from the original spectrum (energies below 35 Mev were not considered), the percentage decrease being independent of energy and having the value of approximately 35%. No correction was made for this effect in view of the assumptions in the calculation, but the order of magnitude of the effect has been referred to in the text.

(iv) Summary of Applied Corrections and Uncertainties

The following list gives the uncertainties in the factors used to derive the absolute cross sections:

Solid angle of telescope	1%
Efficiency of copper monitor	3.5%
Statistical uncertainty in copper monitor	1%
Cross section of $Cu^{63}(\gamma,n)Cu^{62}$ reaction	5%
Half-life of Cu^{62}	1.5%
Bremsstrahlung calculations	assumed precise
Tail of $Cu^{63}(\gamma,n)Cu^{62}$ reaction	2%
Proton telescope bracket width	3% for $E_p<60$ Mev 1.3% for $E_p>60$ Mev
Nuclear attentuation correction	1%

The over-all precision of the cross sections excluding counting statistics:

7.6% for $E_p<60$ Mev, 6.4% for $E_p>60$ Mev.

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Natural Radioactivity of V^{50} and Ta^{180}

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The methods of proportional counter and scintillation spectroscopy have been used to examine the possible radioactivity of the naturally occurring nuclides V^{50} and Ta^{180}. Some positive evidence is obtained that V^{50} decays by K capture to the 1.58-Mev excited state of Ti^{50} with a half-life of $(4.8\pm1.2)\times10^{14}$ yr. A search for titanium K x-rays gave negative results. For Ta^{180} a lower limit of $(2.3\pm0.7)\times10^{13}$ years was found for the half-life against decay by K capture and a lower limit of $(1.7\pm0.6)\times10^{13}$ years against beta decay.

1. INTRODUCTION

TWO naturally occurring odd-odd nuclei have been discovered during the last decade in investigations using a mass-spectrometer. They are[1] $_{23}V^{50}$ and[2] $_{73}Ta^{180}$; it may be expected that they are both radioactive.

$_{23}V^{50}$ is the central member of the isobaric triplet $Ti^{50}-V^{50}-Cr^{50}$ and should be unstable against both beta emission and electron capture. Its relative abundance is 0.25%.[3] From direct mass measurements, the energy available for the decay of V^{50} to Ti^{50} is 2.39 ± 0.13 Mev and from V^{50} to Cr^{50} is 1.18 ± 0.12 Mev.[4] The spin of V^{50} in its ground state is known to be 6.[5] A search for the radioactivity of V^{50} by other workers has so far yielded negative results.[6] In an earlier investigation in this laboratory, using Geiger counter technique, a lower limit for beta emission of 10^{12} years was found. In the present work we have searched for K x-rays of Ti with a shielded proportional counter and have examined the gamma-ray spectrum of vanadium using a scintillation spectrometer within an iron and mercury shield.

From measurements of the proton spectra in the (d,p) reaction[7] on Ti^{50}, it appears that the first excited level of Ti^{50} is at 1.58 Mev above the ground state. In accordance with the general behavior of excited states of even-even nuclei, this is almost certainly a 2+ state. Because of the high spin value of V^{50} ($I=6$), it is to be expected that V^{50} will prefer to decay with a high probability to this excited level of Ti^{50}, rather than decay by a direct transition to the 0+ ground state, despite the smaller energy difference. In this case one would expect electron capture to be accompanied by a gamma ray of 1.58 Mev. It was decided, therefore, to look particularly for a 1.58-Mev gamma ray.

$_{73}Ta^{180}$ has the very low abundance of 0.012%.[3] Its stable isobaric neighbors are $_{72}Hf^{180}$ and $_{74}W^{180}$. A short-lived isomer of Ta^{180} has been known for some years. It is produced in the reaction $Ta^{181}(\gamma,n)Ta^{180}$ and has a half-life of 8 hours.[8] Its decay scheme according to Brown *et al.*[8] is shown in Fig. 1. The average total internal conversion coefficient of the two gamma rays of 93 and 102 kev has been found to be 4.6 and the average K to L ratio is 0.15. It is to be expected that in the decay of the natural Ta^{180} the same gamma rays might be found. From the decay of the 5.5-hr isomer $_{72}Hf^{180}$,[9] a series of excited levels of Hf^{180} are known, which fit

