Radiations from Mo^{99} and Tc^{99m}

MERLE E. BUNKER AND ROBERT CANADA
Indiana University, Bloomington, Indiana*
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The radiations from Mo^{99} (67 hours) and its metastable daughter Tc^{99m} (6 hours) have been measured in a magnetic lens spectrograph. The beta-ray spectrum consists of a group with an end-point energy of 1.23 Mev, one of 0.445 Mev and possibly a third lower energy group. The relative intensities of the 1.23-Mev to 0.445-Mev groups are 4:1. Gamma-rays have been found at 0.040, 0.140, 0.181, 0.367, 0.741, and 0.780 Mev. The gamma-rays at 0.040, 0.140, and 0.181 Mev are internally converted. The spectrum of Tc^{99} consists of one internally converted gamma-ray at 0.140 Mev. A disintegration scheme is proposed.

I. INTRODUCTION

If molybdenum is bombarded with either deuterons or slow neutrons, a 67-hour activity is produced which has been attributed[1] to Mo^{99}. Furthermore, it has been shown that this isotope is in equilibrium with a 6-hour daughter element,[1,2] an isomer of Tc^{99}. Most of the preliminary measurements that have been made on the radiations from these two activities have been absorption and coincidence measurements, and no thorough spectrometric study has previously been reported. While this work was in progress, a similar study was reported by Medicus, Maeder, and Schneider[3] with results quite similar to those reported here. The present work, however, shows certain additional lines not obtained by them.

Seaborg and Segrè[1] separated the technetium activity from the molybdenum activity by chemical means, and reported the radiations from the technetium fraction to consist of a highly converted gamma-ray of energy 0.136 Mev. The molybdenum fraction was reported to emit beta-rays with an end-point energy of 1.5 Mev and gamma-rays of about 0.4-Mev energy. However, Miller and Curtiss[4] investigated the 67-hour activity with a thin magnetic lens spectrometer and reported gamma-rays of 0.24 and 0.75-Mev energy. More recently, Mandeville and Scherb,[5] using absorption and coincidence techniques, found that Mo^{99} emits two beta-ray groups with end-point energies of about 0.24 Mev and 1.03 Mev. Their coincidence measurements indicated that the low energy beta-ray group is coupled with gamma-rays but that the high energy group is not. The maximum energy of the gamma-rays was found to be 0.71 Mev and a small gamma-gamma-coincidence rate was detected. In addition, they isolated the 6-hour technetium activity by chemical separation and found no particle-gamma or gamma-gamma-coincidences, which would indicate that the metastable state decays with emission of a single partially converted gamma-ray.

II. APPARATUS AND SOURCE PREPARATION

The apparatus used in this investigation was a magnetic lens spectrometer of conventional design. The general details of the instrument have been described elsewhere.[6] During these experiments, the spectrometer was adjusted to give a resolution of about 2.5 percent. An end window Geiger counter with an aperture 7 mm in diameter served as the detector of beta-particles. The Zapon counter window used in examining the data had a low energy cut-off of 11–15 kev, depending on the type of experiment performed.

The Mo^{99} used in the present investigation was obtained by bombarding metallic molybdenum with deuterons in the Indiana University cyclotron. This bombardment also produces a number of radioactive technetium isotopes, whose combined activity is much greater than the activity of the Mo^{99}. Since the half-lives of the technetium activities are of the same order of magnitude as that of Mo^{99}, a great deal of care was taken in the process of chemical purification to remove all the technetium. The method used in the chemical purification was similar to that described by Seaborg and Segrè.[1] In the final stage of the procedure, the molybdenum was precipitated with 8-hydroxyquinoline solution. A small amount of this precipitate was dissolved in nitric acid for use as beta-ray source material. The remainder was dissolved in nitric acid and the molybdenum was precipitated as the sulfide with hydrogen sulfide, centrifuged, and dried for use as the gamma-ray source material.

Before any measurements were made, at least thirty-six hours were allowed to elapse in order to permit the 6-hour metastable state to come to equilibrium with the parent activity.

