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Neutron Deficient Radioactive Isotopes of Tantalum and Wolfram*

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Neutron deficient radioactive isotopes of tantalum have been produced by bombardment of lutetium with α-particles, of hafnium with 10 Mev protons, and of tantalum with fast neutrons and protons from 10 to 70 Mev energy. In addition, wolfram isotopes produced by 10 to 70 Mev proton bombardments of tantalum, together with their respective tantalum daughter activities were studied. The radiation characteristics of the new activities of tantalum and wolfram are described.

I. EXPERIMENTAL

USING the 60-inch Crocker Laboratory cyclotron, bombardments were made of lutetium with 38, 30 and 20 Mev α-particles, of hafnium with 10 Mev protons, and of tantalum with fast neutrons from bombardment of beryllium with 19 Mev deuterons. Tantalum metal foil was bombarded with different energies of protons from the 184-inch cyclotron and the linear accelerator. The techniques of bombardments and of measurement of radiation characteristics by absorption methods, together with assumptions involved in the interpretation of data, have been described previously.[1,2]

The lutetium oxide targets were dissolved in nitric acid, hafnium oxide and tantalum targets in the minimum of a mixture of nitric and hydrofluoric acids. Where appropriate, carrier solutions of hafnium nitrate, sodium wolframate and tantalum in hydrofluoric acid were added, together with holdback carriers for contaminating activities likely to be formed from target materials; e.g., copper, platinum, etc. Radiochemical exchange of hafnium, tantalum, and wolfram activities with added carriers, was ensured by boiling the target solutions in hydrofluoric acid.

Lutetium and rare earth carriers added for scavenging, were removed by precipitation of the fluorides from hot solutions $3N$ in both nitric and hydrofluoric acids. Wolfram was separated using the following procedure. To the hot hydrofluoric acid solution of the tantalum target, hydrazine sulfate was first added to reduce any nitric acid present. Sufficient strong hydrochloric acid to give a $6N$ solution was added followed by stannous chloride solution, then ammonium thiocyanate solution followed by excess boric acid to complex the fluoride; the green thiocyanate complex of IV valent wolfram formed, was then extracted into ethyl acetate. The washed solvent layer was evaporated, holdback carriers for various elements were added and wolfram VI oxide precipitated by boiling with strong nitric acid. The oxide was dissolved in ammonium hydroxide, thiocyanate added to the solution and the reduction-extraction process repeated after acidification.

Hafnium and tantalum were precipitated as phosphates from boiling $3N$ nitric acid solution after complexing the fluoride ion with boric acid. The phosphates were metathesized with sodium hydroxide.

Hafnium and tantalum were separated by several methods. The mixed hydroxides were fused with sodium hydroxide and the cold melt extracted with water to give an insoluble residue of hafnium oxide and a solution of sodium tantalate, from which tantalum was recovered by acidification and precipitation of the phosphate; hafnium was extracted from $>8N$ hydrochloric acid solutions by tributylphosphate in butanol or from perchloric acid solutions by thenoyltrifluoracetone in benzene—the latter extraction was used in the search for hafnium daughters of tantalum activities where no hafnium carrier was added and the benzene extracts evaporated and ignited on trays for counting. When hafnium was not the target, carrier quantities were separated from tantalum solutions $3N$ in both

* This work was performed under the auspices of the AEC.
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[1] G. Wilkinson, Phys. Rev. 75, 1019 (1949).
[2] G. Wilkinson and H. G. Hicks, Phys. Rev. 75, 1370 (1949).

nitric and hydrofluoric acids by precipitation of barium hafnium fluoride.

After removal of hafnium, tantalum was recovered as the phosphate which was dissolved in the minimum of hydrofluoric acid. Niobium carrier was added and the tantalum separated by precipitation of potassium tantalum fluoride. This precipitate was heated to fuming with strong sulphuric acid to remove the fluoride and the phosphate precipitated from $2N$ acid solution.

A simple separation of tantalum and wolfram used in studying tantalum daughter activities was found to be quantitative, and, further to give a decontamination factor of greater than 10^6. The tantalum daughter activity was separated by scavenging the boiling strongly alkaline wolframate solution with nickel hydroxide. The washed hydroxide was dissolved in acid, excess wolframate solution added and the boiling solution again made alkaline. After three cycles, the active tantalum was finally collected on a small amount of zirconium hydroxide by precipitation from hydrochloric acid solution by addition of ammonium hydroxide; the large amounts of nickel used as a scavenger remain in solution.

In all cases the tantalum and wolfram oxides were prepared for counting and estimation of chemical yields by ignition of the hydrated oxides or phosphates.

