Search for the decay of $^{180}Ta^m$

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A sample of Ta_2O_5 containing 151 mg of Ta enriched to 5.47% in $^{180}Ta^m$ was counted in a well-type high-purity Ge detector. This was enclosed in a large NaI(Tl) detector operated in anticoincidence to reduce the cosmic-ray continuum background. Several runs of ~100 days each were made under different conditions to search for γ-ray singles or summing peaks that would follow the electron capture decay of $^{180}Ta^m$ to ^{180}Hf or the β^- decay to ^{180}W. Lower limits derived were $T_{1/2} > 3.0\times10^{15}$ yr, $\log f_0 t > 22.9$ for electron capture decay and $T_{1/2} > 1.9\times10^{15}$ yr, $\log f_0 t > 21.3$ for β^- decay. The total half-life of $^{180}Ta^m$ is therefore $T_{1/2} > 1.2\times10^{15}$yr.

I. INTRODUCTION

The rare mass-180 isotope of tantalum, of natural abundance[1] 0.012(2)%, was discovered by White, Collins, and Rourke[2] in 1955. However, it has been shown only within the past 10 years that the naturally-occurring isotope is not the ground state but is, in fact, an isomer[3–5] at an excitation energy[6] of 73(2) keV and with a spin parity[4,7] of $J^\pi = 9^-$. The well-known 8.1-h activity[8] is actually the ground state of ^{180}Ta and has $J^\pi = 1^+$. It decays to the ground and first-excited states of ^{180}Hf and ^{180}W. Midway between the ground and isomeric states of ^{180}Ta is a 38(2)-keV level[6] with $J^\pi = 2^+$. Based on the known masses,[9] and an isomeric excitation energy of 73(2) keV, the decay energies for $^{180}Ta^m$ are $Q_{EC} = 926(4)$ keV and $Q_\beta = 781(5)$ keV. Level structures at low energies are well understood in both of the daughter nuclei ^{180}Hf and ^{180}W and, as may be seen from the level diagram in Fig. 1, the three main ways that $^{180}Ta^m$ could decay include EC or β^- nonunique third-forbidden decays to 6^+ states at 640.9 keV in ^{180}Hf or 688.3 keV in ^{180}W, or a γ-ray transition of energy 35(3) keV and multipolarity $E7$ which would be almost completely converted. Decay energies of $^{180}Ta^m$ to the 6^+ states are 285(4) keV for EC and 93(5) keV for β^- decay.

On the EC side, a cascade of three coincident γ rays of 332.3, 215.3, and 93.3 keV would be expected, while on the β^- side there would also be a triple γ cascade with energies of 350.4, 234.3, and 103.6 keV. All of the levels are short lived as far as γ-ray summing effects are concerned, and since the transitions are presumably all of $E2$ multipolarity, their conversion coefficients can be calculated. Table I lists the fractions of unconverted γ rays, in % per transition, for all six of the ^{180}Hf and ^{180}W γ rays.

Previous work on $^{180}Ta^m$ has consisted of searches with conventional Ge(Li) detectors for the various γ rays listed in Table I. In the most recent experiments by Norman,[10] a sample of Ta_2O_5 containing 30.5 mg of Ta enriched to 5.1% in $^{180}Ta^m$ was counted for 14 d with a well-shielded 50-cm^3 coaxial Ge(Li) detector. $\gamma\gamma$ coincidences were also measured but the results were not as sensitive as in singles due to the low coincidence efficiency and background effects. The limits derived were as follows: for EC decay $T_{1/2} > 5.6\times10^{13}$ yr, $\log f_0 t > 20.8$, and for β^- decay $T_{1/2} > 5.6\times10^{13}$ yr, $\log f_0 t > 20.1$.

Our aim was to achieve higher sensitivity in the search for $^{180}Ta^m$ decay by (1) increasing the γ-ray detection efficiency, (2) using five times the amount of source material as in the experiment by Norman, (3) reducing background with an anticoincidence counter, and (4) counting for much longer periods.