[1] D. C. Hess, Jr., and M. G. Inghram, Phys. Rev. **76**, 1717 (1949).
[2] White, Collins, and Rourke, Phys. Rev. **97**, 566 (1955).
[3] White, Collins, and Rourke, Phys. Rev. **101**, 1786 (1956).
[4] W. Johnson, Phys. Rev. **87**, 166 (1952).
[5] Kikuchi, Sirevetz, and Cohen, Phys. Rev. **92**, 109 (1953).
[6] W. T. Leland, Phys. Rev. **76**, 1722 (1949); H. Selig, thesis, Carnegie Institute of Technology, 1954 (unpublished); J. Heintze, Z. Naturforsch. **10A**, 77 (1955).
[7] G. F. Pieper, Phys. Rev. **88**, 1299 (1952).
[8] Brown, Bendel, Shore, and Becker, Phys. Rev. **84**, 292 (1951).
[9] Mihelich, Scharff-Goldhaber, and McKeown, Phys. Rev. **94**, 794 (1954).

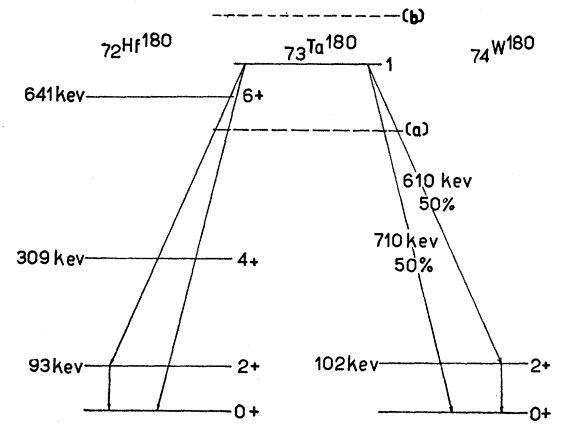

FIG. 1. The decay scheme of 8-hr Ta^{180} and excited levels of Hf^{180}. The energy released in the decay of Ta^{180} to Hf^{180} is unknown.

very well the rotational states of the Bohr-Mottelson model. Because of the high spin value which should certainly be ascribed to the natural long-lived Ta^{180} ($I=1$ was proposed for the 8-hr Ta), it is to be expected that if enough energy is available, Ta^{180} would preferably decay to the higher energy states of its neighbors, with their higher spin values. One then would expect to find gamma rays with energies which equal those appearing in the rotational spectra of Hf^{180}. It is probable that a similar rotational spectrum exists in the excited states of W^{180}, although this has not yet been confirmed experimentally.

A priori, one does not know which of the two levels of Ta^{180} is the ground state and which is the isomeric state. If the naturally occurring Ta^{180} is the ground state [possibility (a) in Fig. 1] then the energy available for beta decay will be less than 700 kev. If, on the other hand, natural Ta^{180} is an excited isomeric state [possibility (b) in Fig. 1], then a possible mode of decay of the latter could be by a direct transition to the ground state of Ta^{180} which would then decay with an 8-hr half-life, and one should be able to detect the known radiations of Ta^{180}.

In this work, a search was made for gamma rays of energy of approximately 100 and 200 kev and for K x-rays from hafnium and tungsten of energy 54 and 58 kev, which would result from K capture and from internal conversion from the 93- and 102-kev energy states.

Eberhardt *et al.*[10] have attempted to find evidence for the radioactivity of Ta^{180} in old minerals. Their results were negative and they obtained a lower limit of 10^{12} years for beta decay.

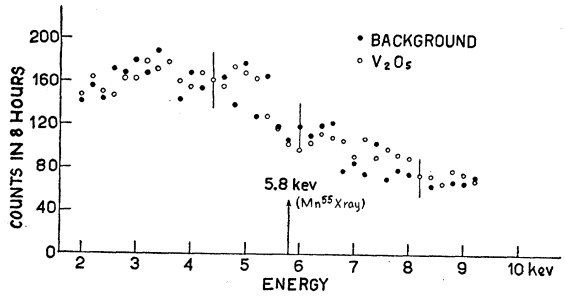

FIG. 2. Spectrum of pulse heights obtained from vanadium oxide and background spectrum, using a proportional counter. The vertical lines indicate typical statistical errors.

