Radiations from Molybdenum (99) and Technetium (99)

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The 67-hour activity was induced in Mo_2O_3 irradiated by slow neutrons in the Clinton pile. Mo^{99} emits two beta-ray spectra, one of low energy, having an end point at 0.057 g/cm² in aluminum, the other having a maximum energy of 1.03 Mev. The beta-rays of the low energy group are coupled with gamma-radiation, whereas those of the spectrum ending at 1.03 Mev are not. The gamma-rays of Mo^{99} have a maximum energy of 0.71 Mev as measured by coincidence absorption, and a small gamma-gamma coincidence rate, 0.085×10^{-3} coincidence per gamma-ray, was detected in Mo^{99}.

The electrons of the highly converted gamma-ray of the 6-hour metastable state of Tc^{99}, separated from Mo^{99}, are non-coincident with gamma-radiation. No gamma-gamma coincidences were observed in the technetium fraction. The conversion electrons have a range of 0.022 g/cm² in aluminum.

1. INTRODUCTION

IRRADIATION of molybdenum by slow neutrons gives rise to a 67-hour activity which has been assigned to molybdenum (99).[1] It has been shown[1,2] that part of the molybdenum disintegrations lead to a 6-hour metastable state of an isotope of element 43, $_{43}Tc^{99}$. A large percentage of the disintegrations go directly to the ground state of Tc^{99}, having a half-period of 4×10^6 years.[2]

For the experiments to be discussed in this paper, Mo^{99} was produced when Mo_2O_3 was irradiated by slow neutrons in the Clinton pile. Chemical separations were performed for the removal of calcium, columbium, iron, and phosphorous as possible impurities. Beta-ray energies were measured by noting the absorption in thin aluminum foils placed before a single G-M counter. Gamma-ray energies were measured by coincidence absorption of the secondary electrons. Two G-M counters in coincidence were employed for the determination of the beta-gamma and gamma-gamma coincidence rates of Mo^{99} and Tc^{99}. The accidental coincidences were calculated by the expression

$$A = N_1N_2(K\tau),$$

where $K\tau$ was 1.3 microseconds. A more detailed description of the general experimental procedure has been previously given.[3]

Mo^{99}

The hard beta-rays of Mo^{99} have been reported to have energies of 1.5 Mev[1] and 1.2 Mev.[2] A number of gamma-ray energies have been reported. Schwarz and Pool[4] have obtained evidence for the presence of gamma-rays having energies of 0.770, 0.815, and 0.840 Mev from an analysis of photoelectric lines in a semicircular focusing spectrometer. Quantum energies of 0.24 and 0.75 Mev were reported by Miller and Curtiss[5] who used a thin magnetic lens spectrometer.

The absorption curve of the primary beta-rays of Mo^{99} is shown in Fig. 1. The absorption limit is seen to occur at 0.40 g/cm², 1.03 Mev as calculated by Feather's equation.[6] Coincidence absorption of the secondary electrons of the gamma-rays of Mo^{99} yielded a maximum quantum energy of 0.71 Mev as calculated from the end point of Fig. 2. The above-quoted values of beta-ray and gamma-ray energies were present in Mo^{99} whether in equilibrium with its daughter isotope of technetium or chemically separated from it.[7]

A thin source of Mo^{99} in equilibrium with the 6-hour metastable state of Tc^{99} was placed between two G-M counters in coincidence, and the beta-gamma coincidence rate was observed

* Assisted by the Office of Naval Research.
[1] G. T. Seaborg and E. Segrè, Phys. Rev. **55**, 808 (1939).
[2] Plutonium Project Report, "Nuclei formed in fission," Rev. Mod. Phys. **18**, 513 (1946).
[3] C. E. Mandeville and M. V. Scherb, Phys. Rev. **73**, 141 (1948).
[4] W. M. Schwarz and M. L. Pool, Phys. Rev. **71**, 122 (1947).
[5] L. C. Miller and L. F. Curtiss, Phys. Rev. **70**, 983 (1946).
[6] N. Feather, Proc. Camb. Phil. Soc. **34**, 599 (1938).
[7] C. Perrier and E. Segrè, J. Chem. Phys. **5**, 712 (1937); *ibid.* **7**, 155 (1939).

