γ Transitions in the Decay of ^{99}Mo

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The internal conversion spectrum of γ transitions in the decay of ^{99}Mo has been re-studied using a high resolution double focusing β-ray spectrometer. In addition to γ-rays previously reported, seven more γ-rays could be observed. Internal conversion coefficients and multipolarities of γ transitions are given. Energies, spins and parities of high excited levels in ^{99}Tc are confirmed.

The decay properties of ^{99}Mo were studied earlier by several groups [1–7] and more recently by BASHANDY et al. [8] and VAN EIJK et al. [9] using different techniques. In spite of the different methods of investigation in [8] and [9], the energies and internal conversion results for the low γ rays are in excellent agreement. The new γ-ray of energy (989.37 ± 1.02) keV observed in the conversion electron spectrum [8] was confirmed by the γ measurements [9]. However, VAN EIJK et al. [9] reported more new γ rays in the region $\sim 300-600$ keV and above one MeV. These γ transitions could not be clearly identified in the conversion electron spectrum [8] due to the high β background and low conversion electron intensities. In our previous paper [8] conversion coefficients of low energy γ rays and some high energy γ rays with known intensities were studied and utilized for assigning possible spins and parities to the levels of ^{99}Tc. According to the extensive studies [9] of β and γ transitions following the decay of ^{99}Mo, new levels at 671, 761, 1004, 1142, 1169, and 1197 keV have been reported [9]. The level structure of ^{99}Tc has been investigated theoretically by GOSWAMI and SHERWOOD [7] who proposed several levels. However, the agreement between the experimental level scheme given by VAN EIJK et al. [9] and that calculated theoretically is extremely poor. Also the spin and parity assignments for most of the new levels are questionable. Therefore, further studies were necessary in order to draw more definite conclusions about the new levels and their properties. In the present contribution interest has been focused on search for the new γ rays and on the multipolarity assignments, with emphasis on transitions in the ^{99}Mo decay. Measurements of several internal conversion coefficients are reported from which information on excited states of ^{99}Tc could be deduced.

1. Experimental Procedures

The radioactive ^{99}Mo sources used in the present work were produced by sputtering molybdenum oxide (enriched to $\sim 99\%$ ^{98}Mo), onto aluminium foils of thickness 0.7 mg/cm^2 which then were irradiated in a thermal neutron flux of $\sim 10^{14}$ n/cm$^2\cdot$sec. The time of irradiation was about 12 days continuously. The thickness of the material deposited was estimated in each source to be $\sim 80\ \mu$g/cm^2. The area was 1.5×0.2 cm^2. Before the measurements were started, the activity was allowed to decay for a necessary period so that the short lived activities, which may be produced would not disturb the measurements.

The study of the conversion electron spectrum was carried out using a double focusing β-ray spectrometer type GП П-2 ($\varrho_0 = 22.5$ cm) of the Nuclear Physics Department. This spectrometer utilizes a Geiger-Müller tube as a detector. The front window was about 2 mg /cm^2 mica. In most of the investigations described here, a resolution in momentum of $\sim 0.3\%$ was used. However, for the high energy electron spectrum it was necessary to use a momentum resolution up to $\sim 0.6\%$ in order to detect the weak internal conversion lines.