In Table I are summarized the production and radiation characteristics of tantalum and wolfram isotopes. The energy of gamma-radiation is obtained from lead and aluminum absorption measurements while the energy of positive and negative electrons were obtained from both aluminum absorption measurements and from a simple magnetic spectrometer. The contribution of soft electromagnetic radiations in aluminum absorption measurements was obtained by removal of electrons by beryllium absorbers.

All the radioactive isotopes of tantalum and wolfram studied decay with the emission of electromagnetic radiations of half thicknesses 16 to 18 mg/cm^2 aluminum and 115 to 130 mg/cm^2 lead, which correspond to the average energies of L and K x-radiation of hafnium and tantalum respectively.

In the bombardment of lutetium with α-particles of 38, 30 and 19 Mev energy isotopes of half-lives 8.0 hours, 53 hours, 2.1 hours, and about 600 days were observed, the latter being in very low yield. The magnitude, and variation in yield of the first three isotopes at the various energies, agree with those observed previously for the production of thulium and rhenium activities by α-particle bombardment of holmium and tantalum respectively, and allow fairly certain mass allocation as in Table I. The long-lived activity is presumably formed from the 2.5 percent abundant Lu^{176} isotope. In the 10 Mev proton bombardment of hafnium, the 2.1 hour, 53 hour and long-lived activities were formed in approximately equal yields; the 8-hour component, which must be a complex of the 8.0-hour Ta^{180} unequivocally assigned by fast neutron bombardment of tantalum and the 8.0-hour Ta^{176}, was in a somewhat higher yield; these yields agree with the present allocations. The 8.0-hour Ta^{176} and 53-hour Ta^{177} have been shown to grow from wolfram parent activities and a further tantalum isotope of half-life 9.35 minutes was found to be the daughter of a 21.5-day wolfram isotope. The wolfram activities were assigned from the yields at various bombarding energies and agree with the above allocations of their daughter activities. The 9.35-min. tantalum must be allocated to Ta^{178} and is thus probably an independent isomer of the 2.1-hour activity; the latter has not been observed to grow from a wolfram parent.

The possibility of a long-lived isomer of Ta^{180} exists, but no evidence for such an isotope has been obtained in fast neutron bombardments where the long-lived background was only the 117-day Ta^{181} formed by neutron capture. No difference in the absorptions or decays of the long-lived activities from $Lu+\alpha$, $Hf+p$, or $Ta+p$ bombardments has been observed and all appear to be identical.

No hafnium daughter activities, which would be isomers of stable hafnium isotopes, with half-lives greater than five minutes have been observed.

II. TANTALUM ISOTOPES

16±2-min. Ta^{182}

This activity was known previously and the cross section for production in thermal neutron capture in tantalum has been measured.[3] The aluminum absorption has been remeasured. The approximate ratio of the radiations, 0.12 Mev e^-: 0.6 Mev β^-: $K+\gamma$ rays $=\sim 0.2:\sim 0.05:1$ suggests that the isotope decays by isomeric transition with a few percent branching by beta particle emission.

8.00±0.05-hour Ta^{180}

This previously known[4] activity, which is produced by fast neutron bombardment of tantalum has been restudied; the isotope was also produced by bombardment of tantalum with protons by the p, pn reaction.

The decays were followed through eleven half-lives. The aluminum and lead absorption curves of the activity show the radiations to consist of beta-particles, range 210 mg/cm^2 aluminum (0.6 Mev), Feather range 238 mg/cm^2 (0.7 Mev) L and K x-radiation and 1.3 Mev gamma-radiation. On the magnetic spectrometer the distribution of the electrons was that of a beta-particle of maximum energy 0.7 Mev. From the measurements, with usual corrections and assumptions counting efficiencies, the following ratios were obtained: 0.7 Mev β^-: L x ray: K x-ray: 1.3 Mev γ-ray$=0.13:\sim 1:1:\sim 0.01$.

[3] Seren, Friedlander, and Turkel, Phys. Rev. **72**, 163 (1947).
[4] G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).

TABLE I. Production and characteristics of tantalum and wolfram isotopes.

