New Neutron-Deficient Isotopes of Tantalum*

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Bombardment of Ho_2O_3 with N^{14} ions in the Berkeley heavy-ion linear accelerator has resulted in the discovery of new isotopes of tantalum which have been assigned as Ta^{173} and Ta^{174}. They have half-lives of 3.7 hr and 1.3 hr, respectively. Tantalum-172 was not observed and is believed to have a half-life shorter than 30 minutes. Gamma-ray spectra have been obtained for these two isotopes and for Ta^{175}. Tantalum-175, with an 11-hr half-life, has also been produced by 48-Mev alpha-particle bombardment of Lu_2O_3, and its conversion-electron spectrum was studied. From these data a decay scheme is proposed using nine of the observed transitions and assigning spins to three members of the ground-state rotational band.

INTRODUCTION

THE Berkeley heavy-ion linear accelerator may be used to produce very neutron-deficient isotopes by such reactions as (N^{14},xn), where x can conveniently be made 4 or greater by energy selection. Excitation functions are very sharp for these reactions, making it possible to produce relatively pure isotopic species. This method has been utilized to produce short-lived isotopes of tungsten which subsequently decay to tantalum isotopes not previously discovered. Recently reported[1] 11-hr Ta^{175} was produced in this manner and also by cross-bombardment with alpha particles to positively mass-assign the isotope and at the same time obtain enough activity to study its decay characteristics in more detail. No evidence was found for the 30-minute isotope reported by Dohan and Naumann.

EXPERIMENTAL PROCEDURE

Thin Ho_2O_3 powder targets were bombarded with N^{14} ions for periods of $\frac{1}{2}$ to 3 hr at energies ranging from 35 to 95 Mev. The alpha-particle bombardment was carried out at 45 Mev on a thin Lu_2O_3 powder target.

After irradiation the target was dissolved in HCl, and HF was added to bring the concentration to about 6*N* HCl and 1*N* HF. Carrier-free tantalum was extracted into di-isopropyl ketone (DIPK). After acid washing of the organic layer, samples were evaporated for study by means of a 3-inch high by 3-inch diameter cylindrical beveled sodium iodide (Tl) scintillation crystal used with a 100-channel pulse-height analyzer. Samples were also evaporated to dryness on a thin aluminum strip for use in the double-focusing electron spectrometer.[2] Samples for the permanent-magnet electron spectrographs as previously described by Smith and Hollander[3] were obtained by back-extraction of the tantalum activity into water, followed by cathode electrodeposition onto a 10-mil platinum wire from $(NH_4)_2C_2O_4$ buffer solution.

EXPERIMENTAL RESULTS

Ta^{173}

Using the nuclear masses computed by Cameron[4] and assuming a compound-nucleus reaction, one estimates the threshold for formation of W^{173} to be about 66 Mev, and the peak of the excitation function of the reaction should be at about 85 Mev. Tantalum-173 should decay to 24-hour Hf^{173}, whose two prominent gamma rays at 124 and 298 kev should be observed growing into the sample.

At bombarding energies above 70 Mev the two peaks are observed. The growth-rate curve of the 298-kev

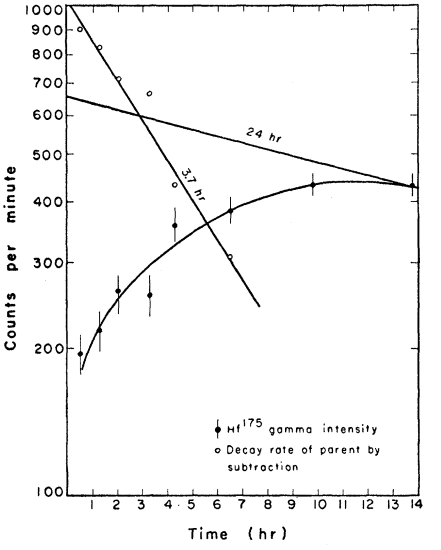

FIG. 1. The half-life of Ta^{173} obtained from the growth rate of the 298-kev gamma-ray photopeak in the daughter nuclide, 24-hour Hf^{173}.

