

what higher energy, we find a second cluster of levels of seniority three. This behavior is almost independent of the relative sizes of the pairing force and the separation between the single-particle levels. We would expect states of the same spin to interact. In the simplest approximation of this interaction, we might expect its effect to be proportional to the available number of states of a given spin. On this basis we would expect $\frac{3}{2}+$ and $\frac{5}{2}+$ levels to be low, but not lower than the $\frac{5}{2}+$ and $\frac{7}{2}+$ levels. Except for the conspicuous absence of a $\frac{7}{2}+$ level, the low-lying levels of Pd^{103} are in accord with such a description. Parenthetically we remark that the $N=57$ isotope Ru^{101} does exhibit a low-lying $\frac{7}{2}+$ state. Quantitative calculations are in progress.

It is a striking fact that for neutron numbers $53 \leq N \leq 59$ the first excited state is $\frac{3}{2}+$ and occurs at an energy of 100–300 keV whenever the ground state is $\frac{5}{2}+$. Six cases are now known: Zr^{93} and Mo^{95} ($N=53$); Ru^{99} ($N=55$); Ru^{101} and Pd^{103} ($N=57$); Pd^{105} ($N=59$). A calculation similar to that for the $N=57$ isotones would also yield a low-lying $\frac{3}{2}+$ state throughout the region $53 \leq N \leq 59$.

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New Isomer: 5.3-h $\text{Mo}^{103\ddagger}$

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Fission of U^{238} and Th^{232} induced by 14.8-MeV neutrons was found to produce a new radioactive nuclide which was assigned to Mo^{103} isomeric state. The following radiation characteristics have been observed to belong to the decay of Mo^{103} isomer: half-life 5.3 ± 0.2 h; gamma-ray energies 0.54 ± 0.03 , 1.13 ± 0.06 , 1.26 ± 0.06 , 1.46 ± 0.10 , and 1.70 ± 0.10 MeV; and relative intensities 100, 28.9 ± 2.0 , 42.9 ± 4.0 , 13.6 ± 2.0 , and 36.8 ± 4.0 , respectively. The production ratio of this new nuclide to Mo^{99} in the 14.8-MeV neutron-induced fission of U^{238} and Th^{232} was found to be 0.010 ± 0.002 and 0.015 ± 0.002 , respectively.

I. INTRODUCTION

IN connection with a systematic study on the production ratios of various isomers resulting from 14.8-MeV neutron-induced fission of heavy elements, we found a new 5.3-h activity associated with molybdenum. All the experimental evidences secured so far lead to the conclusion that this new nuclide has to be assigned to an isomeric state of Mo^{103} . Kienle *et al.*,¹ in 1963, reported the existence of a 70-sec Mo^{103} . According to Mayer and Jensen,² however, the levels $h_{11/2}$, $d_{3/2}$, and $s_{1/2}$ may lie relatively close in energy, and the possibility of the occurrence of isomers seems to exist just before the levels $d_{5/2}$ and $g_{7/2}$ are completely filled at $N=64$. Nevertheless, the occurrence of an isomer with a half-life as long as 5 h in this region was quite unexpected, and we present in this report the decay characteristics, genetic relationships, as well as the basis for the assignment of this nuclide to Mo^{103} .

II. EXPERIMENTAL

The U^{238} used in this work was supplied by Dr. H. M. Roth of the Research and Development Division, U. S. Atomic Energy Commission, Oak Ridge, Tennessee. The isotopic composition of the uranium was 99.989% U^{238} and 0.011% U^{235} . The Th^{232} used was a reagent-grade $\text{Th}(\text{NO}_3)_4 \cdot 4\text{H}_2\text{O}$ (Baker's analyzed). The U^{238}O_2 ($\text{NO}_3)_2 \cdot 5\text{H}_2\text{O}$ and $\text{Th}(\text{NO}_3)_4 \cdot 4\text{H}_2\text{O}$ was irradiated with 14.8-MeV neutrons produced by the $\text{T}(d,n)\text{He}^4$ reaction in the University of Arkansas 400-kV Cockcroft-Walton positive ion accelerator.³

