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HALF-LIVES OF Tm^{167}, Tm^{168}, Tm^{170}, Lu^{171}, Lu^{172}, Lu^{173}, Lu^{174}, Lu^{174m}, Au^{195}, Au^{196}, AND Au^{196m}

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ABSTRACT

The following half-lives have been measured: Tm^{167}, 9.25 ± 0.1 d; Tm^{168}, 93 ± 0.5 d; Tm^{170}, 125 ± 2 d; Lu^{171}, 8.2 ± 0.1 d; Lu^{172}, 6.7 ± 0.1 d; Lu^{173}, 499 ± 5 d; Lu^{174} and Lu^{174m}, 140 ± 10 d and 1300 ± 150 d; Au^{195}, 185 ± 1 d; Au^{196}, 6.17 ± 0.05 d; Au^{196m}, 9.7 ± 0.1 h.

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In the course of our work over the past few years we have made fairly accurate measurements of the half-lives of a number of nuclides. The ones that differ from published values[1] are listed in Table I. Also included in Table I are the production method and the principal radiation counted for each nuclide.

EXPERIMENTAL

A. Chemical Purifications

With the exception of the Au^{196} and Au^{196m} samples produced with the Cockcroft-Walton, all samples were chemically purified. All chemical procedures had been tested to ensure adequate radiochemical purity. The rare earths were separated on Dowex-50 columns with lactate elution at controlled pH.

B. Irradiations

Alpha-particle bombardments were done in the Crocker Laboratory 60-in. cyclotron in Berkeley.

Thermal neutron irradiations were done in the Livermore Water Boiler Reactor. The 14-Mev neutrons were produced by the $H^3(d,n)He^4$ reaction in the Livermore Cockcroft-Walton accelerator.

Materials irradiated in bomb spectrum neutrons were placed in or near a thermonuclear bomb. The desired element was then chemically separated from bomb debris collected on aircraft filters. Multiple samples from each detonation were counted.

C. Counting

The roman numerals below correspond to the entries in column 5 of Table I. All samples were mounted on aluminum plates. If they were gamma-counted, enough Al or Be absorber was used to absorb any betas present. The crystals used in methods I through VII below were all NaI(Tl), either 1-1/2 or 1-3/4 inches in diameter. The dimension given in the description is the crystal thickness. The photomultiplier tubes and associated circuitry were more or less standard. The energies given are the limits, set by the discriminator circuits, to the gamma energy accepted. For example, in method I the crystal is 1-1/2 inches in diameter and 1/8 inch thick, and the discriminators are set to accept pulses from gammas in the energy range between 35 and 75 kev only.

I. 1/8-in. crystal, 35 to 75 kev.

II. 1/8-in. crystal, 50 to 95 kev.

III. 1/8-in. crystal, 150 to 250 kev.

IV. 1-in. crystal, 29.5 kev or greater.

V. 2-in. crystal, 150 to 300 kev.

VI. 2-in. crystal, 600 to 900 kev.

VII. Coincidence counter. Two 2-in. crystals back to back, each set for 294 to 414 kev.

VIII. Beta counter. A methane-filled (1 atm) proportional counter with 1-in.-diameter window. Window was Al-covered Mylar, ~1 mg/cm^2.

D. Calculations and Errors

The half-lives were determined by a least-squares analysis of the counting data. An IBM 650 computer was used for the calculations.

The errors quoted in Table I fall in two categories. For the cases where a reasonably large number of samples were counted (Tm^{168}, Lu^{174}, Au^{195}, and Au^{196}), the error given is the mean deviation of the individually measured half-lives from the average.

In the other cases, the error was estimated from the counting statistics, the difficulty of resolving the decay curves, and the number of half-lives over which the samples were counted.

E. Miscellaneous Details of Individual Nuclides

Tm^{167}. The alpha bombardment of Ho produced 7.7-h Tm^{166} and 93-d Tm^{168} as well as the 9.25-d Tm^{167}. Only one sample was prepared. Twenty-four days were allowed for the 7.7-h activity to decay, and the sample was counted for 1 year on both counters. The long counting time was to allow sufficient decay so that the 93-day tail could be accurately measured and subtracted. The 93-d activity accounted for only a few percent of the total at first count. The two counting methods gave good agreement on the half-life, 9.28 and 9.24 d.