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[1] P. Bro. F. C. Dohan and R. A. Naumann, Second Regional Meeting, Delaware Valley Section of the American Chemical Society, February 5, 1958. Titles of papers listed in Chem. Eng. News, January 13, 1958, Vol. 36, p. 72.

[2] G. D. O'Kelley, University of California Radiation Laboratory Report, UCRL-1243, June, 1951 (unpublished).

[3] W. G. Smith and J. M. Hollander, Phys. Rev. **101**, 746 (1956).

[4] A. G. W. Cameron, Atomic Energy of Canada Limited Report 433, CRP-690 (unpublished).

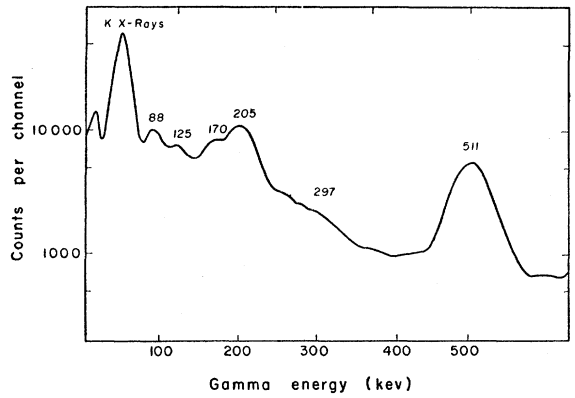

FIG. 2. Gamma pulse-height spectrum of Ta^{173} and Ta^{174} produced by N^{14} bombardment of holmium at 95 Mev.

photon peak is shown in Fig. 1. Resolution of the curve gives a decay rate for the parent Ta^{173} nuclide of 3.7 hr. Figure 2 is a gamma-ray spectrum of the tantalum activity produced by this bombardment, which was at 95 Mev. Two photopeaks, one at 170 kev and one at 90-kev energy, decay with the right half-life to be identified as belonging to this isotope. The other gamma rays and nearly all of the 511-kev annihilation radiation peak decay more rapidly. No gamma rays were seen with energies above 500 kev.

On the basis of this evidence we assign to Ta^{173} a half-life of about 3.7 hr, a high electron-capture-to-positron ratio, and gammas of 90 and 170 kev. Gamma-gamma coincidence measurements indicate that these two transitions are in coincidence with each other.

No additional radiations are seen at higher bombarding energies, and from the time elapsed between bombardment and measurement one may conclude that isotopes of tantalum of mass less that 173 have half-lives shorter than about 30 min.

Ta^{174}

At bombarding energies of 60 Mev and higher an isotope is formed which decays with a half-life of about 1 hr. Energy considerations predict that W^{174} can be formed above about 55 Mev with an excitation function peak around 75 Mev. Figure 3 shows the gamma spectrum of the tantalum activity 1 hr after N^{14} bombardment at 70 Mev. One notes a very large 511-kev annihilation radiation peak, indicating a large amount of positron emission in this isotope. Cameron[4] estimates that there is about 4.5 Mev of decay energy for Ta^{174}, which would produce a large positron-to-electron-capture ratio.

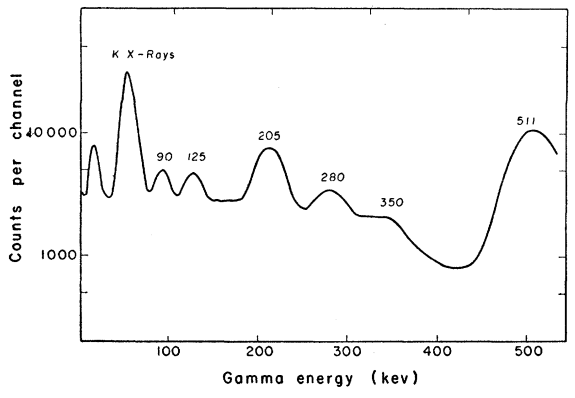

FIG. 3. Gamma pulse-height spectrum of Ta^{174} produced by N^{14} bombardment of holmium at 70 Mev.