APPARATUS

For the search for K x-rays of Ti, which would follow the K-capture decay of V^{50} to Ti^{50}, a shielded proportional counter was used. The proportional counter was 30 cm long and 5 cm in diameter and the high-tension electrode was equipped with guard rings to eliminate spurious counts. The counter was calibrated with Mn x-rays from Fe^{55}, which entered the counter through a 4 mg/cm^2 beryllium window. For these x-rays (5.8 kev) the resolution was 18%. The counter was shielded with anticoincidence counters and a 2-ton iron shield. The integral background from the shielded counter was about 16 counts/minute, above a bias corresponding to 2 kev energy.

The pulse-height spectrum was analyzed by photographic methods. The pulses from the proportional counter, after linear amplification, were displayed on an oscilloscope screen after being stretched to 50 microseconds. The pulses from the anticoincidence counters, after being lengthened, served to modulate the intensity of the oscilloscope, so that simultaneous proportional counter pulses were not recorded. The pulses were then photographed on a slowly moving film and analyzed after projection.

For the analysis of the gamma-ray spectra, NaI(Tl) crystals mounted on a Dumont $K1234$ photomultiplier were used. The crystals were shielded by 15 cm of iron, 2 cm of mercury, and 7 mm nickel. The function of the nickel was to absorb the characteristic x-radiation of mercury of about 70–80 kev, which results from the K ionization arising in the interaction of the background radiation with the mercury shield.

For the investigation of vanadium, a large crystal was used ($1\frac{3}{4}$ in. in diameter and 2 in. thick), which gave a resolution of 8% for 660-kev gamma radiation. For the measurements on tantalum, where we are concerned with low-energy radiation, a small crystal of NaI is more advantageous in order to diminish background effects. In this case a crystal of $\frac{1}{2}$ in. diameter and $\frac{1}{2}$ in. in thickness was used. In order to diminish the absorption at low energies a light reflector of aluminum without magnesium oxide was used. The resolution of the apparatus with this crystal was 9.5% at 660 kev, 18% at 100 kev, and 24% at 40 kev.

The pulse-height spectra were analyzed first by means of the photographic method mentioned above, and later by an 80-channel pulse-height analyzer of the Hutchinson-Scarrott type.

[10] Eberhardt, Geiss, and Lang, Z. Naturforsch. **10a**, 796 (1955).

3. EXPERIMENTAL RESULTS WITH VANADIUM

The background spectrum of the proportional counter was measured with a clean aluminum sheet inside the counter, since aluminum foil was used as a support for the vanadium sample. It was found that the aluminum available was slightly contaminated and increased the integral background rate above 2 kev by 2–3 counts per minute. In the search for x-rays from vanadium, 4 g of spectrosocopically pure V_2O_5 were spread uniformly over an area of 370 cm^2 on the aluminum sheet by using a solution of 5% collodion in butyl acetate. This was formed into a cylinder and placed into the counter. The results are shown in Fig. 2. There is no evidence for K x-rays. A lower limit of $(3.3\pm1.2)\times10^{13}$ years for decay by K capture was deduced from these results.

In the search for gamma rays from V^{50}, 32 g of spectroscopically pure V_2O_5 were used. This was firmly packed around the crystal with 14 g placed in front of the crystal on an area of approximately 20 cm^2 and 18 g surrounding the crystal on the cylindrical surface, on an area of 60 cm^2. This distribution of the source was estimated to be the most effective, after measuring the relative efficiency of the crystal for 1.33-Mev gamma rays with a point source of Co^{60} placed at different points around the crystal. In order to calibrate the energy scale, the gamma rays from Co^{60} (1.17, 1.33 Mev), Cs^{137} (660 kev), and K^{40} (1.46 Mev) were used. The spectrum of the vanadium sample and the background were taken during frequently alternating periods for a total counting time of 306 hours. The over-all stability of the counter and electronic equipment was checked constantly throughout the measurements, by examining at frequent intervals the gamma spectrum produced by Cs^{137} and Co^{60}.

TABLE I. Total counts recorded for different energy ranges for the background and in presence of vandium oxide.

Energy range in Mev	1.2–1.65	1.65–2.2	1.5–1.65
Background counts	32 950±180	18 310±130	7780±90
V_2O_5 counts	33 960±180	18 350±130	8030±90
Difference	1010±250	40±180	250±120

The experimentally recorded spectra between the energy limits of 1200 and 2200 kev are shown in Fig. 3. The ordinate shows separately the total number of counts recorded per channel for background and also in the presence of the V_2O_5 sample. The period of counting in each case was 153 hours.