After irradiation, the length of which was varied from 15 min up to 1h, the sample was dissolved in 6M hydrochloric acid containing appropriate amounts of inactive carriers of Mo(VI), Te(IV), Te(VI), and Fe(III). Molybdenum was separated from other fission products using standard radiochemical procedures.^{4–6}

³ W. L. Bronner, K. E. Ehlers, W. W. Eukel, H. S. Gordon, R. C. Marker, F. Woelker, and R. W. Fink, *Nucleonics* **17**, 94 (1959).

⁴ J. Kleinberg, Report No. LA-1721 (1954).

⁵ C. D. Coryell and N. Sugarman, in *Radiochemical Studies: The Fission Products* edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, New York, 1951); National Nuclear Energy Series, Division 4, Vol. 9.

⁶ Nucl. Sci. Ser. NAS-NS 3009 (National Academy of Sciences-National Research Council, Washington 25, D. C.).

[†] Work supported by U. S. Atomic Energy Commission.

¹ Von P. Kienle, F. Baumgartner, B. Weckermann, and U. Zahn, *Radiochim. Acta* **1**, 84, (1963).

² M. G. Mayer and J. H. D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley & Sons, Inc., New York, 1955), p. 214.

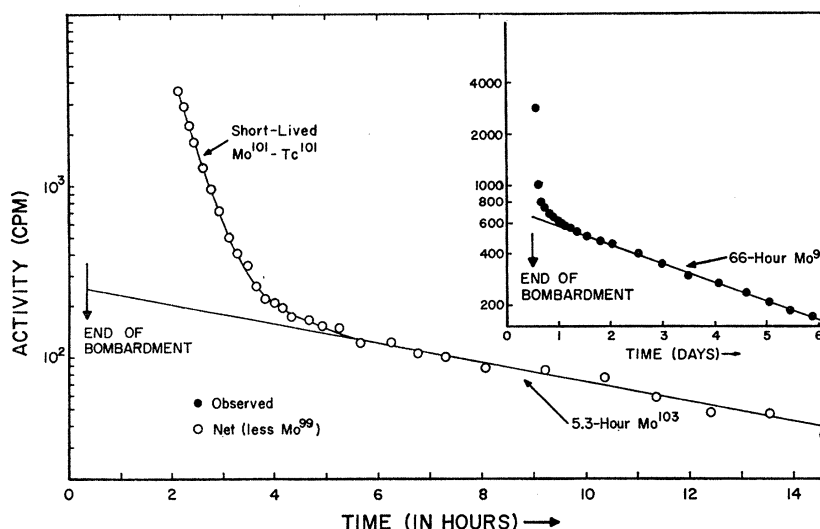


FIG. 1. Gross beta-decay curve of molybdenum isotopes separated from U^{238} irradiated with 14.8-MeV neutrons.

Briefly, the procedure used is as follows: Molybdenum was separated from the fission mixture by extraction into diethyl ether from 6M hydrochloric acid. After back extraction into water containing a few drops of NH_4OH , several $\text{Fe}(\text{OH})_3$ scavengings were carried out. The resultant supernate was then acidified to methyl red end point with dilute HNO_3 acid, and rhenium carrier was added to act as hold back carrier for technetium. Finally molybdenum was precipitated as PbMoO_4 .

Beta counting was carried out in a 2π end-window methane flow proportional counter. All the beta countings were performed through a 15 mg/cm² aluminum foil to eliminate the 6.0-h Tc^{99m} conversion electrons. Gamma ray spectra were taken using a 3×3-in. NaI(Tl) crystal.