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Present work Transition energy keV	VAN EIJK et al. [9] Transition energy keV	VAN EIJK et al. [9] Relative γ-intensities	Present work Experimental α_k	SLIV and BAND [10] Theoretical α_k	Multi-polarity
40.69 ± 0.05	40.585 ± 0.002		3.7 ± 0.5	3.2 (M 1)	M 1
139.90 ± 0.14	140.511 ± 0.006	100			
142.35 ± 0.14	142.63 ± 0.03	0.03 ± 0.006	30 ± 3	29.5 (M 4)	M 4
180.92 ± 0.18	181.06 ± 0.04	7.5 ± 0.2	0.133 ± 0.02	0.126 (E 2)	E 2
	249 ± 1	0.006 ± 0.003			
	344 ± 2	0.0013 ± 0.0005			
368.71 ± 0.40	366.4 ± 0.1	1.63 ± 0.10	0.072 ± 0.001	0.0077 (M 1)	M 1
381.22 ± 0.42	380.7 ± 0.2	0.011 ± 0.003	0.009 ± 0.001	0.007 (M 1) 0.01 (E 2)	M 1 + E 2
409.13 ± 0.62	409 ± 2	0.0016 ± 0.0007	0.006 ± 0.0008	0.0058 (M 1)	M 1
411.31 ± 0.50	411.5 ± 0.5	0.027 ± 0.003	0.003 ± 0.0005	0.00226 (E 1)	E 1
460.01 ± 0.63	458 ± 1	0.006 ± 0.003	0.0054 ± 0.0006	0.0056 (E 2)	E 2
528.74 ± 0.53	528.9 ± 0.2	0.06 ± 0.007	0.005 ± 0.0006	0.0041 (E 2)	E 2
620.05 ± 0.64	620.7 ± 0.2	0.027 ± 0.004	0.002 ± 0.0004	0.0022 (M 1)	M 1
	620.7 ± 0.2	0.005 ± 0.002			
741.37 ± 0.76	739.7 ± 0.1	15.4 ± 0.6	0.0016 ± 0.0004	0.0015 (E 2 or M 1)	M 1 or E 2
780.23 ± 0.80	778.2 ± 0.1	5.4 ± 0.3	0.0005 ± 0.0001	0.0004 (E 1)	E 1
824.50 ± 0.92	823.1 ± 0.1	0.16 ± 0.01	0.0004 ± 0.0001	0.00037 (E 1)	E 1
950.55 ± 0.97	961.0 ± 0.2	0.12 ± 0.01	0.0024 ± 0.0005	0.0021 (M 2)	M 2
989.37 ± 1.02	988.2 ± 0.5	0.0020 ± 0.0007			
1001.9 ± 1.2	1001.7 ± 0.2	0.007 ± 0.002	0.0018 ± 0.0003	0.0017 (M 2)	M 2
1017.3 ± 1.5	1016 ± 2	0.0010 ± 0.0005			

Table 1. γ transitions in the decay of ^{99}Mo.

1.1. Internal Conversion Electron Measurements

The study of the ^{99}Mo decay was initiated by scanning the conversion electron spectrum in the energy range where the new γ transitions recently reported [9] could be found. In our previous paper [8] we reported precise conversion electron energies especially in the low energy range. However, in the intermediate and high energy range where the β background is high the conversion lines were very weak and it was difficult to determine them precisely. It is necessary to use more strong sources as well as more standard sources for calibrations.

In this investigations the energy region from 200 – 1100 keV was searched for conversion lines. The conversion lines of the 84 keV transition in ^{170}Yb, the 412 keV transition in ^{198}Hg and the 661 keV transition in ^{137}Ba as well as high transitions in ^{187}Re previously measured with the instrument, were used for energy calibrations. The momentum resolution was varying between 0.3 and 0.6 percent depending on the accuracy required and the need to decrease the continuous β-ray spectrum as much as possible, in some energy region, without spoiling the statistics. Possible source asymmetry was compansated for. Lines that were not fully resolved, by the spectrometer, were separated graphically making use of the known shape of a monoenergetic line. A large number of sufficiently isolated lines in the electron spectrum itself yielded the necessary information about the line shapes in the various energy regions. A typical example of the data taken during this investigation is displayed in Figures 1 and 2. The observed conversion electron spectrum was carefully analysed and several γ transitions are ascribed to the decay of ^{99}Mo.

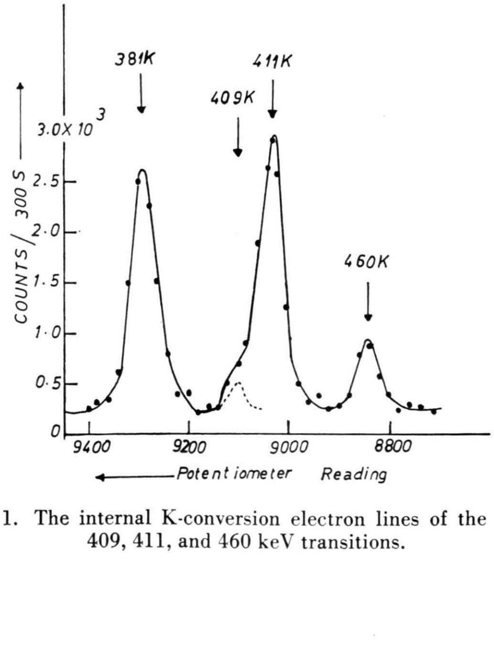

Fig. 1. The internal K-conversion electron lines of the 381, 409, 411, and 460 keV transitions.

