Neutron Deficient Radioactive Isotopes of Rhenium

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A study has been made of neutron deficient radioactive isotopes of rhenium. Using the 60-inch Crocker Laboratory cyclotron, bombardments have been made of tantalum with 38, 30, and 19 Mev α-particles, of wolfram with 10-Mev protons, and of rhenium with fast neutrons.

The radiation characteristics of four new radioactive isotopes and of the previously known 50-day Re^{184} isotope have been studied.

I. EXPERIMENTAL

TANTALUM was bombarded on the 60-inch Crocker Laboratory cyclotron as the spectroscopically pure metal foil. After bombardment, the foil was dissolved in hydrofluoric acid, with the addition of the minimum of strong nitric acid. Rhenium carrier was then added, and hydrogen sulfide passed into the boiling solution for fifteen minutes. The rhenium sulfide was dissolved in a solution of hydrogen peroxide and sodium hydroxide, which was then "scavenged" by ferric hydroxide precipitations. The rhenium was then reprecipitated from strong hydrochloric acid solution as the sulfide. While this procedure has been found to give radiochemically pure rhenium, the following additional chemistry was performed in early bombardments: Rhenium was distilled as the volatile chloride or oxide and recovered from the distillate by precipitation as sulfide. The latter was dissolved in a solution of hydrogen peroxide and sodium hydroxide and the rhenium then precipitated from the solution as nitron perrhenate by the well-known procedure. Similar chemical methods were used for neutron bombarded rhenium metal and for proton bombarded wolfram.

The radiation characteristics of radioactive isotopes have been obtained from aluminum and lead absorptions. The general procedure has been to measure the absorption of the gross radiations from a small thin source of active material mounted on thin mica in aluminum absorbers placed directly below the counter window; beryllium foil sufficient to remove electrons completely was then placed above the source and the aluminum absorption of soft electromagnetic radiation measured. A correction for reduction in intensity of the latter by the beryllium was made and a composite curve of total electromagnetic radiation constructed; subtraction of this curve from the measured absorption allowed resolution of electron radiations.

The ratios of the various components of the complex radiations usually observed in orbital electron capturing isotopes were obtained from lead and aluminum absorption data after correction for counting efficiencies, absorption in counter window and other absorbers, etc. The techniques and various assumptions made in interpreting results have been discussed in detail previously.[1,2]

II. RHENIUM ISOTOPES

Four new rhenium activities of half-lives 12.7 hours, 64.0 hours, ~240 days, and 2.2 days have been produced by α-particle bombardment of tantalum and have been allocated respectively to masses 182, 182, 183, and 184. The allocation of a previously reported 50-day rhenium

TABLE I. Production and properties of rhenium isotopes.

Isotope	Type of radiation	Half-life	Energy of radiation in Mev: Particles	Energy of radiation in Mev: γ-rays	Produced by
Re^{182}	K or I.T., e^-, γ	64.0±0.5 hours	0.11, 0.24(abs)	L, K x-rays 0.22, 1.5(abs)	Ta-α-3n W-p-n
Re^{182}	K or I.T., e^-, γ	12.7±0.2 hours	0.16, ~1(abs)	L, K x-rays 0.4, 1.6(abs)	Ta-α-3n W-p-n
Re^{183}	K, e^-, γ	~240 days	0.16(abs)	L, K x-rays 1.0 (abs)	Ta-α-2n W-p-n
Re^{184}	K, e^-, γ	50±2 days	0.2, ~0.7(abs)	L, K x-rays 0.17, 1.0(abs)	Ta-α-n W-p-n Re-n-2n
Re^{184}	K or I.T., e^-, γ	2.2±0.1 days	0.2, ~1(abs)	L, K x-rays	Ta-α-n W-p-n

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[1] G. Wilkinson, Phys. Rev. **75**, 1019 (1949).
[2] G. Wilkinson and H. G. Hicks, Phys. Rev. **75**, 1370 (1949).

activity to mass 184 has also been confirmed. The present data is summarized in Table I.