III. RESULTS AND DISCUSSION

A typical gross beta decay of the molybdenum fraction radiochemically separated from uranium shortly after the end of irradiation is shown in Fig. 1. Between the short-lived component of $\text{Mo}^{101}\text{-Tc}^{101}$ and 66-h decay of Mo^{99} , the existence of a 5.3-h molybdenum activity is clearly seen in Fig. 1. Chemical separations for molybdenum were performed on irradiated uranium samples immediately after the end of irradiation and also 3 h after the end of irradiation. The ratio of the 5.3-h activity to the 66-h activity remained unchanged in these experiments, indicating that the 5.3-h activity is due to molybdenum, and not technetium.

Figure 2 shows the tail part of the gross beta decay of the molybdenum fraction separated from irradiated uranium. After about 70 days, the decay of Mo^{99} was complete and a "tail" of approximately 4 counts per minute (cpm) was observed. The ratio of the 5.3-h activity present in the sample to the residual activity was found to be in agreement with the half-life ratio

of 40-day Ru^{103} and 5.3-h molybdenum when corrected for counting efficiencies.

Typical gamma-ray spectra of the molybdenum fraction separated from the irradiated uranium are shown in Fig. 3. The peaks at 0.370, 0.740, and 0.920 MeV were due to 66-h Mo^{99} . All the other peaks showed the half-lives between 5.0 and 5.5 h; their energies and intensities are listed in Table I. The gamma spectrum taken about 70 days later, after the 66-h Mo^{99} had decayed away completely, showed the presence of 0.498- and 0.615-MeV peaks of 40-day Ru^{103} .

In order to eliminate the further possibility that the 5.3-h activity was due to the contamination, the following experiment was carried out. After the irradiation

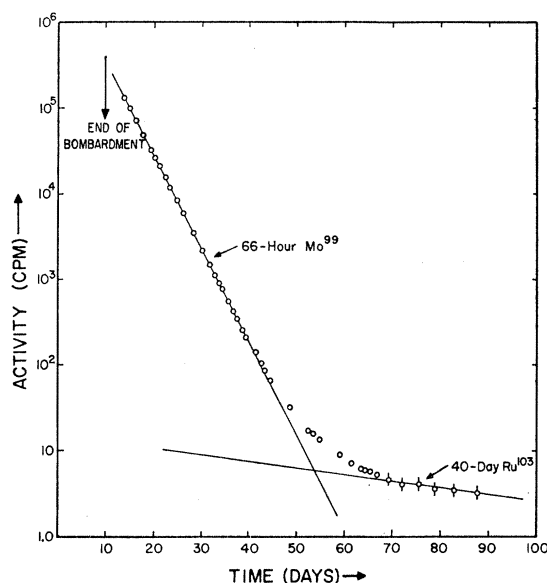


FIG. 2. Tail part of the beta-decay curve of the molybdenum fraction resulting from the fission of U^{238} by 14.8-MeV neutrons.