It is seen from Fig. 3 that throughout the energy region from 1.2 to 1.6 Mev more counts are recorded in the presence of the vanadium oxide sample. The excess number of total counts recorded in the presence of V_2O_5 in this energy range during the whole counting interval was 1010±250. No excess counts were recorded above 1.65 Mev. In this region the pulse spectra recorded in the two cases are seen to be identical. The numerical results for the integral number of counts are summarized in Table I.

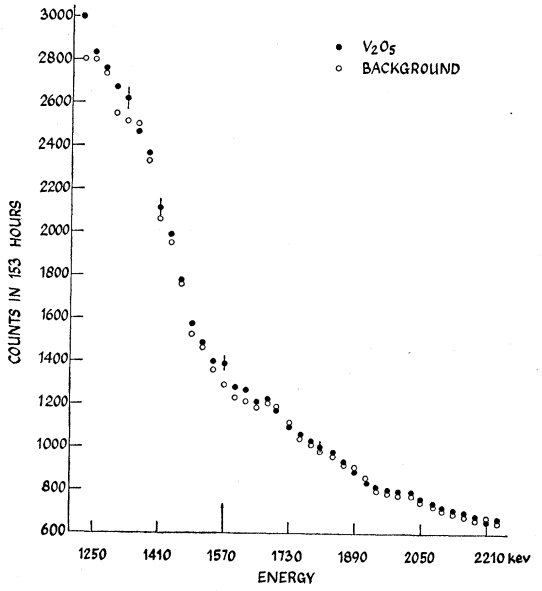

FIG. 3. Spectrum of pulse heights obtained from vanadium oxide and background spectrum, using a scintillator.

These results indicate with a high degree of likelihood that gamma radiation is emmitted from the vanadium sample, producing a scintillation pulse spectrum with an upper limit at about 1.65 Mev. This conclusion is consistent with the assumption that a gamma ray of 1.58 Mev is emitted. Because of the statistical errors it is difficult to draw definite conclusions from the actual shape of the difference between the two recorded spectra shown in Fig. 3. This difference spectrum is shown in Fig. 4(a). It is seen that more counts are recorded in the region of the expected full peak at 1.58 Mev. The calibration spectrum obtained with K^{40} is shown in Fig. 4(b) on the same energy scale. The smooth line drawn in Fig. 4(a) would be the expected

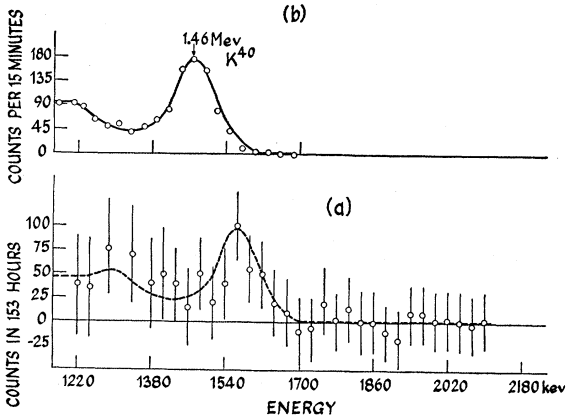

FIG. 4. (a) Pulse-height spectrum due to V_2O_5 after subtraction of background. (b) Pulse-height spectrum due to K^{40} from sample of potassium metabisulphite, on same energy scale.

shape of the scintillation pulse spectrum for the 1.58-Mev gamma ray. The total excess number of counts recorded in the region of the expected photopeak is 250 ± 120 (see Table I). In very many runs which were carried out over a long period of time, the essential shape of the spectra described above was found to be reproducible and hence would seem to be a real effect.

Since a 1.46-Mev gamma ray appears in the disintegration of K^{40}, which has a half-life of 1.28×10^9 years and a relative abundance of 0.012%,[11] and since this energy is close to the energy of the expected gamma ray from V^{50}, it was necessary to exclude the possibility of contamination by potassium. In fact, in the early stages of this work excessive background in the energy range concerned was obtained because of the 1.46 gamma ray of K^{40}, arising from the potassium content in the glass of the photomultiplier tube which was originally used. This effect was not found in the Dumont tube which was finally used. In order to estimate accurately the concentration of potassium which it would be necessary to assume exists in the vanadium sample in order to explain the observed results, the gamma-ray spectrum arising from K^{40} was measured after surrounding the crystal with 41 g of potassium metabisulphite ($K_2S_2O_5$) which was distributed in exactly the same geometry as the vanadium [Fig. 4(b)]. The measurement of the ratio of the intensity of the gamma rays in the two experiments then gives the ratio of the amounts of K^{40} in the two samples, under the assumption that the observed counts in the vanadium are due completely to potassium contamination. In this way it was found that one must assume a concentration of 1 part in 10^4 of potassium by weight in the vanadium sample, in order to explain the results by contamination. This is at least two orders of magnitude greater than the upper limit of the concentration of potassium estimated by spectroscopic analysis.