A. 64.0-hour Re^{182}; 12.7-hour Re^{182}

In the bombardment of tantalum with 38-Mev α-particles, two activities of half-lives 12.7 hours and 64.0 hours were observed, in addition to the longer-lived isotopes. The two short-lived isotopes were observed in the same ratio, but with greatly lowered intensities at 30 Mev, and not at all at 20-Mev bombarding energy. This observation is consistent with production by $\alpha,3n$ reaction. That the 12.7-hour activity cannot be Re^{181} produced by $\alpha,4n$ reaction is shown by the constancy of the 12.7-hour to 64-hour intensity ratio (Table II), and particularly by the production of the 12.7-hour activity by 9-Mev proton bombardment of tungsten where the $p,2n$ reaction does not occur. Further, the yield of the 140-day W^{181},[3] which would be produced as the daughter of any Re^{181} formed in an $\alpha,4n$ reaction of tantalum, agrees with that to be expected from the known deuteron contamination of the α-particle beam (<1 percent) and the measured cross section (5×10^{-2} barn) for the Ta-d-$2n$ reaction.

64.0±0.5-hour Re^{182}.

The gross decay of this activity, which comprises the bulk of the radioactivity in the 38-Mev α-particle bombardments of tantalum, has been followed through twelve half-lives to give a value of 64.0±0.5 hours for the half-life. The radiations consist of electrons, x-ray and hard γ-radiation, all of which have been separately followed and the same half-life obtained. The aluminum absorption of the radiations is shown in Fig. 1. The electromagnetic radiation contribution was obtained from aluminum absorption measurements after removal of the electrons by beryllium absorbers. Two electron components have been resolved, of ranges 16 mg/cm² (~110 kev), and 60 mg/cm² (0.24 Mev). The lead absorption curve shows complex hard electromagnetic radiation of half-thicknesses 137±5 mg/cm² (62 kev), 550 mg/cm² (220 kev) and 13.5 g/cm² lead (1.5 Mev) in addition to the soft radiation half-thickness 21±1 mg/cm² aluminum measured in the aluminum absorption; the two soft components agree well with the expected L and K x-radiation of wolfram or rhenium. No positrons were detected in a very active sample studied on a crude beta-ray spectrometer.

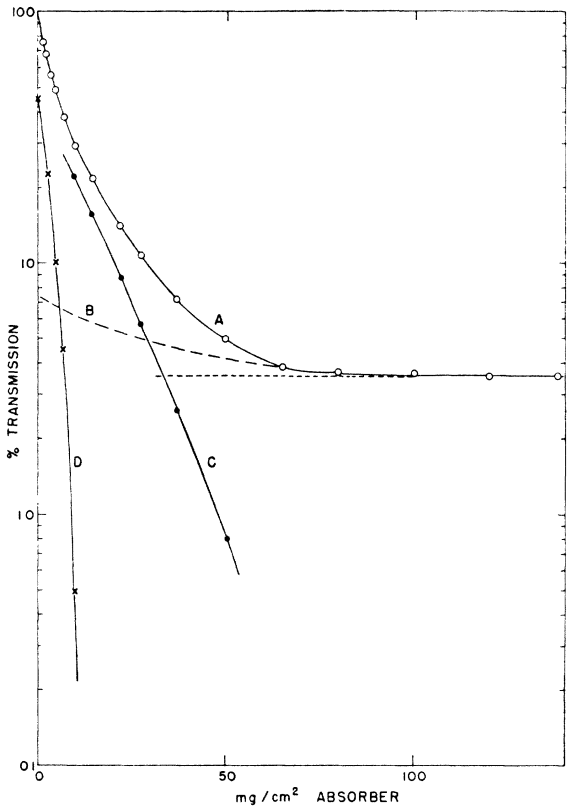

FIG. 1. Aluminum absorption of radiations of 64.0-hour Re^{182} activity (A). The contribution (B) of electromagnetic radiations was obtained from the aluminum absorption curve of this radiation measured after removal of electrons with a 100 mg/cm² beryllium foil; a correction was made for the absorption of the L x-rays in the beryllium and the curve B was constructed by addition of the intensity corrected L X-ray absorption curve to the K x-ray and γ-ray background. The electron lines C and D were resolved after subtraction of the electromagnetic contribution from the curve A.