There are five photopeaks at energies of 90, 125, 205, 280, and 350 kev, shown in Fig. 3. These peaks all decay with about the correct 1-hr half-life and are believed to belong to this isotope. Coincidence measurements show another peak at about 160 kev which is not seen clearly in this spectrum. The half-life of the isotope can readily be determined by following the decay rates of the gross activity of the material as measured by an end-window G-M counter and of a single photon in the gamma spectrum. The results of these two methods indicate a half-life of 1.3 ± 0.1 hr.

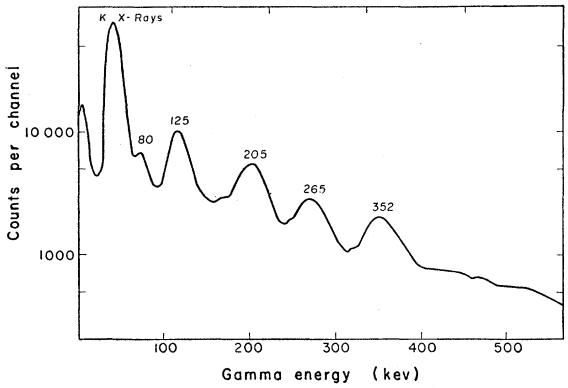

FIG. 4. Gamma pulse-height spectrum of Ta^{175} produced by N^{14} bombardment of holmium at 60 Mev.

Gamma-gamma coincidence measurements were made on this spectrum; gating on the 511-kev photons showed the 90-, 205-, and 160-kev gammas in coincidence, and gating on the 205-kev photon indicated that the 90- and the 280-kev gammas were in coincidence with it. No gamma rays of sufficient abundance to be detected are to be seen at energies above 500 kev. Attempts to produce enough of the isotope to study its conversion electrons spectroscopically have not been successful.

Ta^{175}

At bombarding energies just above 45 Mev, the nuclear reaction threshold for an isotope whose half-life is 11 hr is observed. Figure 4 is a tantalum gamma-ray spectrum taken 10 hr after a 60-Mev bombardment. The time lapse allowed for the decay of Ta^{174}. This spectrum was obtained on a 1-inch high by $1\frac{1}{2}$-inch diameter cylindrical NaI(Tl) crystal. Nearly all the peaks are distorted, indicating a very complex spectrum.

The decay rate of this isotope, as obtained from following both gross activity and single photopeaks, is 11 hr. The lack of evidence for appreciable 511-kev radiation suggests a small positron branching ratio and thus a relatively small decay energy. The Cameron mass equation gives 1.8 Mev as the decay energy of Ta^{175}. Following the decay of this 11-hr isotope, a long-lived activity remains which has a prominent 340-kev gamma ray. Since this is presumably 70-day Hf^{175}, the mass assignment seems quite certain. The assignment is further confirmed by cross-bombardment with alpha particles as discussed below.

Ta^{175} Conversion-Electron Spectra

In Ta^{175} one is not limited to the relatively small amounts of activity produceable in the heavy-ion accelerator; a much larger amount of activity can be obtained by the use of the Berkeley 60-inch cyclotron. The reaction employed was $Lu^{175}(\alpha,4n)Ta^{175}$. Following bombardment in the intense cyclotron alpha-particle beam, samples of sufficient strength for conversion-electron measurements were obtained. Table I lists the transitions whose conversion-electron lines have been seen and confirmed as having the correct half-life. Spectra were obtained on three different permanent-magnet electron spectrographs. In addition to the transitions listed, a number of others were seen and identified as belonging to other isotopes. The most intense lines in the spectrum were those due to the presence of Ta^{176}. A weak line whose half-life could not be confirmed was used in the decay scheme as the K line for the 140.9-kev transition.

Relative electron line intensities were determined in two ways. First the lines on the photographic plates described above were compared visually with lines of known relative intensities on a set of standard plates made by exposing a source of Pa^{233} for varying known time intervals. Second, the Ta^{175} spectrum was measured in the double-focusing electron spectrometer, which gave accurate intensity data but less resolution. In both methods measurement of the K line of the 81.6-kev transition was so uncertain as to make impossible any estimate of the K/L ratio for the transition. A summary of the intensity measurements is given in Table II. The data from the electron spectrometer have been normalized to the visual comparison readings.