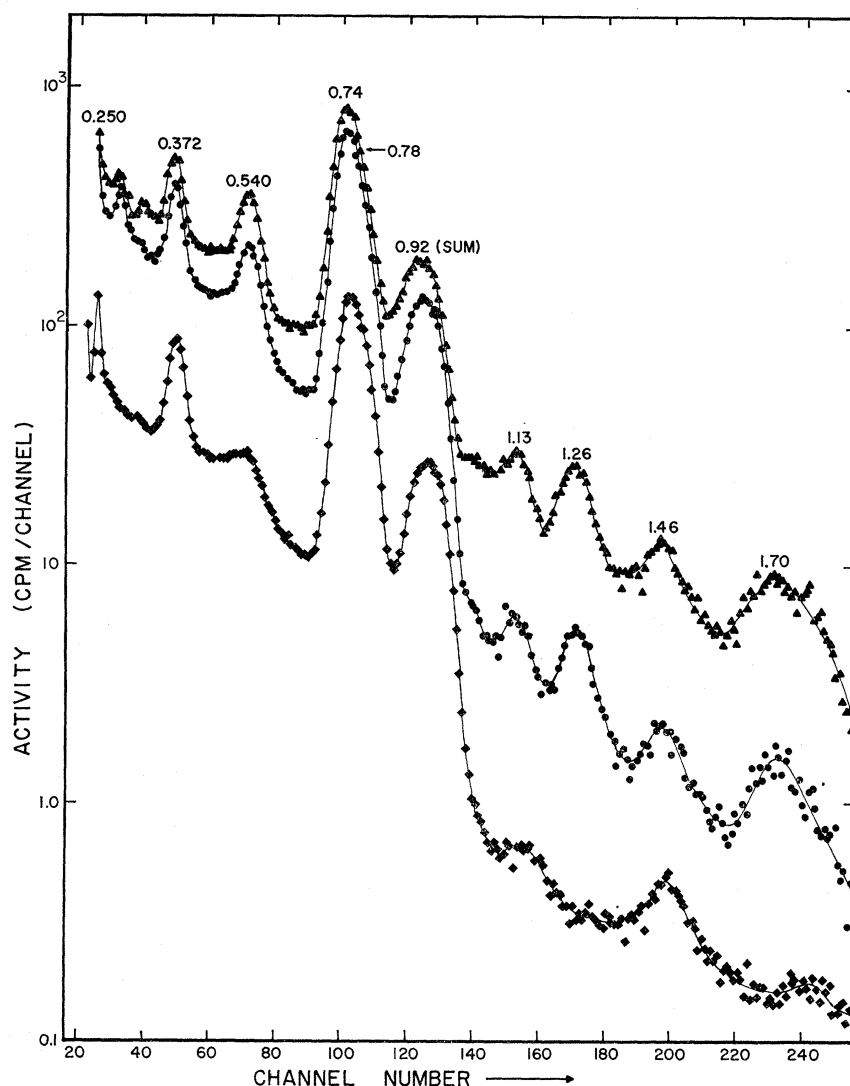


FIG. 3. Gamma spectra of the molybdenum isotopes from the 14.8-MeV neutron fission of U^{238} . Δ 5 h after the end of bombardment; $\bullet$ 19 h after the end of bombardment; $\blacklozenge$ 124 h after the end of bombardment.

the uranium sample was dissolved in 6M hydrochloric acid, containing appropriate amounts of inactive carriers of Mo(VI), Te(IV), Fe(III), and Te(VI). Molybdenum was extracted into diethyl ether and then back extracted into water containing a few drops of NH_4OH . After several $Fe(OH)_3$ scavengings the resultant supernate was divided into two fractions, from one of which molybdenum was precipitated as $PbMoO_4$ as described above. Then from the other fraction

molybdenum was precipitated with alpha-benzoin oxime. The precipitate was dissolved in a small amount of fuming nitric acid, and alpha-benzoin oxime precipitations were repeated three times. The final precipitate was dissolved in fuming nitric acid and treated with hot concentrated perchloric acid. After one more ferric hydroxide scavenging molybdenum was precipitated as lead molybdate.

The gamma spectra of these samples about 5 h after the end of irradiation showed identical gamma-ray peaks. The ratios of 5.3-h gamma-ray peaks to the 0.74-MeV peak of 66-h Mo^{99} were found to be constant in both samples. These results confirmed that the 5.3-h activity was due to the molybdenum isotope, and not due to the contamination.

The 5.3-h activity was also observed when thorium, instead of uranium, was irradiated with 14.8-MeV neutrons. In all measurements performed using varying

TABLE I. Gamma rays from Mo^{108} isomer.

Gamma ray energy (MeV)	Relative Intensity
0.54 ± 0.03	100
1.13 ± 0.06	28.9 ± 2.0
1.26 ± 0.06	42.9 ± 4.0
1.46 ± 0.10	13.6 ± 2.0
1.70 ± 0.10	36.8 ± 4.0

quantities of uranium and thorium, and changing irradiation times, the production ratios of the 5.3-h activity to 66-h Mo⁹⁹ remained constant for uranium and thorium as shown in Table II.