From the aluminum absorption of the radiations from a "carrierless" rhenium sample mounted on very thin mica backing, the ratios of the two electrons and L x-radiations to the harder electromagnetic radiations were determined; corrections were made for absorption of the soft radiations in the air gap and counter window were made. The ratios of the hard electromagnetic radiations were obtained from the lead absorption. Counting efficiencies assumed were 2.5 percent for L x-radiation, 0.5 percent for K x-radiation and 220 kev γ-rays, and 1.5 percent for the 1.5 Mev γ-ray. Fluorescence yields of 0.8 and 0.5 were assumed for K and L x-rays, respectively. The following ratios were obtained:

110 kev e^-	270 kev e^-	L x-rays	K x-rays	220 kev γ	1.5 Mev γ
= 1.4	: 0.2	: ~2	: 1	: 0.7	: 0.45 .

In view of the complex nature of the radiations it was thought that a shorter-lived lower isomer of the 64-hour activity might be present—possibly the 12.7-hour activity observed in the bombardments along with the 64-hour rhenium. An attempt was made to separate the nuclear isomers. A carrierless solution of rhenium was prepared by removal of tantalum from a bombarded target after solution in nitric and hydro-

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

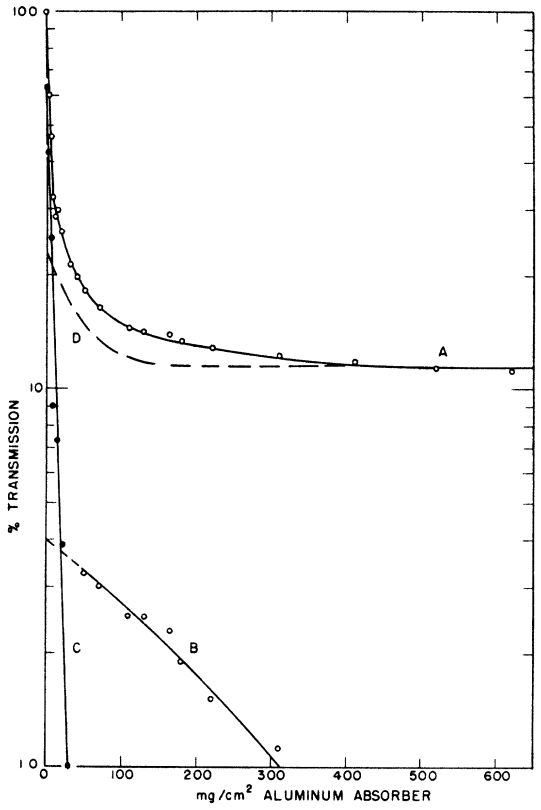

FIG. 2. Aluminum absorption (A) of 12.7-hour Re^{182} activity; electromagnetic contribution (D), ~1 Mev electrons (B), 0.16 Mev electron (C).

fluoric acids, by addition of boric acid followed by ammonia. The filtrate was evaporated with strong nitric acid and the solution, which should contain the radioactive rhenium as perrhenate, was diluted and neutralized with ammonia. If the 64-hour activity produced a lower isomeric daughter, part of the activity of the latter would be expected to remain in the III or IV oxidation states after decay of the parent. Any activity in the III or IV oxidation state could then be removed by scavenging the solution with ferric hydroxide. No evidence for such an isomer was observed, and it may be presumed that the very soft electrons of the 64-hour activity arise from conversion in γ-ray transitions from metastable levels in the daughter nucleus following orbital electron capture. It is somewhat difficult to decide what radiations constitute one disintegration, since x-rays can arise from conversion as well as from L or K orbital electron capture.

The relative yields for production of the 64-hour activity at the various energies of bombarding α-particles (Table II), can, however, be compared by taking the K x-rays as a reference.