While the statistical errors do not permit absolutely certain conclusions regarding the energy and intensity of the detected radiations from the vanadium sample, we feel that the experimental results provide fair evidence for radioactivity of V^{50} by the expected mode of decay.

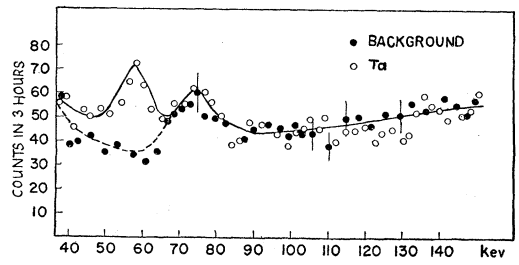

FIG. 5. Pulse-height spectrum obtained from tantalum and background.

[11] McNair, Glover, and Wilson, Phil. Mag. **1**, 199 (1956).

The same peak at 1.58 Mev was also observed with far better statistics and in the same relative intensity in a much larger sample of 500 g of V_2O_5. This material was not spectroscopically standardized, but since it gave essentially the same results, we regard this as a confirmation of the existence of a 1.58-Mev gamma ray.

From these results one can arrive at an estimate of the half-life of V^{50} in terms of the half-life of K^{40}, using the measured intensity of the 1.46-Mev gamma ray obtained from a known quantity of potassium sulphite under similar geometrical conditions, as described in the previous paragraph. After making small corrections for the change in efficiency of the scintillator crystal between energies 1.46 Mev and 1.58 Mev and also for small changes in self-absorption of the gamma rays in the solid samples for the two energies, we arrive at the following result for the ratio of the half-lives:

$$T_{\frac{1}{2}}(V^{50})/T_{\frac{1}{2}}(K^{40})=(3.8\pm0.9)\times10^5.$$

Using the most recent estimate of the half-life of K^{40}, $(1.28\pm0.02)\times10^9$ yr,[11] we obtain the value of $(4.8\pm1.2)\times10^{14}$ yr for the half-life of V^{50} for decay by K capture to the 1.58-Mev level of Ti^{50}.

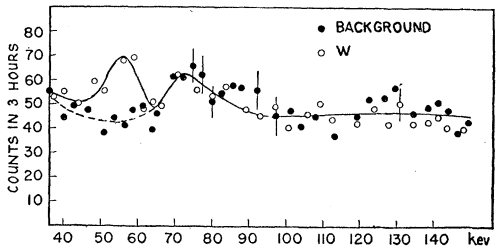

FIG. 6. Pulse-height spectrum obtained from tungsten and background.

In a search for the radioactivity of vanadium, using Geiger counter techniques, Heintze[6] obtained some excess counts, which he ascribed to impurities. In the light of the present results it seems probable that his effect was a real one.

After completion of this work, a recent paper of Glover and Watt[12] has appeared in which a search for a 1.58-Mev gamma ray from V^{50} was made, using a scintillation counter in a manner similar to the above. In this work again some positive evidence for the expected gamma ray is obtained, especially in the Compton region. In the present work, the evidence for the full peak at 1.58 Mev seems more convincing. The half-life estimated by these authors is $(4.0\pm1.1)\times10^{14}$ yr, in fair agreement with the present results.

4. EXPERIMENTAL RESULTS WITH TANTALUM

The energy calibrations of the scintillator in this experiment were made using a source of Gd^{153}, which

[12] R. N. Glover and D. E. Watt, Phil. Mag. **2**, 697 (1957).

gives radiations at about 42 kev and 103 kev. The Gd spectra were photographed during the experiment each hour and showed that the stability of the equipment was excellent.

The tantalum was examined in the form of three sheets of pure material, each of thickness 300 mg/cm². These sheets were disposed on the plane surface of the cylindrical crystal and also around the curved surface over an over-all area of about 32 cm². Similar sheets of pure tungsten were examined for comparison.