III. THE BETA-RAY SPECTRUM

The beta-ray source had a surface density of 0.2 mg/cm^2 and was about 6 mm in diameter. It was mounted on a thin Zapon film of 0.08 mg/cm^2 surface density.

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[1] G. T. Seaborg and E. Segrè, Phys. Rev. **55**, 808 (1939).
[2] Plutonium Project Report, Rev. Mod. Phys. **18**, 513 (1946).
[3] Maeder, Medicus, and Schneider, Helv. Phys. Acta **22**, 603 (1949).
[4] L. C. Miller and L. F. Curtiss, Phys. Rev. **70**, 983 (1946).
[5] C. E. Mandeville and M. V. Scherb, Phys. Rev. **73**, 848 (1948).
[6] Bunker, Canada, and Mitchell, Phys. Rev. **79**, 610 (1950).

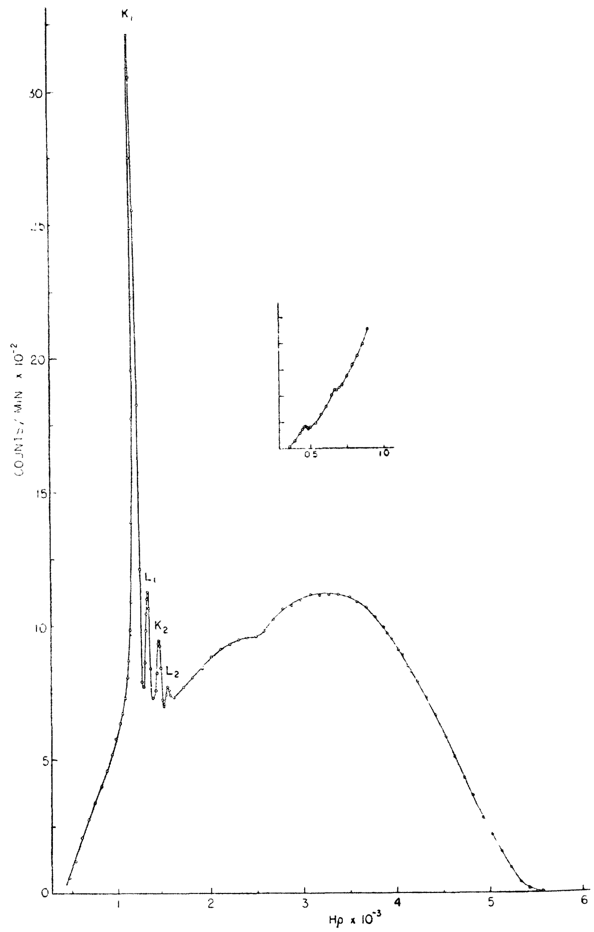

FIG. 1. Beta-ray spectrum of Mo^{99}.

The beta-ray spectrum is shown in Fig. 1. Here the number of counts per minute, corrected for decay, is plotted as ordinate against $H\rho$ (gauss-cm) as abscissa. It can be seen at once that the continuous part of the spectrum is complex. A Fermi analysis has been made of the beta-ray data using the approximation for the Coulomb correction factor, $F(Z, \eta)$, given by Bethe and Bacher.[7] The results are shown in Fig. 2. The end-point energy of the high energy group is 1.23±0.01 Mev and that of the low energy group is 0.445±0.01 Mev. The contribution of these two groups to the low energy portion of the spectrum was calculated by extrapolating the straight lines of the Fermi plot back to zero energy. The ratio of the intensity of the high energy group to that of the low energy group is 4:1. Judging from the appearance of the spectrum below and to the left of the internal conversion lines, it would seem that there is an additional low energy beta-ray group, although it is not possible to say much about its end point energy or intensity due to interference from the group of lines.

Referring to Fig. 1, the peaks at 1228 and 1328 gauss-cm are K and L internal conversion lines corresponding to a gamma-ray energy of 0.140 Mev. Similarly, the peaks at 1457 and 1535 gauss-cm are K and L conversion lines corresponding to a gamma-ray energy of 0.181 Mev. The ratio N_K/N_L was calculated in both cases, although a certain amount of arbitrariness exists in choosing base levels for the lines. For this reason, maximum and minimum values were calculated in the case of the pair of lines of lower energy. The results are shown in Table I.