Table III summarizes the multipolarity assignments of eight of the transitions. The best-characterized transition seems to be that at 126.2 kev. Therefore, the gamma-ray-intensity data are normalized to the observed intensity of the 126-kev gamma ray. These gamma-intensity figures are only approximate because of the complexity of the spectrum as seen in Fig. 4. The 70.5-kev and 81.6-kev transitions must be characterized by L-subshell ratios only, since their K-line intensities are not known. The 81.6-kev transition might be $E1$, for which the theoretical subshell ratio is

TABLE I. Ta^{175} conversion-electron energy; summary for electron lines seen in three different spectrographs.

Transition energy (kev)	100-gauss magnet	200-gauss magnet	350-gauss magnet
50.50	$L_I\ L_{II}\ L_{III}\ M_I\ M_{II}\ M_{III}\ N$		
70.53	$L_I\ L_{II}\ L_{III}\ \ M_{II}\ M_{III}$		
77.43	$L_I\ L_{II}\ L_{III}$		
81.57	$K\ L_I\ L_{II}\ L_{III}\ M\ \ N\ \ 0$		
104.3	$K\ L_I\ L_{II}\ L_{III}$		
126.2	$K\ L_I\ L_{II}\ L_{III}\ M\ \ N$		
157.1	$K\ L$		
162.5	$K\ L$		
179.4	$K\ \ L_{II}\ L_{III}$	$L\ M\ N$	
186.0	$K\ L$	L	
190.8	K	$L\ M$	
193.3	K	L	
207.9	$K\ L$	$K\ L$	
235.6	K	L	
267.2		$K\ L\ M\ N$	
349.0		$K\ L\ M$	$K\ L\ M$
394.1		K	$K\ L$
437.3		$K\ L$	$K\ L$

TABLE II. Ta^{175} conversion-electron intensity; summary.

Transition energy (kev)	Subshell	Intensity: Visual comparison method[a]	Intensity: Double-focussing spectrometer
50.5	L_I	10.2	
	L_{II}	9.5	
	L_{III}	11.5	
70.5	L_I	3.0	
	L_{II}	7.3	
	L_{III}	4.5	
81.6	L_I	28	
	L_{II}	8.8	51.3
	L_{III}	8.7	
104.3	K	40	39.4
	L_I	9.4	
	L_{II}	2.3	10.3
	L_{III}	1	
126.2	K	20	
	L_I	2.1	
	L_{II}	9.3	19.7
	L_{III}	8.0	
157.1	K	1	
162.5	K	9.6	<11.7
179.4	K	1.9	
	L	0.67	
186.0	K	2.8	
190.8	K	3.0	
207.9	K	4.7	
267.2	K	15	14.9
	L		2.3

[a] Intensities have a probable error of about 20% except that relative subshell intensities (close-lying lines) are accurate to about 15%.

TABLE III. Ta^{175} conversion coefficients and multipolarity assignments.

Transition energy (kev)	Relative electron intensity	Normalized gamma intensity[a]	Multipolarity[b]	α Theoretical	α Observed	K/L Theoretical	K/L Observed[c]	$L_I/L_{II}/L_{III}$ Theoretical	$L_I/L_{II}/L_{III}$ Observed[d]
50.5	31		60% $E2$ 40% $M1$	4.9				1/0.95/1.0	1/0.90/1.1
70.5	15(L)		35% $E2$ 65% $M1$	9.1		1.0		1/2.0 /1.9	1/2.5 /1.5
81.6	18(L)	<9	7% $E2$ 93% $M1$	6.2		3.9		1/0.30/0.22	1/0.31/0.30
104.3	53		7% $E2$ 93% $M1$	3.5		5.2	3.8	1/0.22/0.14	1/0.24/0.11
126.2	40	29.5	94% $E2$ 6% $M1$	1.35	1.35[a]	0.99	1.0	1/4.9 /3.9	1/4.7 /3.7
162.5	12	7	$M1$	1.14	1.7	7.2	8		
179.4	2.5		$E2$	0.36		1.4	2.8		
207.9	5.5	25	$M1$	0.53	2.2	6.9	5–8		
267.2	17	20	$M1$	0.26	0.85	7.3	6		

[a] Gamma intensities normalized to electron intensities to give correct α for the 126.2-kev transition.
[b] Determined by best fit to observed K/L and $L_I/L_{II}/L_{III}$.
[c] Probable errors are about 30%.
[d] Probable errors in these ratios are about 20%.