as a function of the thickness of aluminum absorber inserted before the beta-ray counter. These data are given in Fig. 3(a) where the genuine beta-gamma coincidence rate is seen to decrease from 0.07×10^{-3} coincidence per beta-ray at zero absorber thickness to zero at an absorber thickness of 0.057 g/cm^2. This would indicate the presence of a group of beta-rays of low energy (about 240 kev) which is coupled with gamma-rays. It follows also that the hard beta-rays, reported in this paper as having an end point at 1.03 Mev, are non-coincident with gamma-radiation. The beta-transition of high energy then leads to the ground state of one or both of the technetium isomers. The beta-gamma coincidence rate for Mo^{99} chemically separated from its daughter isotopes of Tc^{99} is given in Fig. 3(b), where it is seen to be essentially the same as prior to separation.

The gamma-gamma coincidence rate for the molybdenum fraction was found to be 0.085 $\times10^{-3}$ coincidence per gamma-ray, suggesting cascade emission of gamma-rays following the beta-ray group of low energy.

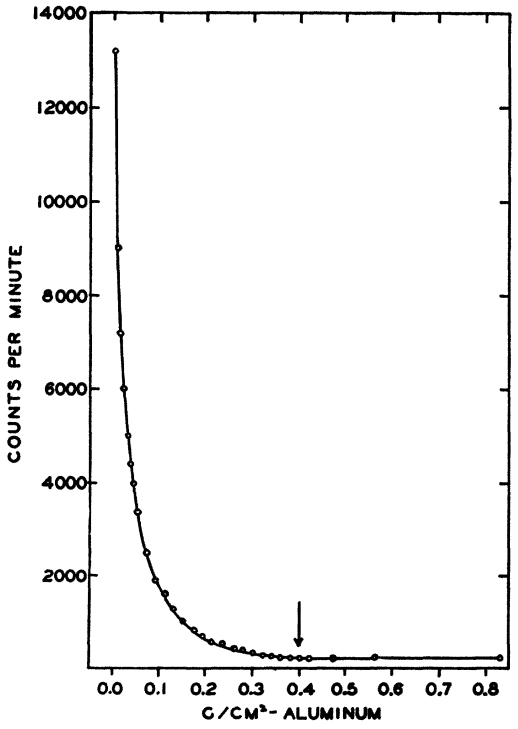

FIG. 1. Absorption in aluminum of the beta-rays of Mo^{99}.

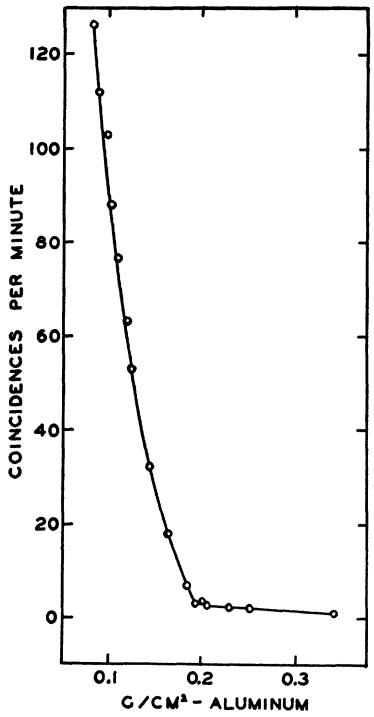

FIG. 2. Coincidence absorption of the secondary electrons of the gamma-rays of Mo^{99}. The quantum energy calculated from the end point is 0.71 Mev.

Tc^{99}

As has already been pointed out, Mo^{99} has been shown to be in equilibrium with a 6-hour daughter element,[1,2] an isomer of Tc^{99}. The 6-hour activity decays with the emission of gamma-rays and conversion electrons.[1,2] The energy of the converted gamma-ray has been measured to be 0.129 Mev.[8] Some evidence has also been obtained for the presence of gamma-rays of energy 0.18 Mev which were thought to be in cascade with the gamma-rays of the 6-hour isomeric transition.[1] No coincidences were found in the present experiment between the conversion electrons and the gamma-radiation nor were gamma-gamma coincidences observed. These results would indicate that the 6-hour Tc^{99} de-excites with emission of a single partially converted gamma-ray of energy 0.129 Mev. It should be remarked that several hundred coincidences were recorded during each of a number of different runs in the case of both beta-gamma

[8] D. C. Kalbfell, Phys. Rev. **54**, 543 (1938).