12.7-hour Re^{182}

In bombardments of tantalum with α-particles, an activity of 12.7-hour half-life was found to accompany the 64-hour Re^{182}. The activity was found also in the bombardment of wolfram with 10-Mev protons. The decay of the gross and electromagnetic radiations from both Ta+α and W+p bombardments were followed through four and eight half-lives respectively to give a value of 12.7±0.2 hours for the half-life. The radiation characteristics were obtained by resolution of aluminum, beryllium and lead absorption curves, after the contribution of the longer-lived activities at the time of measurement had been subtracted. The aluminum (Fig. 2) and lead absorption curves for the 12.7-hour activity show the radiations to consist of electrons of ranges 35 mg/cm² (160 kev) and ~400 mg/cm² (~1 Mev), and electromagnetic radiations of half-thicknesses 20 mg/cm² aluminum (9.3 kev), 140 mg/cm² lead (62 kev), 3.0 g/cm² lead (400 kev) and 15 g/cm² (1.8 Mev). The two soft electromagnetic radiations correspond well with wolfram or rhenium L and K x-radiation. From the measurements, the following ratios were obtained:

160 kev e^-	~1 Mev e^-	L x-rays	K x-rays	0.4 Mev γ	1.8 Mev γ
= 0.04	: 0.003	: 0.5	: 1	0.25	: 0.15 .

The isotope thus appears to decay by orbital electron capture, although partial decay by isomeric transition to the 64-hour activity is not excluded.

B. 50-day Re^{184}: ~240-day Re^{183}

After decay of the shorter lived periods in Ta+α and W+p bombardments, a complex long-lived activity remains in the rhenium fractions. This activity has been resolved into two components of half-lives 50±2 days and ~240 days, the measurements being carried through several half-lives. The 50-day activity has also been produced by fast neutron bombardment of rhenium. The radiation characteristics obtained by resolution of absorption (Fig. 3) and decay curves are the same in all bombardments, and agree well with those reported by other workers.[4] The approximate ratios of the various radiations of the 50-day activity are:

0.2 Mev e^-	0.7 Mev e^-	L x-rays	K x-rays	~1 Mev γ
0.35	: 0.004	: 0.35	: 1	: 0.2

In the rhenium fraction from Ta+α and W+p bombardments, an activity of ~240 days half-life has been observed after decay of the 50-day period. A pure intense source of this isotope was obtained from aged wolfram exit strips which had received deuterons,

[4] See G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).

protons and α-particles from the 60-inch Crocker Laboratory cyclotron.

The aluminum absorption of an "infinitely thin" sample of the ~240-day activity mounted on thin mica and the lead absorption on a more active sample is shown in Fig. 4. The radiations consists of electrons, total range 35 mg/cm^2 (0.16 Mev) and electromagnetic radiations of half-thicknesses ~21 mg/cm^2 aluminum (9.4 kev), 140 mg/cm^2 lead (62 kev) and 10 g/cm^2 lead (1.0 Mev). On the crude beta-ray spectrograph only a single peak of very soft electrons of ~0.15 Mev energy were observed; no positrons were detected in a very active sample. The following ratios were obtained by the usual procedure.

$$0.16 \text{ Mev } e^-: L \text{ x-rays}: K \text{ x-rays}: 1 \text{ Mev } \gamma$$
$$= \sim 0.4 : \sim 1.1 : 1 : 0.1 .$$

The isotope thus appears to decay by orbital electron capture, with electrons arising from subsequent γ-ray transitions. The isotope has been allocated to mass 183 on the basis of yields in the α-particle bombardments.

C. 2.2-day Re^{184}

In the 19-Mev α-particle bombardment of tantalum, an activity of 2.2 days half-life has been observed in addition to the longer periods. The activity which was not observed in the higher energy bombardments because of the low yields and masking by other activities, is allocated to Re^{184} on the basis of its production by α,n reaction on tantalum. That the activity was not detected by other workers who studied the $n,2n$ reactions in rhenium and p,n reactions in wolfram is not surprising in view of the similarity in half-life to the 98.2-hour Re^{186} which would effectively mask the shorter half-life.