On the basis of the reports of other investigators, it was assumed that the 0.140-Mev gamma-ray is associated with the 6-hour isomeric state of Tc^{99}. In order to confirm this assumption and to investigate the possibility of the 0.181-Mev gamma-ray also being associated with this (or another) metastable level, the technetium was removed chemically from a portion of the source material, and observation of the internal conversion lines emitted by the molybdenum fraction was begun within three hours after the purification. It was observed that the conversion lines associated with the 0.140-Mev gamma-ray increased in intensity with time at approximately the rate expected from the growth of a 6-hour activity, but that the lines associated with the 0.181-Mev gamma-ray decay with a 67-hour half-life from the beginning of the observations. It is thus concluded that if the 0.181-Mev gamma-ray is associated with a metastable state, this state has a much shorter half-life than 6 hours. Experiments on the Tc^{99} fractions show only the 0.140-Mev gamma-ray (see below).

A later experiment on the beta-ray spectrum of Mo^{99}, in which a thinner window was used, showed two weak internal conversion lines corresponding to K and L lines for a 40-kev gamma-ray. The intensity of the K-line is, no doubt, decreased by window absorption (see inset of Fig. 1).

IV. GAMMA-RAY MEASUREMENTS

The distribution of secondary electrons ejected from a 7 mg/cm² lead radiator by the gamma-radiation of Mo^{99} was examined in the lens, and the result is shown in Figs. 3 and 4. The peaks at 789, 1262, and 1326 denoted by $K_1L_1M_1$, are the K, L, and M photo-electron peaks corresponding to a gamma-ray of 0.140 Mev. The K, L, and M peaks at 1075, 1480 and 1535 gauss-cm, denoted by $K_2L_2M_2$, are due to a gamma-ray of energy 0.181 Mev. Due to the low intensity of the high-energy region of the spectrum the portion of the curve above 1570 gauss-cm is plotted on a larger scale than is used for the rest of the spectrum and is shown in Fig. 4. The K and L peaks which appear at 2014 and 2326 gauss-cm (K_3L_3) are due to a 0.367-Mev gamma-ray and the K and L peaks at 3500 and 3768 gauss-cm (K_4, L_4) correspond to a 0.741-Mev gamma-ray. The peak at 3642 gauss-cm (K_5) is apparently a K line due to a gamma-ray of 0.780-Mev energy. The L line for this gamma-ray should come at 3906 gauss-cm, but it was not resolved and appears only as a bulge on the

[7] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. 8, 194 (1936).

high energy side of the L line of the 0.741-Mev gamma-ray.

Owing to the numerous weak lines obtained and the uncertainties involved in the chemistry of technetium, the decay of the various lines was followed for a considerable time. All lines appeared to decay with a period of 67 hours. In addition, in some of the earlier runs in which the chemistry was not well carried out, certain strong lines of technetium, produced by the (dn) reaction, were observed. In the present samples these lines were not observed and it is therefore felt that the sample is free of technetium. All lines apparently belong to Mo^{99}.

The relative intensities of the lines of energy greater than 0.300 Mev were calculated with the help of Gray's[8] empirical formula and are shown in Table II.

V. EXPERIMENTS WITH Tc^{99}

In order to investigate the spectrum of Tc^{99} the following procedure was carried out. The Mo^{99} was first separated from technetium. The Tc^{99} was allowed to come into equilibrium with the parent Mo^{99} and was then chemically separated and measured.

The "beta-ray" spectrum, shown in Fig. 5, is seen to consist of K and L internal conversion lines for a gamma-ray of 0.140 Mev. No evidence could be found for an internally converted gamma-ray of 0.181 Mev. In addition, in an incomplete experiment in which a stronger source was used, an Auger line was observed at 14 kev corresponding to the ejection of an L electron by the $K\alpha$ radiation of Tc.

The gamma-rays from Tc^{99} were measured using a lead radiator of 16 mg/cm^2. As will be seen in Fig. 6, only the K, L, and M photo-lines for the gamma-ray at 0.140 Mev are seen.