Isotope	Type of radiation	Half-life	Energy of radiation in Mev: Particles	Energy of radiation in Mev: γ-rays	Produced by
Ta^{176}	K, e^-, γ	8.0±0.1 hr.	0.12, 0.18, ~1	~2	Lu-α-3n W^{176} K decay
Ta^{177}	K, e^-, γ	53±2 hr.	0.11	~1.4 (weak)	Lu-α-2n, 3n Hf-p-n W^{177} K decay
Ta^{178}	K, e^-, β^+(~3%), γ	2.1±0.1 hr.	~0.1 e^- ~1 β^+	1.3–1.5	Lu-α-n Hf-p-n Ta-p-$p3n$
Ta^{178}	K, e^-, β^+(~6%)γ	9.35±0.05 min.	0.08 e^-1.06 β^+	~1.5	Hf-p-n W^{178} K decay
Ta^{179}	K, e^-, γ	~600 days	~0.1	~0.7 (weak)	Lu-α-n Hf-p-n W^{179} K decay?
Ta^{180}	K, e^-, β^-(~15%)γ	8.00±0.05 hr.	0.7 β^-	1.3	Ta-p-$p2n$ Ta-n-$2n$
Ta^{182}	I.T. e^-, γ, β^-(<5%)	16±2 min.	0.12 e^-0.6 β^-		Ta-p-pn Ta-n-γ
W^{176}	K, e^-, β^+(~0.5%)γ	80±5 min.	~0.1, ~0.2, e^-~2β^+	~1.3	Ta-p-6n
W^{177}	K, e^-, γ	130±3 min.	0.13 e^-~0.4 e^-	~0.45, 1.2	Ta-p-5n
W^{178}	K, γ	21.5±0.1 days		~0.27 (weak)	Ta-p-4n
W^{179}	K	30±1 min.	No e^-		Ta-p-3n
W^{179}	K or I.T.	5.2±0.2 min.			Ta-p-3n

The isotope thus appears to decay predominantly by orbital electron capture with about 15 percent branching by negative beta-particle emission.

~600-day Ta^{179}

After decay of shorter-lived activities the tantalum fractions from Lu+α bombardments show a very weak long lived activity. This activity was also found 10 Mev proton bombardments of hafnium and in tantalum bombarded with high energy protons. The isotope is not formed by decay of wolfram parents of half-life greater than one hour, and since no short-lived tantalum daughters of the 30-min. wolfram activity definitely allocated to mass 179 have been found, it is most likely that the 600-day activity has mass 179. This allocation is consistent with the yields in α-particle bombardments of lutetium and in proton bombardments of tantalum.

The radiation characteristics were measured on samples from 10 Mev proton bombardments of hafnium and from 40–50 Mev proton bombardments of tantalum. The approximate ratio of the various radiations were: 0.1 Mev e^-: L x-rays: K x-rays: ~0.7 Mev γ-ray = ~0.03: 1:1: ~0.03. The isotope thus appears to decay predominantly by orbital electron capture.

9.35±0.03-min. Ta^{178}

The decay of this activity separated from its 21.5-day wolfram parent was followed through 13 half-lives; aluminum, beryllium and lead absorptions of the radiations were made, corrections for decay during time of measurement being required. Study on a simple magnetic spectrometer showed conversion electrons of energy ~80 kev together with positrons of maximum energy 1.06 Mev; the energies agree with those obtained from the aluminum absorption.

The ratios of the various radiations, necessarily approximate in view of the various assumptions made, are e^-: β^+: L x-ray: K x-ray: 1.5 Mev γ-ray = ~0.5: 0.03: ~1:1: 0.03.

As discussed below, L and K x-radiation arise from both orbital electron capture in tantalum and from conversion in γ-ray transitions in the hafnium product, and it is reasonable to assume that ~0.5 of the K x-radiation results from orbital electron capture. On this basis, the 9.3-min. activity shows approximately 6 percent positron branching decay.

The 9.3-min. activity has been observed directly in the bombardment of hafnium with 10 Mev protons. Allocation of the 21.5-day wolfram parent is made to mass 178 on the basis of reaction yields; since a longer-lived tantalum has been allocated to mass 178 from Lu+α bombardments the 9.35 min. activity must be an isomer decaying independently. Since no long-lived activity has been observed to form from high intensities of the 9.3-min. activity, an upper limit for decay by isomeric transition can be set as 10^{-4} that for decay by orbital electron capture.

2.1±0.1-hour Ta^{178}

This activity was produced in 19 Mev α-particle bombardments of lutetium and also in proton bombardments of hafnium when it was formed in high yields corresponding to its allocation to mass 178. The radiation characteristics obtained from resolution of absorption curves gave approximate ratios of ~0.1 Mev e^-: ~1 Mev β^+: L x-ray: K x-ray: ~1.4 Mev γ-ray = ~0.3: ~0.02: ~1:1: ~0.5 showing decay by orbital electron capture with about 3 percent positron branching.

