2$ MeV, we relied upon the assigned source calibrations and branching ratios and the expected behavior of the efficiency curves. The absolute uncertainty in $\epsilon(E_\gamma)$ for $E_\gamma < 0.2$ MeV is assigned as 2 1/2% at the 1 σ confidence level for the 90 cm³ detector and as 3% for the other two detectors based upon the scatter in the efficiency data about the chosen curves.

The ⁹⁹Mo sources were made by the ⁹⁹Mo (n, γ) reaction and utilizing standard chemical preparation techniques. The main source was deposited upon a 1 mg/cm² Mylar foil and covered with a similar foil. The initial source strength was ~ 0.15 MBq. Several weaker sources were prepared by aliquot methods on different mountings; one in particular was studied using a standardized beta-ray proportional counter to give its strength to about 5%. The present γ -ray measurements were initiated 3 days after chemical preparations to ensure equilibrium of the ⁹⁹Tc daughter with the ⁹⁹Mo parent.

EXPERIMENTAL RESULTS

The present data are given in Table 1, along with recent evaluated values and data from three previous experiments which has been obtained using Ge(Li) high-resolution detectors. The present results were obtained for $E_\gamma > 180$ keV using the 90 cm³ detector, for the x-rays of $E_\gamma = 18.3$ and 20.6 keV using the LEPS detector, and for the other two lines an average of measurements using two, or all three, detectors. In addition, we took advantage of the very good LEPS resolution to obtain information on the very weak 142-keV transition.

Table 1. ^{99}Mo Decay: Relative Yields for Principal Gamma and X-rays at Equilibrium with ^{99}Tc Isomer

E_γ (keV)	Experimental				Evaluated	
	Doyle ^a	Halford ^b	APC ^c	Present	ORNL ^d	CRSRR ^e
18.3				78.7 ± 3.2	76.3 ± 3.2	
20.6				14.3 ± 0.6	14.1 ± 1.0	
40.3		9.9 ± 0.8	4.9 ± 1.8	8.66 ± 0.27	10.5 ± 0.8	6.62 ± 0.44
140.5	636	706 ± 45	730 ± 22	761 ± 12	667	726 ± 26
142.6	48.7 ± 1.1	49.9 ± 3.4	49.8 ± 2.9	50.1 ± 0.7	50.2 ± 1.1	53.5 ± 2.1
146.3	10.8 ± 0.7	10.7 ± 0.6	10.5 ± 0.7	9.52 ± 0.32	10.5 ± 0.4	11.6 ± 0.8
170.4	100 ± 4	100	100 ± 5	100	100 ± 3	100
777.9	35.5 ± 1.0	36.6 ± 1.6	35.8 ± 2.2	35.8 ± 0.9	35.5 ± 1.5	36.9 ± 2.6
823.8	1.06 ± 0.07	1.11 ± 0.08	1.04 ± 0.07	1.09 ± 0.05	1.08 ± 0.05	1.15 ± 0.03
960.7	0.79 ± 0.06	0.79 ± 0.06	0.80 ± 0.06	0.86 ± 0.04	0.78 ± 0.06	0.82 ± 0.01
	^a Ref. 2	^b Ref. 3	^c Ref. 1		^d Ref. 7	^e Ref. 8

The relevant portion of the spectrum is shown in Fig. 1. These data required about 60 hours of counting time, in which about every twelve hours the source was moved closer to the detector, while keeping the overall analyzer deadtime to < 2%. A polyethylene absorber (150 mg/cm²) was used to reduce the beta-ray contribution to the spectrum. The solid curve shown in the figure indicates the estimated "peak" due to the 142-keV gamma ray assuming (a) that the underlying Compton distribution can be correctly estimated from the slightly higher energy data, (b) that the peak shape is the same as for the 140-keV gamma ray, and (c) that only the 142-keV gamma ray accounts for the observed "peak." Under these conditions we obtain

$$I_\gamma(142.6)/I_\gamma(140.5) = (2.0 \pm 0.3) \times 10^{-4},$$

a value smaller than $(3.0 \pm 0.6) \times 10^{-4}$ reported by Van Eijk *et al.*²

Fig. 1. Portion of Gamma-ray Spectrum vic. $E_\gamma \sim 140 \text{ keV}$. This spectrum was obtained using a high-resolution intrinsic Ge detector to determine the magnitude of the $E_\gamma = 142.6 \text{ keV}$ contribution with respect to the $E_\gamma = 140.5 \text{ keV}$ yield. The vertical dot-dash line vic. channel 460.5 is the expected centroid of the 142.6-keV peak. The horizontal dashed line vic. 271 counts/channel is the average of the data between channels 470 and 480, and is assumed to be the magnitude of Compton distribution between channels 453 and 469. With this assumption, and assuming that a peak corresponding to a 142.6-keV gamma ray has the same shape as that observed for the 140.5-keV gamma ray but centered at channel 460.5, the solid line is the best fit to the observed data.