VI. DISCUSSION

The beta-ray spectrum of Mo^{99} consists of two beta-ray groups, the energies of whose end points are 1.23 $\pm$0.01 and 0.445$\pm$0.01 Mev. The ratio of the intensity of the high energy group to that lower energy is 4:1. There is some indication of a weaker group of low energy, estimated to have an end point of $\sim$0.08 Mev. The gamma-ray spectrum consists of lines at 0.140 Mev, owing to Tc^{99}, 0.181, 0.367, 0.741, and 0.780 Mev with some evidence for a 0.040-Mev line. The line at 0.741 Mev is about 10 times as strong as the lines at 0.367 and

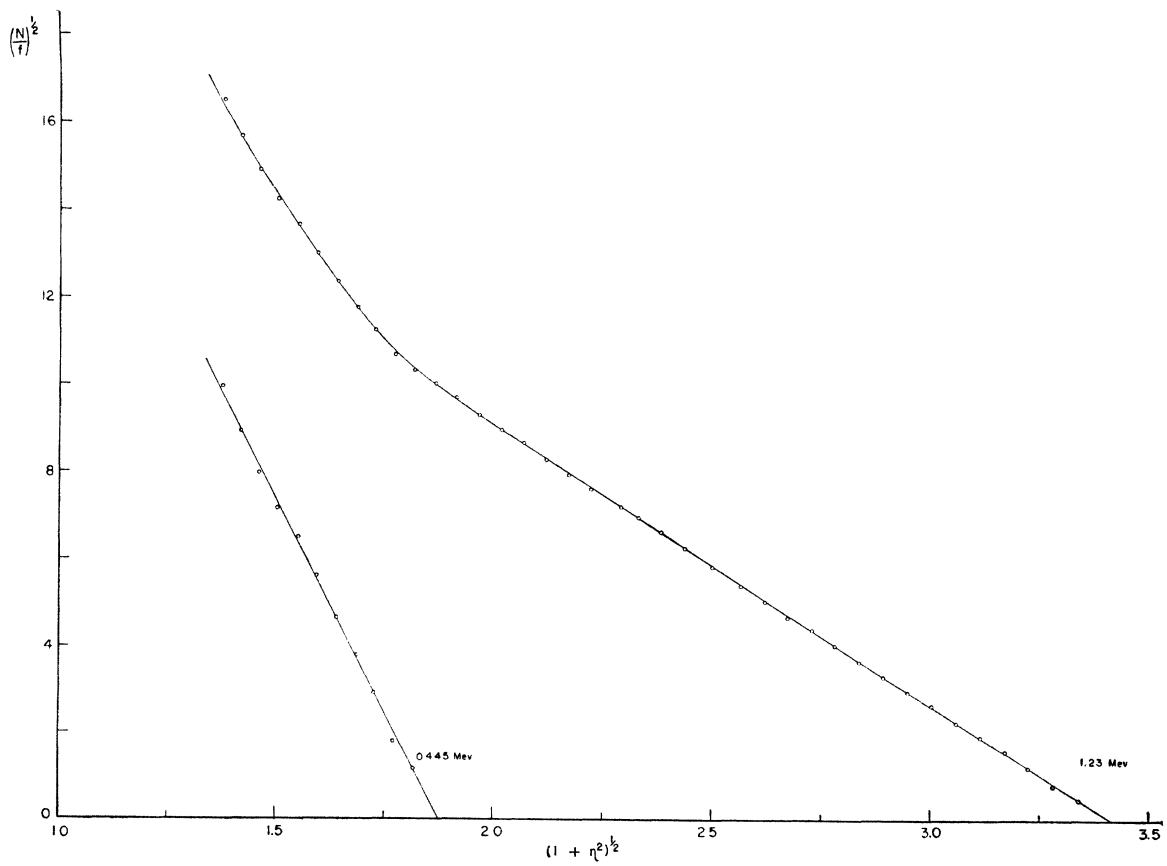

FIG. 2. Fermi plot of beta-ray spectrum of Mo^{99}.

[8] L. H. Gray, Proc. Camb. Phil. Soc. 27, 103 (1931).