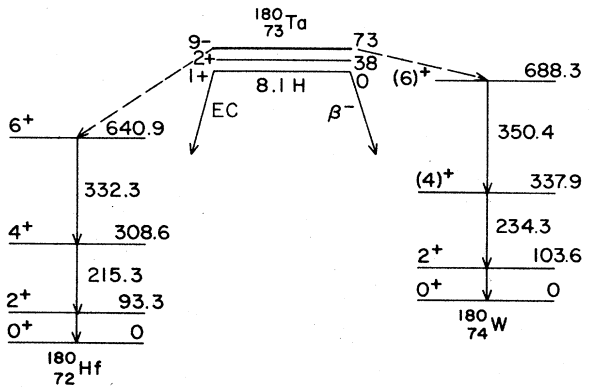

FIG. 1. Energy levels and γ transitions relevant to the search for the decay of $^{180}Ta^m$. Energies are in keV.

TABLE I. Fractions of unconverted γ rays, I_γ, for $E2$ transitions in ^{180}Hf, ^{180}W, and ^{176}Hf, in % per transition. γ energies are in keV.

^{180}Hf		^{180}W		^{176}Hf	
E_γ	I_γ	E_γ	I_γ	E_γ	I_γ
93.3	17.5	103.6	22.5	88.4	14.6
215.3	81.5	234.3	84.4	201.8	78.0
332.3	94.4	350.4	94.8	306.9	93.1

II. EXPERIMENTAL PROCEDURES AND γ-RAY EFFICIENCY CALIBRATION

Because of the low energies of the γ rays that would occur in the decay of $^{180}Ta^m$, high photopeak efficiencies can be expected, especially if a well-type detector is used. Accordingly a high-purity Ge detector of this type was procured. This same detector was recently used in a successful search[11] for the decay of ^{50}V. As described in the ^{50}V paper the characteristics of the detector are as follows: active volume $\gtrsim 100$ cm^3, efficiency for 1333-keV γ rays 25.5% relative to a 7.6×7.6-cm NaI(Tl) detector, and pulse-height resolution (FWHM) of 1.92 keV at 1333 keV. The well is 1.0 cm in diameter and 5.0 cm deep.

Studies of background in this detector when surrounded with Fe shielding showed that the dominant continuum background, in the energy region up to a few MeV, was due to cosmic rays. In order to reduce this yield the HPGe detector was enclosed in a large NaI(Tl) detector operated in anticoincidence. Two such detectors were used, the first on loan from Davis and Cleveland which was a single crystal 30.5 cm in diameter by 22.9 cm long with a well for the vertically-mounted HPGe detector to fit into. Tests with and without the NaI(Tl) detector in operation, when the latter was biased at a lower limit of 200 keV, showed that in the 200—500-keV region of the HPGe γ-ray spectrum the continuum background was reduced by a factor of about 5.5 when the anticoincidence condition was imposed. At the end of the loan period, another large NaI(Tl) system was substituted, a detector that had previously been used by der Mateosian and Goldhaber in some low-background applications. This detector is 25.4 cm in diameter by 30.5 cm long and has an 8.5-cm diameter hole all the way through. A standard 7.6×7.6-cm NaI(Tl) detector was inserted to fill the hole opposite to the HPGe detector and pulses from the six phototubes on the large NaI(Tl) and the phototube on the 7.6×7.6-cm NaI(Tl) were gain matched and added. Unfortunately the backgrounds of contaminant peaks and continuum levels in the HPGe detector with this NaI(Tl) system operating in anticoincidence were more than twice as great as in the case of the Davis-Cleveland crystal, so the results were of limited usefulness.