There are ~ 20 previously reported^{2,3} gamma rays not presented in this report; their ~ 0.5% contribution will be considered in aggregate in the next section.

As mentioned above the gamma-ray intensity data, I_Y , are given in Table 1, normalized to the 740-keV gamma-ray yield. This normalization emphasizes an important feature, viz that for the six highest-energy gamma rays in Table 1 the relative yields are in good agreement. The relative values for the dominant 140.5-keV gamma ray show poor agreement, but it is just this gamma ray for which we need an accurate value of photons/decay. The difficulty, of course, is the effect on any measurements due to the 6.0-hr lifetime of the isomer in ⁹⁹Tc. If a measurement is made before the daughter isomer has reached equilibrium with the parent, the ratio $I_Y(140.5)/I_Y(739.4)$ will be smaller than the ratio at equilibrium. The same effect can be caused by unsuspected loss of the ⁹⁹Tc* isomer from the source.

There is also a difference between the present I_Y for $E_Y = 40.5$ keV and the previous experimental value as well as the two evaluated values. The present value was determined from measurements using the two planar detectors. For the LEPS detector the efficiency, $\epsilon(E_Y = 40.5 \text{ keV})$, is very near the maximum efficiency for this detector. The efficiency for $E_Y = 43.5$ keV was obtained to an accuracy better than 1% using a ²³⁸Pu metal source for which the source strength had been determined by α counting.

EVALUATION OF THE PRESENT DATA

Figure 2 presents the relevant portion of the ^{99}Tc level structure and the several transitions being considered in this section. The band labelled "other levels" is intended to represent all the other levels, some of which are lower in excitation energy than the 920-keV level shown, and the dashed lines shown emanating from the "other levels" and the 920-keV level represent the gamma rays not measured by us. The dashed 2.12-keV transition depopulating the 6.0-hr isomer was also not studied.

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Fig. 2. Partial Schematic Level Diagram of ^{99}Tc . The principal gamma rays discussed in the present report are indicated by the arrows having widths roughly proportional to intensity. Energies and log ft values for three beta groups are also shown. Intensities are discussed in the text.

a. Internal-conversion Coefficients.

Needed to obtain transition rates are the total conversion-electron coefficients α_T , and to compare with our x-ray data α_K values are needed. Those used are given in Table 2. We summarize briefly the reasons for using the values listed.

Table 2. ^{99}Mo Decay: K- and Total-Conversion-electron Ratios for the Principal Transitions Observed

E_t (keV)	α_K	α_T
40.5	3.27 ± 0.10	3.76 ± 0.12
140.5	0.099 ± 0.005	0.113 ± 0.006
181.1	0.126 ± 0.004	0.140 ± 0.005
366.5	0.072 ± 0.001	0.081 ± 0.002
739.4	0.0016 ± 0.0004	0.0018 ± 0.0004

The most critical value given in the table is $\alpha_K(E_t = 140.5) = 0.099 \pm 0.005$. This value for α_K was computed from the tables of Hager and Seltzer¹¹ for an M1 transition with a 1.4% E2 admixture.¹² The 5% uncertainty assigned to α_K reflects the added uncertainty to the usual 3% assignment¹³ because of the rapid change in α_K with admixture. This value of α_K is within error of the experimental value obtained by Van Eijk *et al.*² Our x-ray yield data (discussed below) might favor a somewhat larger value of α_K . For $E_t = 140.5$ keV, α_T is determined from α_K and the reported K/L/MN values.^{6,11}

For $E_t = 181$ keV, $\alpha_K = 0.126$ as computed for a pure E2 transition using the tables of Hager and Seltzer,¹¹ since the measured mixing ratio

is very small.¹² This value is within the experimental uncertainties of prior measurements.^{3,14} α_T was computed using the measured K/L ratio and computing K/M for an E2 transition. As discussed below, the evaluation of the contribution to the total conversion-electron spectrum indicates that α_K and α_T for $E_t = 181$ keV are not very important.

More important are the α_K and α_T for the $E_t = 40.5$ keV transition. The reported¹⁴ measured value for α_K is 3.7 ± 0.5 , but we used 3.27 calculated¹¹ for a pure M1 transition because the reported E2 mixing is very small.¹² The computed α_T is 3.76, which value is in reasonable accord with the present data as applied to the population and depopulation of the $E_x = 181$ keV level.

The remaining conversion-electron coefficients are small, and have a minor effect on the overall evaluation. They become important for a discussion only when the accuracy goal is $\ll 1\%$.

b. The 181-keV Level.