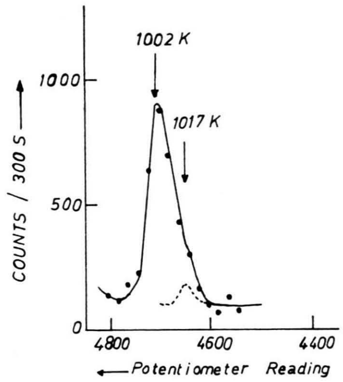

Fig. 2. The internal K-conversion electron lines of the 1002 and 1017 keV transitions.

The intensities of the internal conversion lines were determined from the measured areas under the peaks, taken into consideration the corrections for the absorption in the mica counter window as well as the corrections for the decay. In deducing the areas of partially resolved lines, it was found convenient to subtract the background and β-continuum rates. The electron line intensities were determined from measurements relative to the K-conversion line of the 140 keV transition.

1.2. Internal Conversion Coefficients and Multipole Assignments

In our previous work, due to the lack of gamma intensities, the internal conversion coefficients of only five transitions could be determined. After the work published by VAN EIJK et al. [9] on γ intensities in the decay of ^{99}Mo it is possible, by combination with our persent conversion study, to determine the internal conversion coefficients of most γ transitions in the decay of ^{99}Mo. By means of the photon intensities and conversion electron data, we have calculated the absolute conversion coefficients which are present ed in the table. Normalization between the two series of data is obtained by assuming that the 140 keV transition is a pure M 1 transition as it is proved from its K/L ratio. In order to determine the multipole orders of the transitions, the values of conversion coefficients are compared with theoretical ones interpolated from tables calculated by SLIV and BAND [10].

[10] L. SLIV and I. M. BAND, Alpha-, Beta- and Gamma-Ray Spectroscopy (ed. by K. SIEGBAHN), North Holland Publishing Co., Amsterdam 1965.

2. Results and Discussion

From the carefully studied internal conversion spectrum of the transitions in ^{99}Tc, more γ rays could be observed and added to our previous [8] data. The transition energies determined in the present investigation are summarized in the table together with the transition energies measured by VAN EIJK et al. [9]. There is excellent agreement between the two. However, the two γ-rays of energies 249 and 344 keV were very difficult to confirm from our study, since they are embedded in a high β background. The 368.71 keV transition was reported in our previous work [8] as 372.05 keV. Due to the precision studies and recalibration of the instrument especially in the energy range above 300 keV, it was possible to detect the γ rays of energies 368.71, 381.22, 409.13, 411.31, 460.01, 528.74 and 620.05 keV. The 620 keV transition was reported [9] as a doublet from coincidence measurements, but this is difficult to confirm. At higher energies, the results are in good agreement except for the 950 keV transition which was reported [9] as 961.0 keV.

The proposed [9] level scheme of ^{99}Tc is based on the studies of γ and β transitions following the decay of ^{99}Mo and coincidence measurements. In order to obtain the properties of these levels, the multipolarities of the transitions feeding and deexciting them has to be determined. The internal K-conversion coefficients and the multipole order of the γ transitions in the decay of ^{99}Mo were determined and given in the table. The absolute K-conversion coefficients of the low energy 40 keV, 142 keV and 181 keV transitions were determined. The multipolarities obtained are consistent with the previous results [8] obtained from the K/L-conversion ratios.

The low lying levels in ^{99}Tc have been fully discussed in [8]. Therefore, the discussions here will be restricted on levels above the 511 keV level, see Fig. 3.