Tm^{168}. Seven Tm^{168} samples were prepared by four different methods. Four samples were bomb-produced, one was prepared by Tm^{169} (a, an), one by Tm^{169} (n, 2n) with 14-Mev neutrons from the Cockcroft-Walton, and one by Tm^{169} (n, 2n) from a wide spectrum of neutrons from 24-Mev deuterons on Be.

Counting of the bomb-produced samples was started 600 days after irradiation and continued for 300 days. The average half-life of these samples was

93.0 days, with a mean deviation of 0.2 day. Counting of the other three samples was started 35 to 50 days after irradiation (to allow Tm^{167} to decay) and continued for 350 to 550 days. The average half-life for these three samples was 92.7 days, with a mean deviation of 1 day.

At the time the samples were counted, the only contaminant was 125-day Tm^{170}. This decays by β^-, with an 84-kev gamma associated with 24% of the events. A Be absorber stopped the betas and the discriminator settings eliminated counts from the 84-kev gammas. The counting method discriminated against Tm^{170} by a factor greater than 4400. No Tm^{170} activity could be detected in any of the decay curves.

Tm^{170}. The sample had no interfering activities, and the decay was followed for 3-1/2 years.

Lu^{171}. The alpha irradiation of Tm would be expected to produce 1.7-d Lu^{170} and 6.7-d Lu^{172} as well as Lu^{171}. The sample was allowed to decay for 5 weeks after irradiation, so the amount of 1.7-d isotope was negligible. A gamma pulse analysis showed no evidence for the presence of any of the Lu^{172} gammas. The sensitivity was such that if 1% of the total activity had been due to Lu^{172} it could have been detected. Apparently the (α, n) cross section is much smaller than the (α, 2n) cross section under the irradiation conditions used.

The sample was counted for 5 months on the two counters. The decay on the gamma counter showed a single half-life of 8.15 days. The x-ray counter gave a half-life of 8.23 d, but with a very small long-lived tail. The tail amounted to about 15 counts/min compared to the initial counting rate of 150,000 counts/min. This long-lived component could possibly be the 1.6-y isomer previously reported. However, considering the extremely small yield, it seems

more likely that it is 499-d Lu^{173} formed by (α, pxn) reactions on a small amount of Yb impurity in the Tm target. It thus appears that the 1.6-y isomer probably does not exist.

Lu^{172}. A target of Yb_2O_3 was irradiated with 46-Mev alphas to produce a mixture of Hf isotopes. After 3 months, when the 16-h Lu^{171} and the 24-h Lu^{173} had decayed, the Hf and Lu were separated. At this point the only Hf isotope decaying to Lu was the ~5-y Hf^{172}. The Lu^{172} was allowed to grow in for about 2 weeks; then it was separated from the Hf parent and its decay was followed for 3 months. The two counting methods gave the same half-life.

Lu^{173}. A sample of Hf^{173} was made by irradiating Hf metal with 24-Mev deuterons. Within a few hours after irradiation, rare earths were separated from the sample and discarded. Lu^{173} was allowed to grow into the purified Hf for 8 days and then separated from Hf, (and from Y^{88} which had grown in from Zr^{88} produced from Zr impurity in the target).

After the 6.7-d Lu^{172} (which had grown in from the 5-year Hf^{172}) had decayed, the sample decayed with a single half-life. It was followed for 500 days. The two counting methods gave half-life values of 501 and 498 d.

Lu^{174} and Lu^{174m}. Four bomb-produced Lu samples were used for the half-life determinations. The samples contained 6.8-d Lu^{177} and 499-d Lu^{173} as well as the Lu^{174} isomers. The first counts were taken 600 days after irradiation, so the Lu^{177} had all decayed. The decay was followed for 550 days. Lu^{173} was counted efficiently by the x-ray counter used, but its contribution to the counting rate could be measured independently and subtracted. This was done by counting the Lu^{173} gammas (Lu^{174} has no appreciable amount of gamma radiation), measuring the ratio of gamma counts to x-ray counts from a sample of pure Lu^{173}, and then calculating the contributions of Lu^{173} to the total counting rate of the Lu^{174} sample.