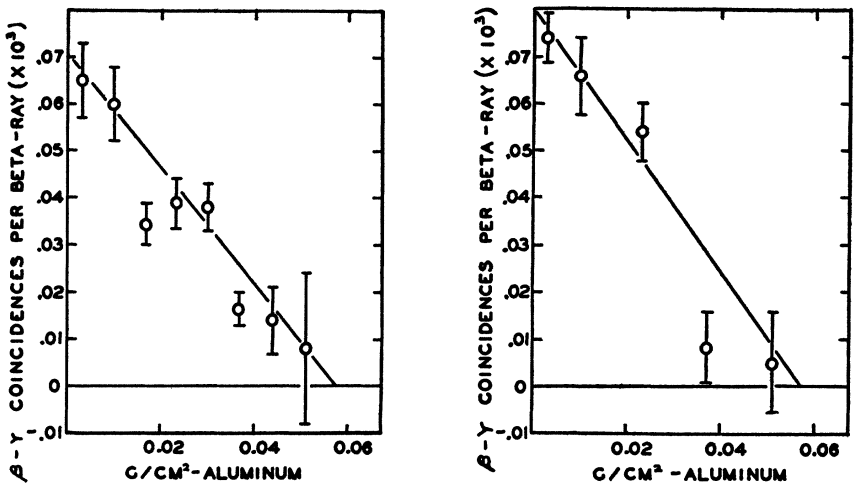

FIG. 3. Beta-gamma coincidence rate of Mo^{99} as a function of the surface density of aluminum absorber placed before the beta-ray counter. The data of 3(a) were taken before the technetium isomers were separated from their molybdenum parent, those of 3(b) immediately after the chemical separation. From the curves, it is clear that the presence of technetium had little effect on the coincidence rate.

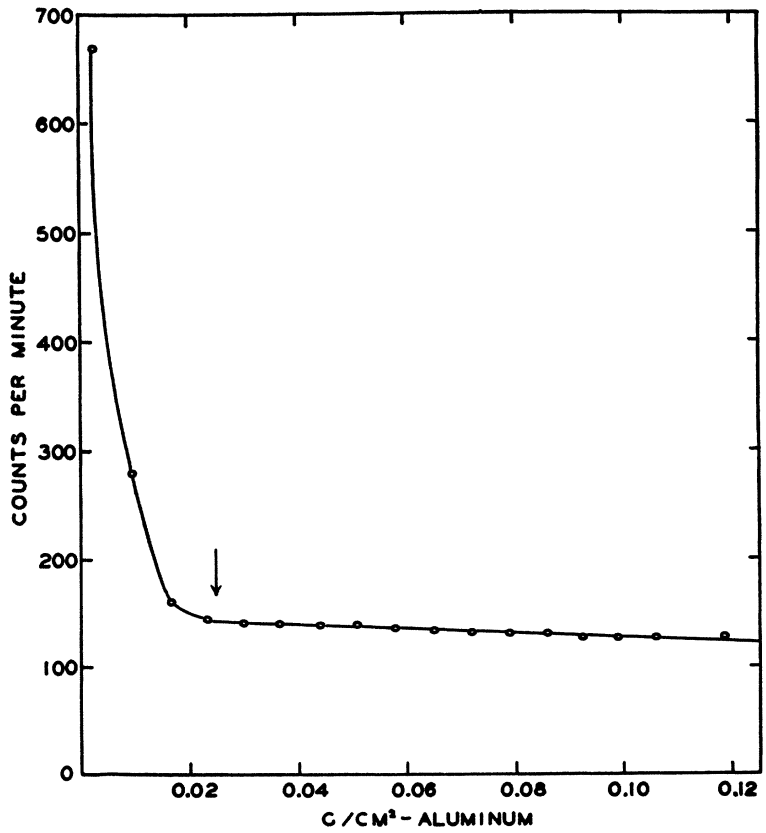

FIG. 4. Absorption in aluminum of the electrons of the converted gamma-ray of the 6-hour isomer of Tc^{99}.