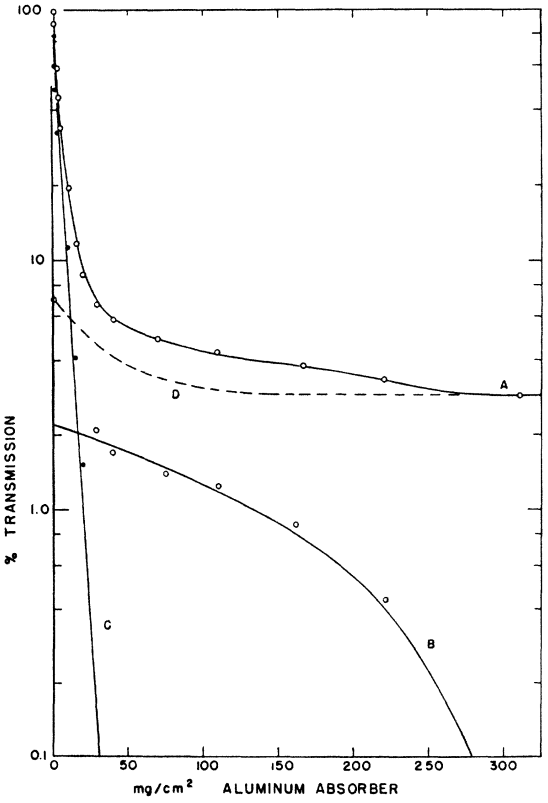

FIG. 3. Aluminum absorption (A) of 50-day Re^{184} activity; K electromagnetic radiation contribution (D), 0.7 Mev electron (B), and 0.2 Mev electron (C).

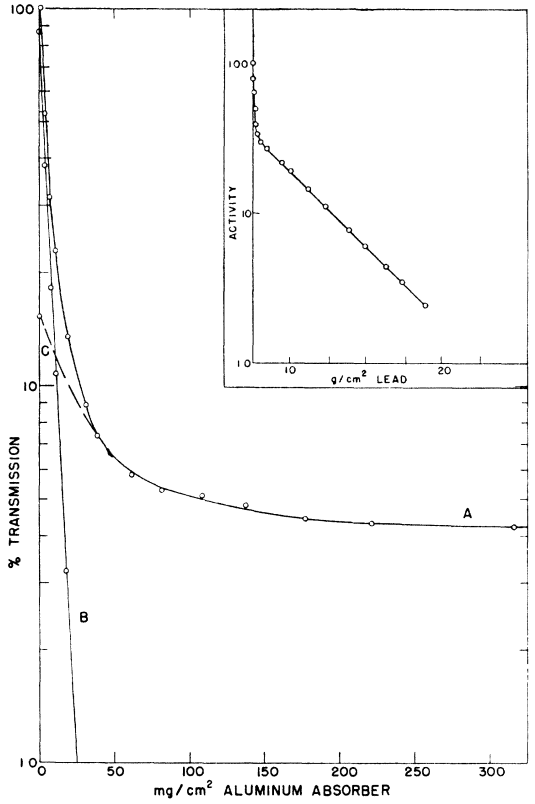

FIG. 4. Aluminum absorption curve (A) of ~240-day Re^{183} activity; curve C shows soft electromagnetic radiation contribution obtained after removal of electrons by beryllium. B is the electron contribution. The lead absorption curve shows K x-rays and 1.0 Mev γ-rays.

The half-life of the activity from Ta+α bombardment is 2.2±0.1 days measured through seven periods. The radiation characteristics were obtained by resolution of aluminum absorption (Fig. 5) and decay curves. The radiations consist of electrons of ranges 50 mg/cm^2 (0.2 Mev) and ~450 mg/cm^2 aluminum (1.1 Mev), soft electromagnetic radiation of half-thickness 21 mg/cm^2 (9.4 kev) and hard electromagnetic radiation. Lead absorptions were not taken because of insufficient activity. The following approximate ratios were calculated from the aluminum absorption:

$$0.2 \text{ Mev } e^-: 1.1 \text{ Mev } e^-: L \text{ x-rays}: K+\gamma\text{-rays}$$
$$0.02 : 0.003 : 0.09 : 1$$

III. DISCUSSION

In order to clarify the status of the five rhenium activities, very active samples were prepared for study