There is no measured direct beta decay to this level, and the sensitivity of the experiment of Nagarajan et al.⁴ was such that they would likely have identified such a transition of $> 0.5\%$. Therefore, we assume that the sum of the intensities of the transitions populating the 181-keV level must balance the sum of the intensities depopulating the level. For the former we have

$$\Sigma_{in} = I_t(740) + I_t(823) + I_t(961) + \text{others.}$$

Combining our I_γ of Table 1 with α_T of Table 2 and estimating the "others" contribution from Van Eijk et al.,² and/or Cook et al.,³ gives

$$\Sigma_{in}(181) = 102.1 \pm 0.4$$

based on $I_Y(740) = 100.0$, and no assigned uncertainty to $I_Y(740)$.

For deexcitation we have

$$\Sigma_{out} = I_Y(181) + I_e(181) + I_Y(40.5) + I_e(40.5).$$

Combining the I_Y of Table 1 with α_T of Table 2,

$$\Sigma_{out} = 98.9 \pm 2.6,$$

where most of the error is associated with $I_e(40.5)$.

c. The 140.5-keV Level.

There is no measured direct beta decay to this level, but there is the contribution from decay of the 142-keV 6-hr isomer. The estimate for the total $I_t(2.1 \text{ keV})$ is

$$\begin{aligned} I_t(2.1) &= I_Y(140) + I_e(140) - I_t(40.5) - \text{others} \\ &= 789.8 \pm 16.6 \end{aligned} \tag{1}$$

where most of the uncertainty is due to the uncertainty in $I_Y(140)$ and where the "others" consists of two very weak transitions (total intensity ≈ 0.22) assigned as decays to the 140.5-keV level.^{2,3}

d. K x-ray Yields.

From Table 1, the total measured K x-ray yield is

$$\begin{aligned} \Sigma_x &= I_Y(18.3) + I_Y(20.6) \\ &= 93.0 \pm 3.8. \end{aligned}$$

This value may be compared with

$$\Sigma_x = 0.793 \Sigma_K, \text{ and}$$

$$\Sigma_K = I_K(40) + I_K(140) + I_K(142) + I_K(181) + I_K(\text{others}),$$

where the factor 0.793 ± 0.031 is the K-fluorescence yield obtained from Bambynek et al.¹⁵ We obtained $I_K(142)$ from the measurement of Geiger¹⁶ of the ratio $K(142)/K(140) = 0.0746 \pm 0.0011$. The remaining I_K were obtained from the data in Tables 1 and 2.

$$\Sigma_K = 115.1 \pm 4.7,$$

so therefore

$$\Sigma_x = 91.3 \pm 5.0.$$

The agreement is very good, even to the point of suggesting that some of the assigned uncertainties have been overestimated.

e. Total Relative Ground-state Decay and the Normalization to 100 ⁹⁹Mo Decays.

The total relative ⁹⁹Mo decay is given by

$$\Sigma_I = I_t^*(140) + I_t^*(142) + I_t(181), \quad (2)$$

ignoring a very weak 762-keV gamma-ray reported by Cook et al.,³ and assigned as a ground-state transition. The asterisks were used to call attention to the need to correct these transitions for the 6.0-hr lifetime of ⁹⁹Tc* as follows:

$$I_t^*(140) = I_t(40.5) + I_t(2.1)/1.10;$$

$$I_t^*(142) = I_t(142)/1.10 .$$

The factor 1.10 is the ratio of the $^{99}\text{Tc}^*$ activity to that of the parent ^{99}Mo when equilibrium has been reached. $I_{\gamma}(142)$ is obtained from $K(142)$ from the last section, from measured K/L and estimated K/M ratios, and from the measured I_{γ} discussed above.

$$\Sigma_T = 823.6 \pm 18.0$$

$$N = 100/\Sigma_T = 0.1214 \pm 0.0027 . \quad (3)$$

The overall uncertainty in the normalization per 100 ^{99}Mo decays (N) is dominated by the uncertainty assigned to $I_{\gamma}(140)$ in Table 1.

f. Beta-ray Branching Ratios.

From these data, the intensities for the principal beta rays may be determined. These are presented in Table 3, along with comparisons from recent evaluations^{7,8} and measurements.⁴

Table 3. ^{99}Mo Decay: Intensities for Principal Beta Rays Per 100 Decays

E_{β} (keV)	Measured I_{β}	Evaluated I_{β}		
	Andhra ^a	ORNL ^b	CDNSNR ^c	Present
437	14	17.6 ± 0.5	15.6 ± 0.5	16.5 ± 0.3
848	2	1.33 ± 0.06	1.4 ± 0.1	0.8 ± 0.1
1214	84	80.7 ± 0.8	83.0 ± 1.0	82.7 ± 0.6

^aRef. 4.