1:0.35:0.42, but if this were the case, the intensity of the gamma ray would have to be 550, in contradiction to the relative photon intensities (Fig. 4). The same argument may be used for the 104.3-kev transition. The large K/L ratio for the 162.5-kev transition indicates either an $M1$ or $E1$, the quadrupole transitions being ruled out. Since the conversion coefficient for an $E1$ transition is only 0.096, an $M1$ is indicated with perhaps some $E2$ admixture. The 179.4-kev transition has a K/L ratio of less than 2.8, since only one L line is included, and an $E2$ assignment is given since the next lowest value is for $M2$ and is 4.6. The last two transitions, at 207.9 and 267.6 kev, both have high K/L ratios and have been assigned as $M1$. $E1$ conversion coefficients would be 0.051 and 0.028, respectively, and the quadrupole transitions are ruled out by their low K/L ratios. Both these transitions may have some $E2$ admixture, since the conversion coefficients appear to be somewhat high.

Theoretical conversion coefficients were obtained from the tables of Sliv and Band.[5]

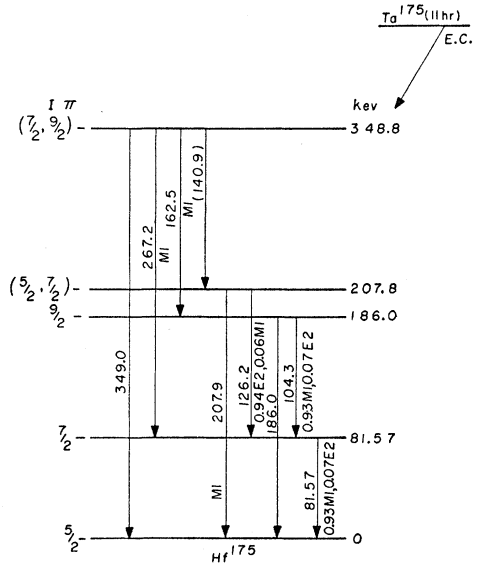

FIG. 5. Proposed decay scheme of Ta^{175}.

Ta^{175} Decay Scheme

On the assumption that the multipolarities given in Table III are correct, the most intense transition is at 81.6 kev. On this basis the transition is assumed to proceed to the ground state. From transition-energy sums one may then assign levels at 186.0, 207.8, and 348.8 kev, as shown in Fig. 5.

Since Hf^{175} is in the region of spheroidal nuclei, we seek neutron-state assignments in the Nilsson scheme. Hafnium-175 has 103 neutrons, and the asymptotic states [based on the $K\pi(N,n_z,\Lambda)$ notation] most probable as the ground states in this region[6] are the $\frac{5}{2}-(512)$ as seen in Yb^{173} or the $\frac{7}{2}-(514)$ as seen in W^{177}, which has 105 neutrons. Other near-lying intrinsic states as seen in neighboring nuclei are the $\frac{7}{2}+(633)$ and $9/2+(624)$. The energy of the first excited state of the $\frac{7}{2}$ band is around 113 kev, while that in the $\frac{5}{2}$ band

[5] L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Report, 1956 [translation: Report 57 ICCK1, issued by Physics Department, University of Illinois, Urbana, Illinois (unpublished)]; and Report 58ICCL1, 1958.
[6] B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat-fys Medd. (to be published).

is 79. Since the 81.6-kev transition in Ta^{175} decays with an $M1$–$E2$ mixture which is typical of intraband transitions, it is believed that this state is the $\frac{7}{2}$ member of the $K=\frac{5}{2}$ rotational band. The $I(I+1)$ energy dependence predicts that the energy of the 9/2 member should be 187 kev, which is very close to the observed level at 186.0. This level decays mostly by an $M1$–$E2$ mixed transition of 104.3 kev to the 81.6-kev level and by a weaker 186.0-kev crossover. This pattern is entirely consistent with an assignment of the 186-kev level as the 9/2 member of the ground rotational band associated with the $\frac{5}{2}-(512)$ neutron orbital.