Considerations of the expected decay schemes for the EC and β^- decays of $^{180}Ta^m$ (see Fig. 1) suggested precautions that would have to be observed when using a Ge detector of essentially 100% solid angle. On the EC side, x rays emitted in most of the decays are in coincidence with the cascade γ rays. Additional coincident x rays (also in the β^- decay) result from the internal conversion of γ rays. If x rays are absorbed in the HPGe detector, the spectrum is complicated and intensity is removed from the photopeaks of the γ rays. Another complication is the absorption of the 93.3- and 103.6-keV γ rays which might have been useful in searching for triple summing peaks, except that the unconverted fractions of these γ rays are only 17.5% and 22.5%, respectively (Table I). It was therefore felt that the most easily interpreted and sensitive results would be obtained by reducing the x rays and unconverted low-energy γ rays of 93.3 and 103.6 keV to negligible levels by using an absorber, and by searching for the 215.3- and 332.3-keV γ rays and their 547.6-keV sum on the EC side, and the 234.3- and 350.4-keV γ rays and their 584.7-keV sum on the β^- side. Based on absorption calculations, a cylindrical Cu capsule 9.6 mm in diameter with a rounded bottom and a wall thickness of 1.0 mm was fabricated so as to contain the Ta_2O_5 sample.

An ideal source for testing γ-ray efficiencies is ^{176}Lu which decays to ^{176}Hf by β^- emission with a decay scheme very similar to the expected $^{180}Ta^m$ decays. The three main cascade γ rays of ^{176}Lu with their unconverted intensities are included in Table I. There is also a 0.9% β^- branch of ^{176}Lu to an 8^+ state giving a 401.1-keV γ ray in cascade, but this is weak enough to ignore. For the efficiency tests a Lu_2O_3 sample containing 150 mg of Lu was prepared. With such a sample placed in the Cu capsule, all conditions were nearly the same as for the $^{180}Ta^m$ measurements, including γ-ray self-absorption in the sample, absorption of γ's in the Cu, and γ-ray singles and summing efficiencies. Small corrections for differences in the fractions of unconverted γ rays were made to derive the appropriate efficiencies for the $^{180}Ta^m$ γ rays.

^{176}Lu, with an abundance[1] of 2.59(2)%, has had numerous half-life determinations. The three values quoted in the Table of Isotopes[8] (in units of 10^{10} yr), i.e., 3.79(3), 3.27(5), and 3.6(1), have a weighted average of $3.65(16)\times10^{10}$ yr. Meanwhile measurements of 4.08(24) (Ref. 12), 3.53(14) (Ref. 13), and 3.59(5) (Ref. 14) have been reported. We adopt for the half-life of ^{176}Lu the weighted average of these six values, namely $3.64(9)\times10^{10}$ yr. A sample containing 150 mg of Lu therefore decays with 8.07(21) disintegrations per second. Runs were made under a variety of conditions, i.e., with and without the Cu capsule and with and without anticoincidence of the NaI(Tl) detector. Figure 2 shows the spectrum of 201.8, 306.9, and 201.8 + 306.9 peaks when the sample was in the Cu capsule at the bottom of the well and with the anticoincidence in operation. In this case the bias on the NaI(Tl) was set at $\sim$350 keV when it was realized that the 307-keV γ rays escaping from the HPGe detector and absorbed by the NaI(Tl) could cancel the detection of some of the 202-keV γ rays if the bias on the NaI(Tl) was as low as 200 keV. Indeed, this effect of bias was measured in separate tests. The spectrum in Fig. 2 was from a run of 12 000 sec, and from the net peak intensities, combined with the 8.07/sec disintegration rate, the following absolute peak efficiencies, in % per disintegration, were derived:

201.8 12.2(6)% ,

306.9 13.7(7)% ,

201.8+306.9 9.4(5)% .

The spectrum observed with the Lu_2O_3 sample by itself, i.e., without the Cu capsule, was interestingly complex and included the following: a group of K-L and K-M x-ray lines in the range 54—64 keV associated with the internal conversion of γ rays; the summing of x rays giving another group of peaks at $2(K\text{-}L)$ and $2(K\text{-}M)$ due to the simultaneous internal conversion of pairs of the cascade γ rays; the three main γ-ray peaks of 88, 202, and 307 keV above each of which was a satellite structure of