53±2 hours Ta^{177}

This activity has been observed after decay of the shorter-lived isotopes in the tantalum fraction from

Lu+α and Hf+p bombardments. An activity, identical in all respects was shown to be the daughter of the 134-min. wolfram activity by successive chemical separations at fixed time intervals. The decays of samples from the three sources were followed through over nine half lives. The approximate ratios of the various radiations are 0.11 Mev e^-: L x-ray: K x-ray: 1.4 Mev γ-ray = $\sim$0.3: $\sim$0.5: 1: 0.005.

8.0±0.1 hour Ta^{176}

This isotope was formed in high yield in bombardment of lutetium with 38 Mev α-particles, and has also been found to grow from a wolfram parent of half-life 80 min. formed by bombardment of tantalum with protons of energy greater than $\sim$50 Mev. The decays of the several components of the radiation were separately followed through about eight half-lives. The ratios of the various radiations are, approximately 120 kev e^-: 180 kev e^-: $\sim$1 Mev e^-: L x-ray: K x-ray: 1.2 Mev γ-ray = $\sim$0.7: $\sim$0.02: $\sim$0.5: 1: $\sim$0.6.

III. WOLFRAM ISOTOPES

The bombardment of tantalum with protons of energy 10 to 70 Mev has led to the characterization of five new radioactive isotopes of wolfram. The 140-day W^{181} activity produced by p, n and $d, 2n$ reaction in tantalum has been described previously.[5]

30±0.1-min., 5.2±0.2-min., W^{179}

These activities were found in the wolfram fraction from 40–60 Mev proton bombardments of tantalum on the 184-inch cyclotron and, in bombardments with 20 to 30 Mev protons from the linear accelerator in yields corresponding to $p, 3n$ reaction. Chemical separation of tantalum after decay of the wolfram activity showed a very weak long-lived activity which is probably due to the long-lived Ta^{179} discussed previously. No short-lived tantalum daughter activities were found. The decay of the 30-min. activity was followed through seven half-lives. No electrons or hard gamma-rays were observed, and the isotope appears to decay with the emission of L and K x-rays only. The ratio of L to K x-radiation in the 30-min. activity corrected for counting efficiencies, fluorescence yield, etc. was about 0.7: 1 suggesting simple electron capture decay.

The 5.2-min. activity whose decay was followed through six half-lives emits L and K x-rays and soft electrons; the ratio of the activities of the 5.2- and 30-min. activities in various bombardments appeared to be the same, and the activities are most likely isomeric.

21.5±0.1-day W^{178}

This activity was shown to be formed by bombardment of tantalum with protons of energy greater than $\sim$25 Mev in yields corresponding to a $p, 4n$ reaction. The decay of the isotope in equilibrium with the 9.35-min. tantalum daughter activity, has been followed through ten half-lives.

In order to obtain the radiation characteristics of the wolfram activity, the aluminum, beryllium, and lead absorption curves of the gross parent-daughter equilibrium mixture were measured. The daughter activity was then quantitatively separated and its activity measured. The equilibrium contribution of the 9.3-min. tantalum to the absorption curves of the gross activity were estimated, and the absorption of radiations of the wolfram parent obtained by subtraction.

The 21.5-day activity appears to decay by emission of L and K x-rays and a weak gamma-ray only. No electrons were observed. The ratios of the various radiations corrected for counting efficiencies, Auger effect, etc., are: e^-: L x-ray: K x-ray: 0.27 Mev γ-ray = $<$0.05: $\sim$0.8: 1: $\sim$0.02.

From the measurements, the ratios of the L and K x-radiations for the 21.5-day and 9.3-min. activities can be obtained. The ratio of quanta emitted by the tantalum activity to quanta emitted by the wolfram parent activity was 1.7 for K x-radiation and 2.0 for L x-radiation. Thus, if it is assumed that one K x-ray quantum emitted by the wolfram activity represents one disintegration by orbital electron capture (only 0.75 L x-ray quanta are emitted per K electron captured due to direct transitions from M and lower shells), then it is clear that 0.4 of the L and K x-rays of the 9.3-min. tantalum daughter must arise following K shell conversion in γ-ray transitions following orbital electron capture. This figure agrees roughly with the number of electrons per K x-ray observed in the tantalum activity.

No daughter activity other than the 9.3-min. tantalum activity has been found to grow from the 21.5-day wolfram isotope.

130±3-min. W^{177}

This isotope was observed in bombardments of tantalum with protons of about 40 to 65 Mev from the 184-inch cyclotron. The decays of electron and electromagnetic radiations followed separately through seven half-lives.

The ratios of the various radiations, corrected for counting efficiencies, etc. are: 0.13 Mev e^-: 0.4 Mev e^-: L x-ray: K x-ray: 0.4 Mev γ-ray: 1.2 Mev γ-ray = $\sim$0.1: $\sim$0.02: $\sim$0.3: 1: $\sim$0.5: 0.2.