The decay curves after the subtraction of the 173 component still showed curvature. The samples were certainly radiochemically pure Lu, so either there was another isotope present, or the Lu^{174} decayed to a long-lived Hf^{174} or Yb^{174}, or Lu^{174} has two isomers. Lu^{174} made with 14-Mev neutrons showed the same curvature. Since this method produces essentially pure Lu^{174}, the long-lived material must have mass 174. Chemical separations on an old sample showed no activity in the Yb or Hf fractions, so the new half-life must be an isomer of Lu^{174}.

Because of the long half-lives involved and the errors introduced in subtracting the Lu^{173} component, the curve resolution is not very accurate. The half-lives obtained for the two isomers thus have relatively large errors.

We have no information as to which half-life is associated with the isomeric state and which with the ground state.

Au^{195}. Seven samples from bomb debris were counted for periods of 900 to 1400 days after the 6.17-d Au^{196} had decayed to less than 0.1% of the total activity remaining. The decay curves showed only a single component.

Au^{196m}. Three Au foils irradiated in the Cockcroft-Walton were counted intensively for 2 days, then followed for 24 days to determine the amount of 617-d Au^{196} to be subtracted. At first count the 9.17-h isomer was 1/3 to 1/2 of the total activity present.

Au^{196}. Measurements were made on two independent sets of samples. One set, made up of four Au foils, was irradiated with 14.1-Mev neutrons from the Cockcroft-Walton. These were wrapped in Cd foil during irradiation to reduce the amount of Au^{198} formed by neutron capture. The amount of Au^{198} formed in unwrapped Au foils was barely perceptible, and the Cd-wrapped ones used for half-life determinations had no appreciable amount.

Three of the four samples were counted by method IV, and the fourth was counted on a 3-in. × 1-1/2-in. NaI crystal with discriminators set to accept pulses from gammas with energies between 250 and 500 kev.

The samples were counted for periods ranging from 20 to 69 days. All decayed with a single half-life. The half-life values ranged from 6.15 to 6.18 d. The 3-in. crystal gave the 6.15-day value.

The second set was made up of 16 bomb-produced samples. These contained relatively large amounts of 2.7-d Au^{198}, so the counting methods used for the Cockcroft-Walton samples could not be used. Au^{196} has two coincident gammas of similar energy (331 and 354 kev). Counting method VII measures these gammas and discriminates strongly against the Au^{198} radiation. It discriminates completely against Au^{195} radiation.

The 16 samples were counted for periods ranging from 12 to 53 days (averaging about 40 days), starting about 1 week after irradiation. All samples decayed with a single half-life, the average value being 6.17 d. This value is the same, within experimental error, as the value obtained from the four Cockcroft-Walton samples.

REFERENCES

*Work performed under auspices of the U. S. Atomic Energy Commission.

[1]D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. 30, No. 2, Part II (1958).

Table I. Summary of data for Nuclides studied.

Nuclide	Half-life	Production method and source of particles	Principal radiation counted (ref. 1)	Counting method[a]
Tm^{167}	9.25 ± 0.1 d	Ho^{165} (α, 2n), 46-Mev alphas	x rays 208-kev gammas	I III
Tm^{168}	93 ± 0.5 d	Tm^{169} (n, 2n), bomb-spectrum neutrons	198-kev gammas	III
Tm^{170}	125 ± 2 d	Tm^{169} (n, γ), thermal neutrons	β^-	VIII
Lu^{171}	8.2 ± 0.1 d (no evidence for 1.6-y isomer)	Tm^{169} (α, 2n), 46-Mev alphas	x rays 735-kev gammas	I VI
Lu^{172}	6.7 ± 0.1 d	Hf^{172} decay	x rays Auger and conv e^-	I VIII
Lu^{173}	499 ± 5 d	Hf^{173} decay	x rays 172- and 273-kev gammas	I V
Lu^{174} and Lu^{174m}	140 ± 10 d 1300 ± 150 d	Lu^{175} (n, 2n), bomb-spectrum neutrons	x rays	I
Au^{195}	185 ± 1 d	Au^{196} (n, 2n), bomb-spectrum neutrons	x rays	II
Au^{196}	617 ± 0.05 d	Au^{197} (n, 2n), 14-Mev and bomb-spectrum neutrons	x rays 331- and 354-kev gammas	IV VII
Au^{196m}	9.7 ± 0.1 h	Au^{197} (n, 2n), 14-Mev neutrons	x rays and gammas	IV

[a] The counting methods are described in the experimental section.