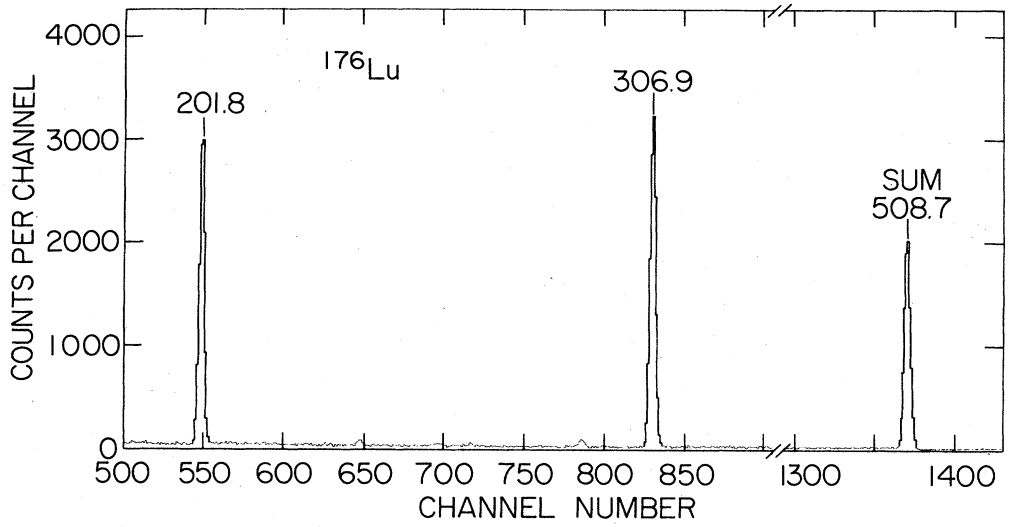

FIG. 2. γ-ray spectrum from a Lu_2O_3 sample containing 150 mg of Lu, enclosed in a Cu capsule and placed in the HPGe well detector. The latter was surrounded by a large NaI(Tl) detector operated in anticoincidence and biased at $\sim$350 keV.

γ-x-ray sum peaks; all three possible double γ-ray sum lines, each with a satellite of x-ray summing lines; and the triple γ-ray sum line, of course, with no satellite of x-ray peaks. In the tests taken with the Lu_2O_3 in the Cu capsule all summing peaks involving x rays and/or the 88.4-keV γ rays were essentially eliminated.

Tests were also made when the Lu_2O_3 source (in Cu) was placed at several positions in the well and at a point just outside the well. For this last condition the efficiencies for detecting the 202, 307, and 202 + 307 keV peaks were reduced by factors of $\sim$3.5 for the singles peaks and $\sim$20 for the sum peak relative to efficiencies when the source was at the bottom of the well. The 202 + 307 summing efficiency of $\sim$0.5% for this condition roughly corresponds to the coincidence efficiency that would be expected for γ's of these energies using two large Ge detectors on either side of a $^{180}Ta^m$ sample.

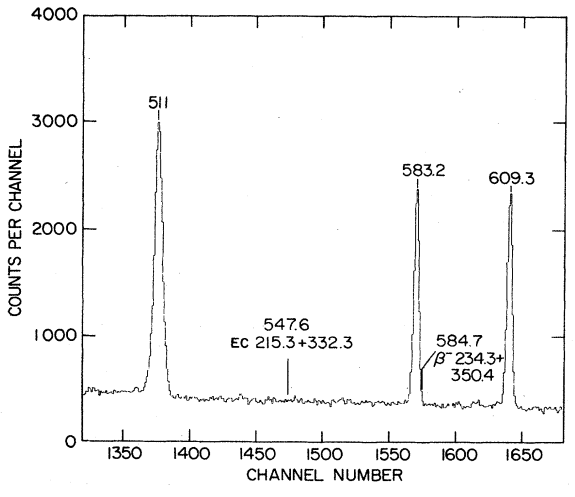

FIG. 3. Portion of the γ-ray spectrum in the region of the expected sum lines of $^{180}Ta^m$ from an 89-d run on a Ta_2O_5 sample containing 151 mg of Ta enriched to 5.47% in $^{180}Ta^m$. The well-type HPGe detector was enclosed in a large NaI(Tl) detector operated in anticoincidence.