The state at 348.8 kev decays by $M1$ transitions to the 9/2- and $\frac{7}{2}$-members of the ground-state band and must therefore be a $\frac{7}{2}$- or 9/2-state belonging to a different intrinsic configuration, presumably the $\frac{7}{2}-(514)$. The state at 207.8 kev must have spin $\frac{5}{2}$ or $\frac{7}{2}$ with negative parity. None of the other observed transitions can be unambiguously assigned to the decay scheme.

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Scattering of High-Energy Nucleons by a Nonlocal Potential*

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The scattering of high-energy nucleons by a simple nonlocal potential is examined in the Born approximation. It is shown that an energy dependent local potential is not fully equivalent to a nonlocal potential. The latter potential introduces an additional angular dependence in the differential cross section which seems to be particularly significant in the backward directions.

THE generalized optical potential has been shown by Feshbach[1] to be both nonlocal and energy-dependent. In the interpretation of the scattering data by an optical model, the empirical potential was, however, commonly taken as local and energy-dependent. In an infinite medium, it is of course true that one can replace the nonlocality of the potential by an explicit energy-dependence.[2] But for the scattering from a finite nucleus, the situation is more complicated, and this simple equivalence should not be expected to hold. The purpose of this note is to show that at least in the Born approximation, the nonlocality of the potential does introduce some modification in the differential cross section also and hence is not reflected entirely by an additional energy dependence.

The nonlocal Schrödinger equation,

$$\hbar^2/2M\nabla^2\psi+E\psi=\int K(\mathbf{r},\mathbf{r}')\psi(\mathbf{r}')d\mathbf{r}', \quad (1)$$

with the phenomenological kernel

$$K(\mathbf{r},\mathbf{r}')=V[(\mathbf{r}+\mathbf{r}')/2]\delta_a(\mathbf{r}-\mathbf{r}') \quad (2)$$

has been used by several authors[3] to describe nuclear bound state problems and scattering phenomena at low energies. Using the effective mass approximation for finite nuclei,[4] it was found that Eq. (1), with the particular choice of kernel given by (2), can indeed yield a fairly satisfactory description in the low-energy region; thus, in this study, we shall retain this special form of the phenomenological kernel. Since our attention will be restricted toward high-energy nucleon-nuclear scattering, the effective mass approximation is no longer valid; hence our results will depend somewhat on the choice of the form factor $\delta_a(\mathbf{r}-\mathbf{r}')$ to be used in Eq. (2).

The nonlocal scattering amplitude f_{NL} may be obtained by examining the asymptotic behavior of ψ in the usual way. The result is

$$f_{\mathrm{NL}}=-(\tfrac{1}{4}\pi)\int \exp(-i\mathbf{k}\cdot\mathbf{r})V[(\mathbf{r}+\mathbf{r}')/2] \times\delta_a(\mathbf{r}-\mathbf{r}')\psi(\mathbf{r}')d\mathbf{r}d\mathbf{r}', \quad (3)$$

where $\mathbf{k}$ is the wave number of the scattered wave, and V contains the factor $2M/\hbar^2$. By introducing relative and "center-of-mass" coordinates and the Fourier

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[1] H. Feshbach, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Palo Alto, 1958), Vol. 8, p. 49; Ann. Phys. **5**, 357 (1958).

[2] W. E. Frahn, Nuovo cimento **4**, 313 (1956).

[3] W. E. Frahn and R. H. Lemmer, Nuovo cimento **6**, 1221 (1957); A. E. S. Green, Revs. Modern Phys. **30**, 569 (1958); A. E. S. Green and P. C. Sood, Phys. Rev. **111**, 1147 (1958).

[4] W. E. Frahn and R. H. Lemmer, Nuovo cimento **5**, 1564 (1957).