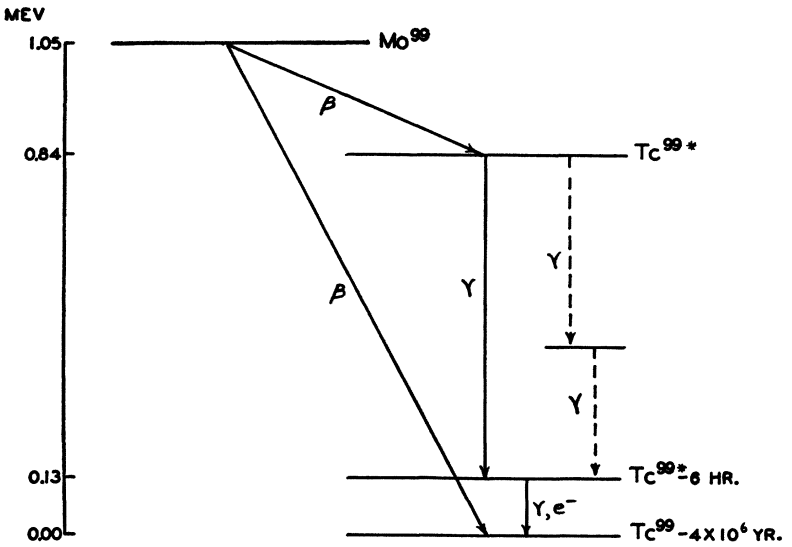

FIG. 5. Disintegration scheme for Mo^{99} and Tc^{99}. Owing to the uncertainties of absorption methods, it was not possible to construct a level diagram using *precisely* the same energy values as are given in the text.

and gamma-gamma coincidences. No† coincidences in excess of the accidental rates were observed. Coincidence experiments on elements having known disintegration schemes have indicated a net efficiency of about 0.13×10^{-3} for a quantum energy of 0.18 Mev. An efficiency of this magnitude would have given as many genuine coincidences as accidentals. The technetium fraction was found to emit no gamma-rays of sufficient energy to measure with the coincidence absorption arrangement. The range of the conversion electrons was measured in aluminum and was found to be 0.022 g/cm². These data are given in Fig. 4.

2. A DISINTEGRATION SCHEME FOR Mo^{99} AND Tc^{99}

The experimental data of this paper are summarized in the level diagram of Fig. 5. The hard beta-rays are shown to lead to the ground state of the long lived isomer of Tc^{99}, and the soft beta-rays, coupled with gamma-radiation, are assumed to lead to the 6-hour isomer. This seems reasonable, since it has been shown that only about ten percent of the molybdenum disintegrations go to the 6-hour metastable state.[2] These experiments would not reveal whether some of the hard beta-rays also go to the 6-hour level. A Fermi plot of spectrometric data might disclose the presence of two high energy spectra having end points differing by 0.129 Mev. It should be pointed out that since the measurements of this paper are subject to all of the inaccuracies of the absorption method, the hard beta-rays may actually have enough energy to permit cascade emission of the two gamma rays reported by Miller and Curtiss.[5]

ACKNOWLEDGMENTS

The writers wish to express their appreciation to Professor W. B. Keighton of Swarthmore College for having carried out the chemical separations.

Note added in proof: A second quantity of Mo_2O_3 has been irradiated by neutrons in the pile. The coincidence measurements of this paper have been repeated with coincidence resolving times of one microsecond and 0.10 microsecond with essentially the same results as reported above. In investigating the coincidence rates of the technetium fraction with the shorter resolving time, the single counting rates in either counter were adjusted so that a genuine (conversion electron)-gamma coincidence rate of 0.02×10^{-3} coincidence per conversion electron would have given four times as many genuine coincidences as accidental coincidences. A gamma-gamma coincidence rate of similar magnitude would have given about twice as many genuine coincidences as accidentals. In neither case were any genuine coincidences observed.