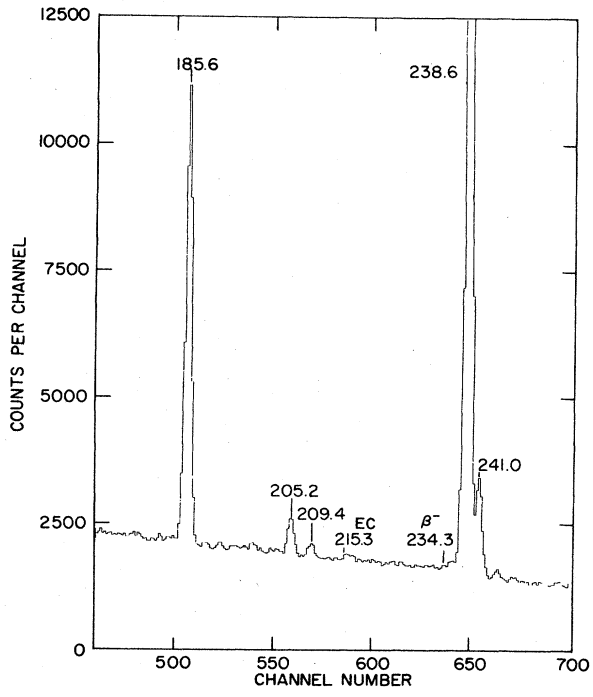

FIG. 4. Portion of the γ-ray spectrum in the region of the expected 215.3- and 234.3-keV peaks from $^{180}Ta^m$. See caption of Fig. 3 for other details.

III. RESULTS AND ANALYSIS

The sample[15] used for measurements on the decay of $^{180}Ta^m$ consisted of Ta_2O_5 containing 151 mg of Ta enriched to 5.47% in $^{180}Ta^m$. It was in the form of a powder enclosed in a plastic "pill" capsule. From the outset of the measurements it was apparent that the Ta_2O_5 sample contained small amounts of ^{152}Eu, ^{154}Eu, ^{133}Ba, and ^{137}Cs activities. Altogether eleven ^{152}Eu peaks and five from ^{154}Eu were observed, but their contributions to the background were negligible. (Based on the spectrum shown in Fig. 5 and the γ-ray efficiencies discussed above the ^{152}Eu strength is $\simeq 1.1\times10^{-7}$ μCi while that of ^{133}Ba is $\simeq 4\times10^{-8}$ μCi.) Note that the ^{152}Eu 344.3-keV peak seems to be present in Norman's spectrum, Fig. 2 of that paper,[10] but was not identified.

γ-ray spectra were recorded in a Nuclear Data 4096-channel pulse-height analyzer operated in an automatic mode such that the spectrum was recorded for 24 h, taped, the analyzer cleared, and a new run started. As described in the ^{50}V paper[11] each spectrum was inspected for pulse-height resolution and then gain shifted to normalize the positions of reference peaks (in this case peaks at 238.6, 583.2, and 1460.8 keV) so that good resolution would be preserved in the grand sum spectrum. Some of the individual spectra exhibited a high 511-keV γ-ray component due to the emission into the atmosphere of ^{15}O activity from the BLIP facility (Brookhaven linac isotope producer). An unfavorable wind direction sometimes results in the intake of the ^{15}O into the air conditioning system of the BNL Chemistry building. Eight spectra containing an excessive 511-keV background were rejected.