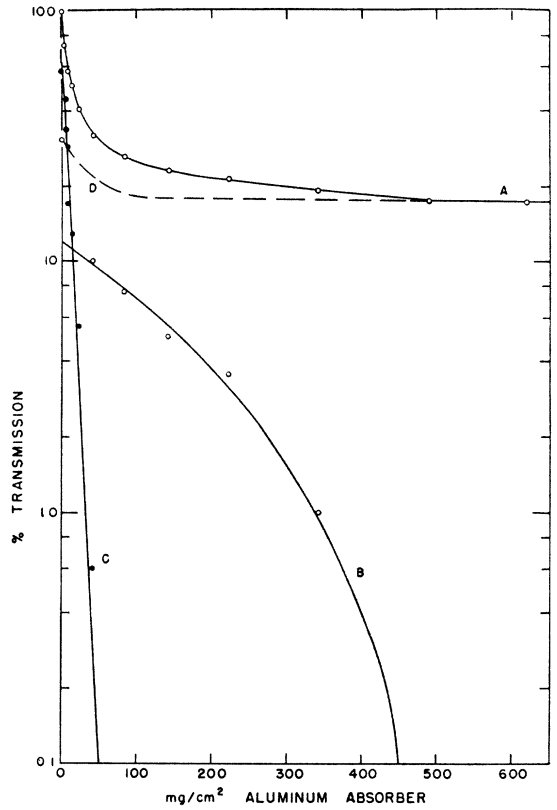

FIG. 5. Aluminum absorption curve (A) of 2.2-day Re^{184} activity; curve D gives the electromagnetic radiation contribution due to L and K x-rays and γ-rays; B and C are the electron contributions.

on a magnetic deflection beta-ray spectrometer, by Professor A. C. Helmholz and Mr. R. W. Hayward. The 64.0-hour and 12.7-hour activities appear to have identical radiations and since the isotopes have been allocated to Re^{182}, it is very probable that they are independent isomers decaying by orbital-electron capture to excited or metastable states of W^{182}. No evidence of beta-particle emission was found in any isotope and the suggestion of W. H. Sullivan[4] that the 50-day Re^{184} decays at least partially by beta-emission is not confirmed. The gamma-ray lines observed in the various isotopes are as follows:

64.0 hours, 12.7 hours: 0.110, 0.127, 0.250 (spec. conv.), 0.222, 0.346 (spec. and spec. conv.).

TABLE II. Relative yields of rhenium isotopes from α-particle bombardment of tantalum.

Activity	Energy of α-particles in Mev 38	30	19	Probable reaction	Mass assignment
12.7 hours	50	7	—	$\alpha,3n$	182
64.0 hours	100	14	—	$\alpha,3n$	182
~240 days	2	7	0.7	$\alpha,2n$	183
50 days	1	1	1	α,n	184
2.2 days	masked	masked	24	α,n	184

50 days: 0.043, 0.205, 0.285 (spec. conv.), 0.159 (spec. and spec. conv.).
~240 days: 0.081, 0.252 (spec. conv.).
2.2 days: 0.043 (spec. conv.), 0.159 (spec. and spec. conv.).

The mass assignments have been made according to measurements on cross sections for production of the isotopes by Ta-α,xn reactions at various bombarding energies. In view of the complex radiations in each case, it is impossible to determine even a rough disintegration scheme by the techniques used, and consequently, the measured K x-radiation is taken as a measure of one disintegration by orbital electron capture. In Table II, the various yields are given relative to those of the 50-day isotope which is unequivocally allocated to mass 184 on the basis of its production by Ta-α,n and Re-$n,2n$ reactions. The variations in yields of the isotopes at α-particle bombarding energies of 38, 30, and 19 Mev, then allow fairly certain mass allocations; the observed yield trends agree with those observed in bombardments of other elements.[2]

The present allocations are in agreement with the formation of the rhenium activities by proton bombardment of wolfram.

V. ACKNOWLEDGMENTS

We wish to thank Professor J. G. Hamilton, Messrs. T. Putnam and B. Rossi, and the crew of the 60-inch Crocker Laboratory cyclotron for their cooperation and assistance in making bombardments. We are grateful to Professor A. C. Helmholtz and Mr. R. W. Hayward for their helpful interest in making measurements on their beta-ray spectrometer. We are indebted to Professors G. T. Seaborg and I. Perlman for their continued encouragement and advice.

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