In order to characterize daughter activities of this isotope, the wolfram fraction containing the 130-min. activity was "milked" for tantalum activities. The tantalum fraction was found to contain only a pure 53-hour activity whose decay was followed through seven half-lives and whose radiations are identical with those of the 53-hour Ta^{177} described above. The yield of the tantalum activity in successive separations shows it to be the daughter of the 130-min. wolfram activity. No evidence for shorter- or longer-lived

[5] G. Wilkinson, Nature **160**, 864 (1947).

daughter activities was obtained. Allocation of the 130-min. activity to mass 177 is made on the basis of recognition of the 53-hour Ta^{177} daughter activity and also from considerations of yield in bombardments at various energies which agree with formation by a p, $5n$ reaction.

80±5-min. W^{176}

This activity was formed in bombardments of tantalum with protons of energy greater than about 50 Mev, and was obtained in a relatively pure yield in short bombardments of 0.2-mil tantalum foil with 70 Mev protons. The radiation characteristics were obtained from absorption measurements carried out immediately after chemical separation. The approximate ratios of the various radiations were ~0.1 Mev e^-: ~0.2 Mev e^-: ~2 Mev β^+: L x-ray: K x-ray: γ-ray = ~0.1: ~0.02: ~0.005: ~1: ~1: ~0.1.

The activity was shown to be the parent of the 8.0-hour Ta^{176} activity by chemical separations at successive time intervals. The half-life of the wolfram activity was obtained by measurement of the activity of aliquots of a stock solution taken at fifteen minute intervals and counted immediately after removal of the tantalum daughter activity. The decay through five half-lives was obtained after correction from the 130-min. W^{177} activity initially present to the extent of about five percent. The 53-hour Ta^{177} activity was found in small yield in the decays of the 8-hour daughter of the 80-min. wolfram as would be expected.

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Resonance Capture of Neutrons by Uranium*

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The resonance capture of slow neutrons by U^{238} has been studied by absorption methods with "good" geometry and thin detectors using the cyclotron as an intense source of neutrons. The experimental cross section for self-absorption for slow neutrons transmitted by cadmium was measured with thin absorbers and found to be 4600×10^{-24} cm²/atom±40 percent. Evidence is given for the existence of more than one level. The predominant level has an energy somewhat under 11 ev, a total natural width somewhat less than 0.20 ev and a neutron width somewhat less than 0.0086 ev. The value $\int\sigma dE/E$ in the resonance region was found to be 290×10^{-24} cm²/atom±40 percent.

I. INTRODUCTION

IT is known that when neutrons are captured by uranium, either a fission of the uranium nucleus may take place, or the neutron may be captured in a typical resonance process, resulting in a radioactive isotope of uranium, U^{239}, which decays by β-emission of 24-minute half-life.[1] Since the latter process absorbs but does not produce neutrons, its investigation is of interest in relation to the problem of obtaining a self-propagating fission process with uranium.[2]

The process of resonance capture is characterized by a large capture cross section for neutrons of energy close to a certain value. According to the theory as given by Breit and Wigner[3] and extended by Bethe and Placzek,[4] the possibility of resonance arises whenever the compound nucleus, which is formed when a neutron falls upon a nucleus, has an energy level corresponding to positive kinetic energy of the neutron. A neutron of energy close to this value may be captured in the level and the subsequent emission of a γ-ray will leave the neutron bound in the nucleus. The width of the level is determined by the probability of γ-ray emission and by the probability of neutron re-emission.

If only one level predominates, the variation of the capture cross section with energy is given by the dispersion formula of Breit and Wigner,[3]

$$\sigma(E)=\pi\lambda^2\frac{2J+1}{(2s+1)(2i+1)}\frac{\Gamma_n\Gamma_\gamma}{(E-E_R)^2+(\Gamma/2)^2}=\left(\frac{E_R}{E}\right)^{\frac{1}{2}}\frac{\sigma_R(\Gamma/2)^2}{(E-E_R)^2+(\Gamma/2)^2},\quad(1)$$

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[1] Meitner, Hahn, and Strassmann, Zeits. f. Physik **106**, 249 (1937). These authors give 23 min. as the half-life.

[2] Anderson, Fermi, and Szilard, Phys. Rev. **56**, 284 (1939).

[3] G. Breit and E. Wigner, Phys. Rev. **49**, 519 (1936).

[4] H. A. Bethe and G. Placzek, Phys. Rev. **51**, 450 (1937).