Additional proofs that the 5.3-h activity could not be assigned to an isomeric state of Mo¹⁰¹ were obtained in the experiment described below. Natural ruthenium chloride was irradiated with 14.8-MeV neutrons, and the molybdenum fraction was separated radiochemically. The molybdenum fraction thus separated did not reveal any sign of 5.3-h activity. In another experiment, a sample of enriched Ru¹⁰⁴ (Ru⁹⁶, 0.24%; Ru⁹⁸, 0.11%; Ru⁹⁹, 0.7%; Ru¹⁰⁰, 0.85%; Ru¹⁰¹, 1.15%; Ru¹⁰², 6.97%; Ru¹⁰⁴, 89.9%) irradiated with 14.8-MeV neutrons did not show any sign of 5.3-h decay of the 0.54-MeV peak in the gamma-ray spectrum.

A milking experiment was carried out in which Ru carrier was added to the molybdenum fraction containing the 5.3-h activity. The Ru was distilled after 48 h from the end of bombardment using perchloric acid. The distillate was collected in 6 M NaOH solution. Ruthenium was precipitated as the hydrous oxide and then reduced to metal. The Ru sample was counted in Tracerlab low-background beta counting equipment (CE-14) which had a background of 1.1 cpm in anti-coincidence with surrounding cosmic-ray counters. Beta-absorption curve using aluminum absorbers resembled the absorption curve of 40-day Ru¹⁰³. Gamma spectrum of the ruthenium sample revealed 0.498- and 0.615-MeV peaks of 40-day Ru¹⁰³. Half-life measurements confirmed the presence of 40-day Ru¹⁰³ in the milked sample.

With the most probable charge Z_p of 41.6 for mass chain 103, following Pappas,⁷ high independent and cumulative yields were expected for Mo¹⁰³.

The 5.3-h activity, however, corresponds to fission yields of 0.03% for Th²³², and 0.056% for U²³⁸. Thus the independent yield of 5.3-h Mo¹⁰³ is very much less than that of 70-sec Mo¹⁰³.

An attempt was made to produce the 5.3-h molybdenum activity in thermal neutron-induced fission of uranium. 1 mg of natural uranium was irradiated at the thimble of CP-5 reactor of Argonne National Laboratory for 30 min at a flux of 6×10^{13} neutrons/cm² sec. A

TABLE II. Production ratio of 5.3-h Mo¹⁰³ to 66-h Mo⁹⁹ in the 14.8-MeV neutron-induced fission of U²³⁸ and Th²³².

Target	Quantity of target material (g)	Length of irradiation (min)	N^{99}/N^{103}
Th ²³²	2	15	66±5
Th ²³²	0.5	15	64±5
Th ²³²	10	30	67±5
Th ²³²	20	15	66±5
U ²³⁸	5	15	100±15
U ²³⁸	25	60	108±15
U ²³⁸	0.2	15	95±15

quick radiochemical separation for molybdenum carried out on this sample did not reveal any 5.3-h decay in the beta counting or any gamma peaks other than 0.184, 0.372, 0.74, and 0.91 MeV, which belong to Mo⁹⁹-Tc^{99m}. Thus it appears that the production ratio of the 5.3-h isomer is very small for thermal neutron-induced fission of uranium.

On the other hand, 21.5-MeV deuteron-induced fission of thorium and uranium, carried out using the beams from the Argonne cyclotron, produced the 5.3-h molybdenum activity, although the yield appeared to be somewhat lower than that in the case of 14.8-MeV neutron induced fission. More detailed studies of the production ratios of the 5.3-h activity will be reported in the near future.

Since neither the decay schemes of 70-sec Mo¹⁰³, nor the 50-sec daughter Tc¹⁰³ are established, it is difficult to construct a decay scheme for the 5.3-h Mo¹⁰³ without further investigations, especially without performing coincidence measurements.

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⁷ A. C. Pappas, MIT Technical Report No. 63, 1953 (unpublished).