The spin and parity of the 511 keV level have been discussed [8] before to be 3/2 —. The only argument is about the value of 511 instead of 509 keV given by other authors. This is due to the 369 keV γ-ray found in our measurements compared to the 366 keV reported by VAN EIJK et al. [9]. However, CRETZU and HOHMUTH [4] have reported a γ ray of energy 370 keV which is in agreement with our result (368.7 ± 0.4) keV. The 671 keV, 5/2—

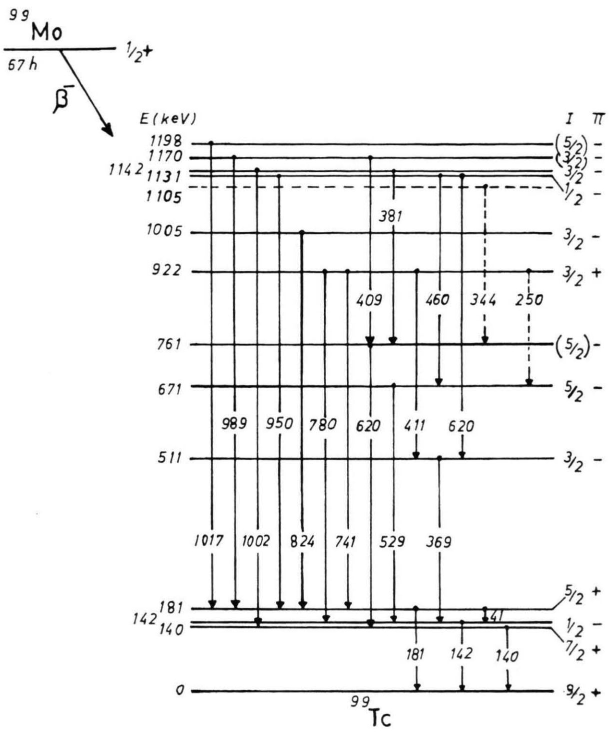

Fig. 3. The decay scheme of ^{99}Mo.

level was proposed by VAN EIJK et al.[9] as a result of coincidence measurements. According to the log *ft* value of the β transition feeding the 671 keV level, a spin assignment of $5/2-$ is suggested. The 671 keV level is de-excited by the 529 keV γ transition to the 142 keV, $1/2-$ level. The K-conversion coefficient of the 529 keV transition shows that it is a pure electric quadrupole transition, see the table, which confirms the $5/2-$ assignment for the 671 keV level in ^{99}Tc. The 761 keV level is also suggested by VAN EIJK et al.[9] due to the coincidence found between the 620 keV transition and the 140 keV, 381 keV, 409 keV γ rays. The appearance of the 381 and 409 keV transitions in the present conversion spectrum confirms the existence of the 761 keV level. From the log *ft* value of the β branch feeding this level together with the multipolarities of γ rays feeding and de-exciting it, $5/2-$ assignment is given to the 761 keV level. The 922 keV level was well established before[8]. A $3/2+$ value is the most probable assignment for the 922 keV excited level which is also compatible with theory. VAN EIJK et al.[9] have reported a new γ ray of energy 249 keV decaying from the 922 keV level to the 671 keV level. This γ ray could not easily be observed in our conversion electron spectrum. The 1005 keV level is unambiguously determined[9] since its position is based upon the fact that the 824 keV transition is coincident with the 181 keV γ ray. Besides the 824 keV transition is not in coincidence with the 369 keV γ ray. The log *ft* value 8.1 of the β transition to the 1005 keV level suggests that its spin is $1/2-$ or $3/2-$. A spin $3/2-$ is proposed, since the 1005 keV level decays to the $5/2+$ level at 181 keV. Also the E 1 character of the 824 keV transition found in our study confirms the $3/2-$ assignment. A new level at 1105 keV was suggested[9] in order to explain the coincidence found between the 620 keV and a weak 344 keV transition. However, this level is doubtful since the 344 keV transition could not be observed in the electron conversion spectrum. And consequently no assignment could be given to it. The 1131 keV level was established[8] before and confirmed by coincidence measurements. γ-γ-conincidence spectra with the gate on 620 keV show that the 369 as well as the 381 keV transitions are coincident. From this result, it was concluded that besides the 620 keV transition depopulating the 761 keV level there exists a 620 keV transition which depopulates a level at 1131 keV. This level makes is possible to fit the 460 keV γ ray which is coincident with the 529 keV transition, into the decay scheme. The log *ft* value of the β branch feeding this level was found to be 7.3. This indicates that this β branch is probably nonunique and first forbidden, which suggests that the spin of the level is $1/2-$ or $3/2-$. In our previous paper[8] the $1/2-$ assignment was excluded, since we expect that the level decays by the 989 keV γ ray to the 142 keV, $1/2-$ level. This explanation should be changed after the 989 keV γ ray was found[9] in coincidence with the 181 keV transition. In the proposed decay scheme Fig. 3 the 1131 keV level decays through the 950 keV, the 620 keV, and the 460 keV transitions to the 181 keV, $5/2+$, the 511 keV, $3/2-$, the 671 keV, $5/2-$ levels respectively. The conversion coefficient data found in the present study proved that the 950 keV transition is M 2, the 460 keV transition is E 2 and the 620 keV transition is M 1 which confirm the $1/2-$ assignment to the 1131 keV level. The position of the 1142 keV level is based on the coincidence relations of the 1002 keV γ ray with the 140 keV transition. In addition, the 381 keV transition fits excellently into the decay scheme, and its position explains the coincidences observed[9] between the 381 and 620 keV γ rays. The log ft value of the β branch feeding the 1142 keV level is 7.5, which indicates that a spin assignment of 1/2− or 3/2− is likely. The K-conversion coefficient of the 1002 keV transition determined gives an M 2 character for that transition, while the 381 keV transition is found to have M 1 + E 2 character, see the table. According to these results, the 1142 keV level is assigned as 3/2−. A level at 1170 keV is introduced, since the 989 keV γ ray was found to be in coincidence with the 181 keV transition. The existence of this level fits the replacement of the weak 409 keV transition which is coincident with the 620 keV transition, in the decay scheme. The log ft value of the β branch feeding the 1170 keV level suggesting a spin 1/2−, 3/2− or 5/2−. A spin of 1/2− can be excluded in view of the feeding to the 5/2+, 181 keV level. Of the remaining spin values, 5/2− is somewhat less probable[9] since the log $1/24(W_0^2-1)\,ft$ value is rather low for a unique first-forbidden β transition. The M 1 character for the 409 keV transition may support the 3/2− assignment for the 1170 keV level. A new level at energy of 1198 keV proposed[9] due to the coincidence relation of the 1017 keV γ ray with the 181 keV transition. The appearance of the K-conversion line of the 1017 keV transition in the conversion spectrum confirms the existence of a level at 1198 keV. The study[9] of β transition feeding the 1198 keV level leads to a spin assignment of 5/2−.