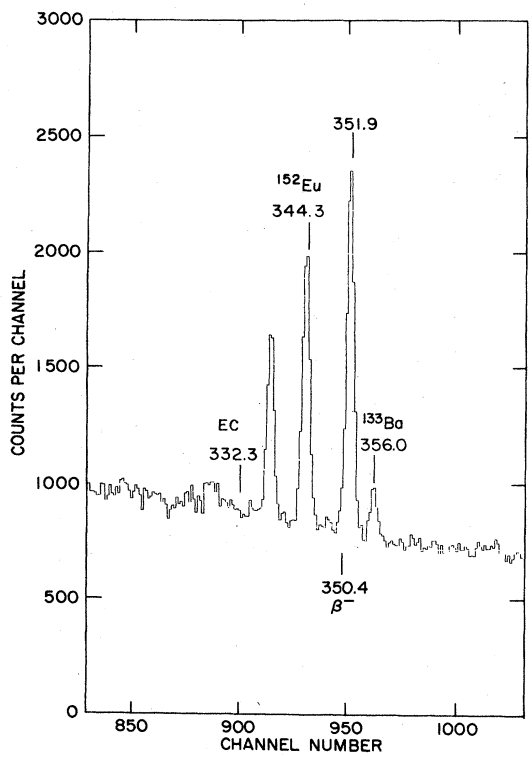

FIG. 5. Portion of the γ-ray spectrum in the region of the expected 332.3- and 350.4-keV peaks from $^{180}Ta^m$. See caption of Fig. 3 for other details. Note the 344.3-keV ^{152}Eu peak and the 356.0-keV ^{133}Ba peak from contaminant activities in the Ta_2O_5 sample (see the text for strengths).

Three main runs were made on the Ta_2O_5 sample, the first being a 110-d run with iron shielding around the HPGe detector and without a Cu capsule. For the second run, totaling 89 d of acceptable spectra, the Cu capsule was used as well as the Davis-Cleveland NaI(Tl) detector in anticoincidence at 200-keV bias. Finally, a run of 116 d was made with the der Mateosian-Goldhaber NaI(Tl), the Cu capsule, and a bias of $\sim$350 keV on the NaI(Tl).

The best results were obtained in the second series of runs. Figure 3 shows the region of this spectrum that would contain the γ-ray sum peaks. In this, as well as in Figs. 4 and 5, all observed peaks could be assigned to known contaminant and background activities, mostly those belonging to the ^{232}Th-^{208}Pb decay chain. The 547.6-keV sum (215.3 + 332.3) on the EC side of $^{180}Ta^m$ is in a very flat region of Fig. 3 free of contaminant lines. A calculation based on an upper limit of 156 counts (95% confidence) for the intensity of the sum peak, the number of $^{180}Ta^m$ atoms present as derived from the sample weight and enrichment factors, the length of the run, and a sum peak efficiency of 10.0% derived from the ^{176}Lu data, with appropriate corrections, gave the half-life limit for EC

$$T_{1/2} > 3.0\times10^{15} \text{ yr} .$$

Unfortunately, the corresponding sum peak of 584.7 keV (234.3 + 350.4) on the β^- side would be separated by only 1.5 keV from the 583.2-keV background peak. An analysis of this region in Fig. 3 gave an upper limit of 500 counts for such a peak.

From an inspection of Figs. 4 and 5, it was found that a somewhat better limit on the β^- decay branch could be obtained from the possible 234.3-keV peak in Fig. 4 which is well separated from the strong 238.6-keV background peak. Here the upper limit on the 234.3-keV line is <320 counts, and when combined with the efficiency and other factors as discussed above, leads to a limit of $T_{1/2} > 1.9\times10^{15}$ yr for β^- decay. The total half-life of $^{180}Ta^m$ is therefore $T_{1/2} > 1.2\times10^{15}$ yr.

IV. DISCUSSION

The results derived above, i.e., $T_{1/2} > 3.0\times10^{15}$ yr for EC and $T_{1/2} > 1.9\times10^{15}$ yr for β^- decay have increased the lower limits on $T_{1/2}$ of $^{180}Ta^m$ by factors of $\sim$50 for EC decay and $\sim$35 for β^- decay as compared with the most recent previous search.[10] Corresponding to these half-lives are the values $\log f_0 t > 22.9$ for EC decay and $\log f_0 t > 21.3$ for β^- decay.

In their summary of forbidden β decays, Raman and Gove[16] listed only one measured nonunique third-forbidden β transition. This is the $^{87}Rb \rightarrow {}^{87}Sr$ β^- decay, $\frac{3}{2}^- \rightarrow \frac{9}{2}^+$ which has a $\log f_0 t = 17.6$ and is within the expected range. Thus, the $\log f_0 t$ limits for both the EC and β^- decay of $^{180}Ta^m$ would appear to be very large and, as

noted by Norman,[10] suggests the possibility that these are highly K-forbidden transitions.

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