First, the scintillation spectrum obtaining with tantalum was compared with the background spectrum (Fig. 5). A clear peak at approximately 55 kev shows up in the tantalum spectrum. This fits quite well the energy of the K x-rays of tantalum. A similar peak, however, was also obtained with the tungsten sheets (Fig. 6). It must be concluded that most of this peak results from the interaction of background activity with the materials, which produces K ionization and accompanying characteristic x-rays.

TABLE II. Characteristics of long-lived K-capture transitions.

Element	Half-life in years	Transition	Energy in Mev	% K-capture	logft
$_{19}K^{40}$	$(1.28\pm0.02)\times10^{9}$	2 yes	0.03 ±0.07	10.8±0.4	13.33
$_{21}Ca^{41}$	$(1.2\pm0.4)\times10^{5}$	2 yes	0.44 ±0.02	100	10.78
$_{28}Ni^{59}$	$(7.5\pm1.3)\times10^{4}$		1.065±0.03	100	11.87
$_{23}V^{50}$	$(2.2\pm0.4)\times10^{14}$	4 yes	0.810±0.13		21.1

In further experiments, the spectra resulting from tantalum and tungsten were measured over a period of 18 hours, 9 hours with tantalum, and 9 hours with tungsten, interchanging the two materials each hour. The results are shown in Fig. 7. The correspondence of the two spectra is seen to be very good, except possibly between 55–60 kev, where there is a small surplus of counts with the tantalum sample. This is indeed the region of energy of the x-rays which would result as a consequence of K capture or internal conversion in the K shell. However, this surplus is only slightly outside the standard error and therefore the results provide only slight evidence for the radioactivity of Ta^{180}. From the results one obtains a lower limit of 50 counts in 9 hours between 55 and 60 kev which might be produced by the tantalum, and therefore a lower limit of $(2.1\pm0.4)\times10^{13}$ yr for the half-life of decay by K capture to the ground state of Hf^{180}. Considering the possibility of decay by K capture to an excited state, one must take into account that some of the K x-rays would result from K conversion. For this possibility we obtain

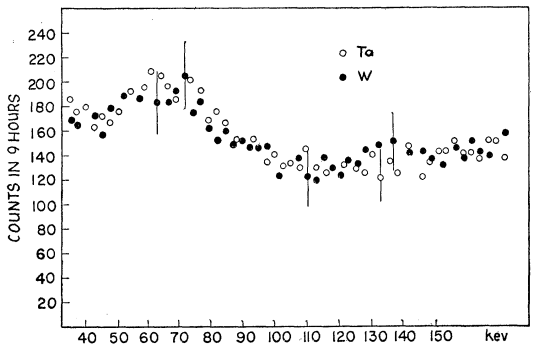

FIG. 7. Pulse-height spectrum obtained from tantalum and tungsten.

a lower limit for the half-life of $(3.5\pm0.7)\times10^{13}$ yr. For the possible decay of Ta^{180} by negatron decay to the first excited level of W^{180} or positron decay to the first excited level of Hf^{180}, we obtain a lower limit of $(1.7\pm0.3)\times10^{13}$ yr.

5. DISCUSSION

The energy difference between the ground state of V^{50} and the 1.58-Mev level of Ti^{50} is about 810 kev. This, together with our results for the half-life for decay by K capture to the 1.58-Mev level of Ti^{50} leads to a logft value of 21.1 for this transition.

In the Table II, we compare the logft values of the few very long-lived K capture transitions that are known, together with the other characteristics of these transitions, when known.[13]

Returning to the possibility mentioned in the introduction that naturally occurring Ta^{180} may be an isomeric state of an 8-hr ground state, it would be interesting to see if 8-hr Ta^{180} could be separated from naturally occurring tantalum. However, a rough estimate shows that very high spin differences between the two levels would be required ($\Delta I\sim8$) in order that the half-life of the level should be greater than 10^{13} years and that simultaneously the decay by direct de-excitation should compete seriously with beta decay to the rotational levels of high spin in W^{180}, which may be assumed to exist. Hence this possibility seems rather unlikely but not impossible. If a good chemical method of separating the isomers could be devised, a very good lower limit to the half-life for this mode of decay could be obtained, because of the concentration of the 8-hr activity.

[13] J. K. Major and L. C. Biedenharn, Revs. Modern Phys. **26**, 321 (1954).