On the Behavior of Short-lived Cosmic Ray Produced Nuclides in the Lower Atmosphere

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Some aspects of short-lived cosmic ray produced nuclides in the lower atmosphere, especially of Na^{24}, and their qualities as tracers for atmospheric motion are studied. The Na^{24} production rate has been estimated. The distribution of this nuclide as a function of altitude in the lower atmosphere has been described by a steady state eddy diffusion model. Measurements of Na^{24} in ground-level air have been carried out. The measured activities varied from $0.07\cdot10^{-3}$ to $0.28\cdot10^{-3}$ dpm/m^3. These figures are in good accordance with the calculated values for reasonable diffusion parameters.

Calculations suggest that CRP-nuclides with life-times in the order of a day are useful as tracers for atmospheric motions in the range of eddy diffusion coefficients of about $5\cdot10^4$ to several 10^6 cm^2 sec^{-1}, and for altitudes below four of five kilometers.

Furthermore some relations between the specific radioactivity of rainwater and the specific activity of cloud-level air, with special respect to short-living CRP-nuclides are found. The evaluation of some measurements of Na^{24} activity in rain water shows good consistency between real atmospheric conditions and calculated values.

General Remarks

In the last ten or fiften years many papers have been published on the behavior of long- and medium-lived cosmic ray produced (CRP-) nuclides in the atmosphere. A survey of the production mechanisms of CRP-nuclides as well as of the investigations about transport and distribution of medium- and long-lived isotopes in the atmosphere and the hydrosphere is given by LAL and PETERS[1].

On the other hand, only a few studies have been carried out on the behavior of short-lived CRP-nuclides in the atmosphere. The first detection of a short-living argon spallation product has been reported in 1956 by WINSBERG[2], who has measured the activity of Cl^{39} (55 min half-life) in rain water. Several years later, in 1963, Na^{24} with 15 hrs half-life has been detected in rain water by ROEDEL[3]. Hereafter, several other short-lived argon spallation products have been found: Mg^{28} (21 hrs), Si^{31} (2.6 hrs), S^{38} (2.9 hrs),

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