^bRef. 7.

^cRef. 8.

g. Overall Uncertainty on $A_Y(140)$.

Labelling the intensity per 100 ^{99}Mo decays of the 140.5-keV gamma ray from the decay of ^{99}Mo in equilibrium with its daughter ^{99}Tc as $A_Y(140)$, then

$$A_Y(140) = N \cdot I_Y(140)$$

where N is given by Eq. (3) and $I_Y(140) = 747$ from Table 1.

$$A_Y(140) = 90.7/100 \text{ decays.}$$

The next step is to estimate the uncertainty on A_Y . To do this we observe from Eq. (2) and remarks following it that

$$A_Y(140) = I_Y(140)/[aI_Y(140) + b]. \quad (4)$$

One may readily verify that

$$a = \frac{1}{1.1} \left\{ 1 + \alpha_K(140) \left[T(140) + \frac{K(142)}{K(140)} T(142) + \frac{K(181)}{K(140)} T(181) \right] \right\}$$

where $T(E_c)$ is the ratio of the total conversion to K conversion for the transition E_c . This equation exhibits the reliance on the $K(142)/K(140)$ conversion-electron ratio measured by Geiger.¹⁶

In addition, the constant of Eq. (4) is

$$b = 0.091 I_c(40.5) + I_Y(181) + 0.91 [I_Y(142) - \text{others}]$$

where the "others" are those in Eq. (1). Numerically,

$$a = 1.03$$

$$b = 54$$

to a precision sufficient for this discussion. Since $I_Y(140) = 747$, it is apparent from Eq. (4) that the uncertainty in $I_Y(140)$ is a small contribution to $\Delta A_Y(140)$. In fact, using the wide variety of $I_Y(140)$ given in Table 1 in just the same fashion as done above does not vary $A_Y(140)$ by more than 1%. Further, a 5% error on the constant b will affect $A_Y(140)$ by only 0.3%, and the uncertainty assigned to b is < 5%.

The largest contribution to the uncertainty of the multiplier a is due to $\alpha_K(140)$ which has an assigned uncertainty of 5%. This uncertainty will give a 0.7% uncertainty in a , which in turn is reflected as 0.6% uncertainty in $A_Y(140)$. [One may argue for reducing the 5% uncertainty assigned to $\alpha_K(140)$ on the strength of the results of the K x-ray yield evaluation.] Including the uncertainty assigned to b results in assigning a 0.7% uncertainty to $A_Y(140)$, an uncertainty consistent with the present accuracy, and by the same token, not overly conservative.

FINAL RESULTS

The final absolute yields are given in Table 4, along with assigned uncertainties. For all but $E_Y = 140$ keV the assigned uncertainties reflect values in Table 1 and the uncertainty in N given in Eq. (3). Also noted in Table 4 is the contributions of the 140.5-keV gamma ray due to population of the $E_x = 142.5$ keV level by the 40.5-keV transition (labelled "prompt"). The remainder is due to decay of the ^{99}Tc isomer in equilibrium.

The present data may also be used to deduce the intensity of 140.5-keV gamma ray decay from the ^{99}Tc isomer in the absence of the ^{99}Mo

parent. The result is 87.2 ± 0.5 140.5-keV gamma rays per 100 ^{99}Tc isomer decays. The analysis of the uncertainty was the same as given above (in section g), and is dominated by the 5% uncertainty assigned to $\alpha_K(140)$.

Table 4. ^{99}Mo Decay: Absolute Yields for Principal Gamma and X-rays, at Equilibrium with ^{99}Tc Isomer

E_γ (keV)	I_γ (per 100 decays)
18.3	9.55 ± 0.49
20.6	1.74 ± 0.09
40.5	1.074 ± 0.044
140.5	90.7 ± 0.6
Prompt 140.5	5.02 ± 0.26
181.1	6.08 ± 0.16
366.5	1.16 ± 0.05
739.4	12.14 ± 0.22
777.9	4.36 ± 0.16
823.0	0.132 ± 0.008
960.7	0.102 ± 0.006

CONCLUSIONS

As mentioned in the Introduction, measuring the yield of the 140.5-keV gamma ray in a gross fission-product spectrum using a standard large-volume Ge detector should provide a means for determining the total number of fissions created in the ^{235}U sample by thermal neutrons. The 0.7% uncertainty associated with $A_Y(140.5)$ is less than the $\sim 1\%$ uncertainty assigned¹⁷ to the cumulative fission yield for ^{99}Mo in thermal-neutron fission of ^{235}U . For the present program⁹ the largest uncertainty in the determination of the number of fissions (n_f) is 2 1/2% associated with the efficiency determination¹⁸ of our Ge detectors. Thus, the $\Delta A_Y(140)$ is a small contribution to Δn_f .

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