These new measurements would confirm the view already expressed in the text of the paper that if the 0.18 Mev

† The actual values of the coincidence rates were $(0.02\pm0.015)\times10^{-3}$ (conversion electron)-gamma-coincidence per conversion electron and $(-0.05\pm0.05)\times10^{-3}$ gamma-gamma coincidence per gamma-ray. It is possible that a small (conversion electron)-(x-ray) coincidence rate was detected, as the first coincidence rate above may indicate. X-rays would be emitted on readjustment of the extra-nuclear structure following internal conversion. Very few (conversion electron)-(x-ray) coincidences would be counted owing to the low detection efficiency of the gamma-ray counter for x-rays.

gamma-ray reported by Seaborg and Segrè is present at all, it is not in cascade with the 0.129 Mev converted gamma-ray of the 6-hour metastable state in Tc^{99}.

APPENDIX

Chemical Purification of Molybdenum

To about two grams of Mo_2O_3 were added 40 mg of Al_2O_3, 20 mg of CaO, and 20 mg of Ta_2O_5 as carriers for iron, calcium, and columbium impurities. The mixture was fused with Na_2CO_3, and the melt was leached with cold water. After filtering out calcium carbonate, 20 mg of Na_3PO_4 were added as a carrier for phosphorous, which was then precipitated from ammoniacal solution as ammonium magnesium phosphate. Most of the aluminum and tantalum should have accompanied this precipitate together with any iron and columbium impurities which might be present. After filtration, the filtrate was acidified with HCl, and most of the molybdenum was precipitated with H_2S as MoS_2. The precipitate was washed with dilute H_2SO_4 (1:99); washed with alcohol, and ignited to MoO_3.

For the separation of Mo from Tc, a second sample of Mo_2O_3 was purified as described above. The filtrate from the magnesium ammonium phosphate precipitation was neutralized and buffered with sodium acetate. Molybdenum was precipitated by 8-hydroxyquinoline. From the filtrate, about 0.5 N in HCl, technetium was precipitated as a sulfide. For details of the molybdenum-technetium separation, see reference 7.

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The Penetration of Gamma-Radiation through Thick Layers

I. Plane Geometry, Klein-Nishina Scattering

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A rough method is developed for estimating the intensity of gamma-rays which have passed through thick targets. Only Klein-Nishina scattering is considered. The treatment is exact both for those gamma-rays which are unscattered and those which are scattered once. The accuracy of the results for rays which are multiple scattered depends upon the fact that most of the scattering takes place in the forward direction with only small angles of deviation. Numerical calculations are made for energies of 2, 6, and $10m_0c^2$ and all thicknesses of target. If A is the intensity of the initial beam, and I is the intensity after passing through a target of D mean-free-paths measured in terms of the initial unscattered gamma ray, it is found that:

initial energy $=2m_0c^2$,
$$I/A=(1+0.487D+0.030D^2)e^{-D},$$
initial energy $=6m_0c^2$,
$$I/A=(1+0.400D+0.0080D^2)e^{-D},$$
initial energy $=10m_0c^2$,
$$I/A=(1+0.33D+0.0040D^2)e^{-D}.$$

The contributions to the intensity are given for the gamma-rays which have been scattered a various number of times. Extensive tables are given which are generally useful for any problems in which only Klein-Nishina scattering is assumed.

I. INTRODUCTION[4]

THE general problem of multiple scattering is exceedingly difficult. However, in some special cases it is possible to develop a rough theory which gives an accurate description of the gamma-radiation that has passed through a thick target. The method presented here is applicable to a wide homogeneous monochromatic beam of gamma-rays impinging at right angles on a flat slab of material. In subsequent papers the photoelectric effect and pair production will be considered, but only Klein-Nishina scattering is considered here. This limits the energy range in which the numerical calculations are valid to a comparatively small range which

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[4] This paper is a thorough revision of and substitution for LADC-70. The method and numerical results have been changed considerably.