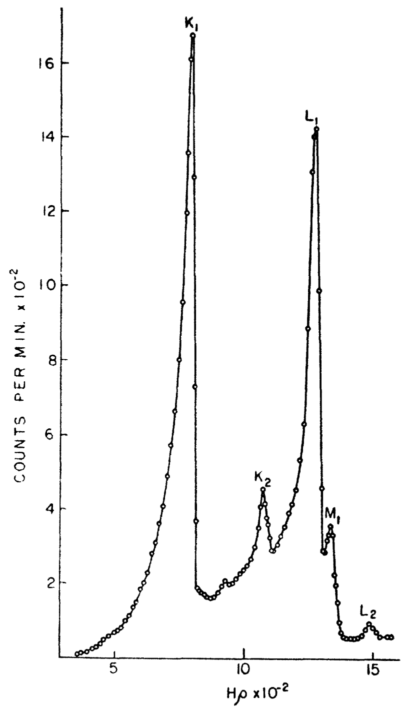

FIG. 3. The spectrum of the secondary electrons ejected from lead by the low energy gamma-rays of Mo^{99}.

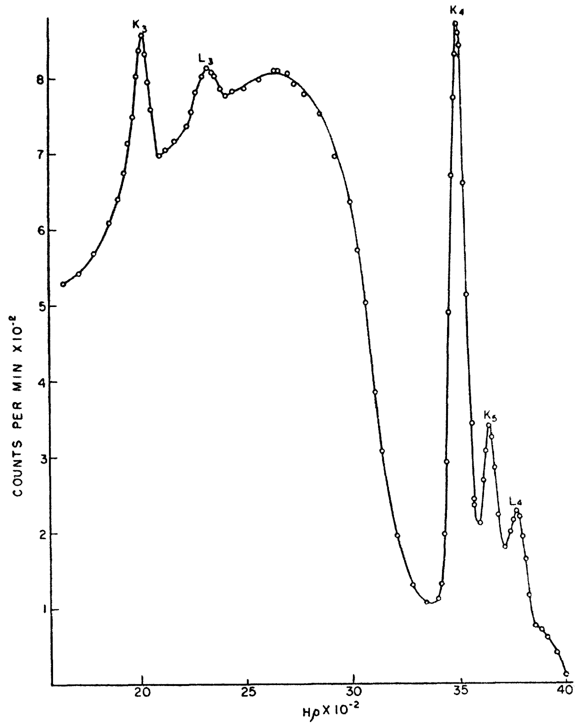

FIG. 4. Secondary electrons ejected from level by the high energy gamma-rays of Mo^{99}.

0.780 Mev. The K to L ratio for the internal conversion electrons from the 0.140-Mev line is approximately 9.

It is difficult to devise a disintegration scheme into which all of the data can be fitted. From a consideration of the energies of the various lines and beta-ray end points, the tentative scheme shown in Fig. 7 is suggested. Unfortunately, in the present experiments, it was not possible to obtain the correct relative intensity for the 0.040-Mev line owing to window cut-off.

If one assumes that the scheme given in Fig. 6 is correct, the internal conversion coefficient for the line at 0.140 Mev is approximately 0.20. According to Axel and Dancoff[9] it is necessary to assume an effective spin change $\Delta I=4$ to account for the half-life of the 6-hour metastable state of Tc^{99}. According to these authors the K to L ratio for the line at 0.140 Mev should be 1.8 for electric 2^4-pole radiation or 6.8 for magnetic 2^3 pole. The present observed value for the ratio of 9 would suggest a magnetic 2^3 pole character for the radiation. However, according to the tables of Rose, Goertzel, Spinrad, Harr, and Strong[10] the conversion ratio, β_3,

TABLE I. Internal conversion data.

Gamma-ray energy (Mev)	N_K/N_L
0.140	9 ± 2
0.181	5

TABLE II. Relative intensities of gamma-ray lines.

E (Mev)	Relative intensity
0.367	0.096
0.741	1.00
0.780	0.144

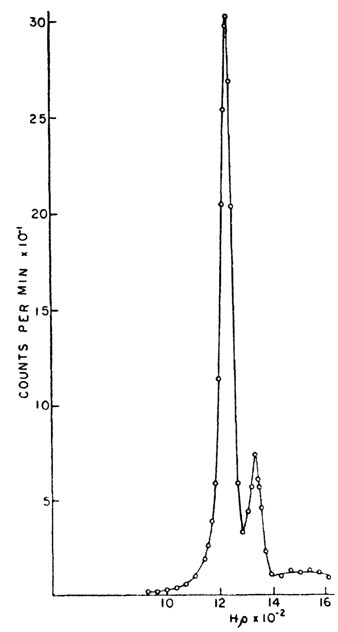

FIG. 5. Internal conversion electron spectrum of Tc^{99m}.

[9] P. Axel and S. M. Dancoff, Phys. Rev. **76**, 892 (1949).

[10] Rose, Goertzel, Spinrad, Harr, and Strong, report privately circulated.

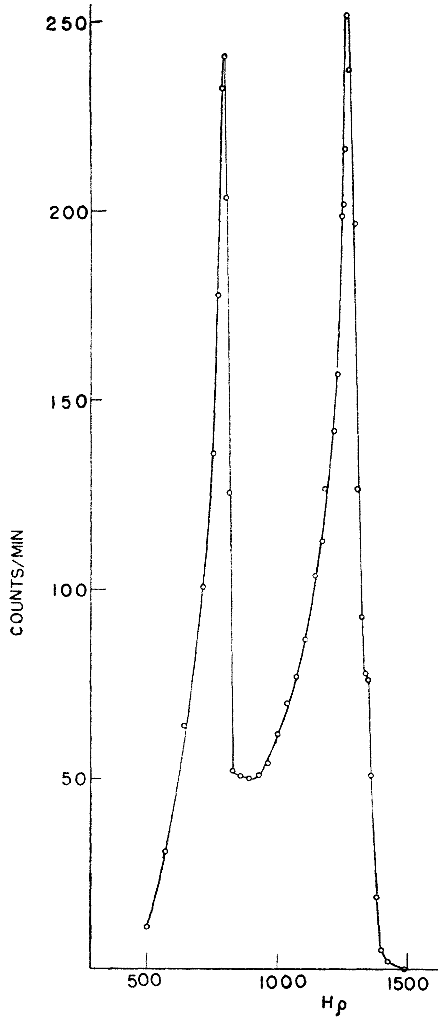

FIG. 6. Secondary electrons ejected from lead by gamma-ray of Tc^{99m}.

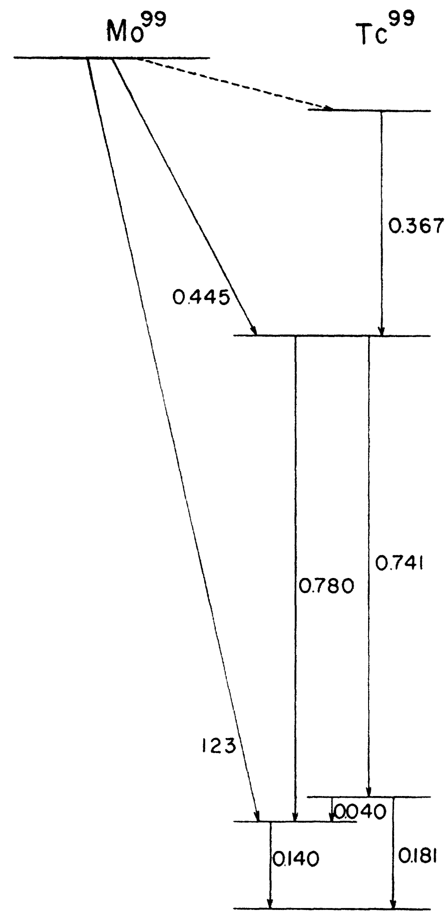

FIG. 7. Suggested energy level diagram for the decay of Mo^{99}.

will be approximately 4, in contrast to the observed value of 0.2. This difficulty appears to have been resolved by Medicus, Maeder, and Schneider,[3] who have evidence for two levels near 140 kev, differing in energy by 1.8 kev. One of these levels is the metastable level responsible for the 6-hour half-life. These two levels are not shown separately in Fig. 7.

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