The Isotopes Os^{185} and Os^{193}

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The half-life of Os^{193} is shown to be 15.0 days, decay being by emission of a beta-particle with range less than 35 milligrams per square centimeter of aluminum (energy not more than 0.165 Mev). A highly converted gamma-ray of about 0.13-Mev energy is probably emitted at each disintegration, producing x-rays in large abundance. Os^{185} decays by orbital capture with a half-life of 97 days. A gamma-ray of 0.75 Mev is probably emitted at each event. The relative cross sections for pile (slow) neutron capture of the isotopes Os^{184}, Os^{190}, and Os^{192} have been estimated as 80:3.5:1, on the basis of measurements on the daughter activities.

I. INTRODUCTION

ZINGG[1] has reported an osmium activity, obtained with neutrons from a radium-beryllium source, of the order of 10-days half-life, in addition to an activity of about 30-hours half-life known from earlier work. Seaborg and Friedlander[2] identified the 32-hour activity they found with cyclotron neutrons as Os^{191}, and reported Os^{193} to emit 0.35-Mev beta-particles with a half-life of 17 days. Goodman and Pool,[3] also using cyclotron neutrons, failed to find the 17-day activity.

The basic fact is that the beta-radiation of Os^{193} is actually very soft. Its aluminum range by the Feather method is not more than 35 milligrams of aluminum, corresponding to a maximum energy of 165 kev. The beta-radiation is accompanied by a soft gamma-ray, and x-rays caused by the internal conversion of the gamma-ray. In all but the thinnest-walled measuring instruments, a significant portion of the activity registered is due to the quantum radiation. The error in the energy assignment by Seaborg and Friedlander is therefore probably due to the influence of the L x-rays. Presumably Goodman and Pool failed to find the activity because of low intensity and heavy-walled measuring equipment.

The nature of the radiation from Os^{193} also misled Seaborg and Friedlander as to its half-life, since together with it we find a long-lived K-capture isotope, whose radiations, blending with those of the 15.0-day isotope, can produce a fictitious 17-day period. Goodman and Pool have described a long-lived isotope from deuterons on rhenium,[3] whose identity with the product of neutron capture we have confirmed, so it is undoubtedly Os^{185}.

II. EXPERIMENTAL

Osmium was activated by several irradiations in the heavy-water moderated chain reactor of the Argonne Laboratory, for 7, 21, and 35 days, respectively. Osmium metal was treated with nitric acid in a distillation apparatus and distilled into an air-cooled condenser. A bubbler trap with concentrated ammonia solution was used as a safety trap, but most of the material was found in the air condensate. This distillation was usually repeated twice more. The osmium was then precipitated by hydrogen sulfide gas, dried, and sealed into a quartz irradiation vessel. At termination of the irradiation the capsule was broken and the osmium distilled again. These purification procedures eliminated small iridium activities. Samples of the final material were precipitated as the sulfide and mounted on mica approximately 3 milligrams per square centimeter surface density. After drying, the samples were made permanent with the aid of a film of zapon approximately 15 micrograms per square centimeter. Experience showed that a post-irradiation decay period of perhaps three weeks had to be allowed to avoid interference by the hard beta-radiation of the 32-hour Os^{191} activity.

With exceptions to be noted below, the activity measurements were made with Geiger counters of the mica end-window type. Windows were usually about 3 milligrams per square centimeter, and

[1] E. Zingg, Helv. Phys. Acta **13**, 219 (1940).
[2] G. T. Seaborg and G. Friedlander, Phys. Rev. **59**, 400 (1941).
[3] L. J. Goodman and M. L. Pool, Phys. Rev. **71**, 288 (1947).

the air path was 1.8 centimeters. The usual counter filling was the standard 9 centimeters pressure of argon, 1 centimeter of alcohol. In the case of absorption curves in which the primary aim was the determination of energies of the quantum components of the radiation, xenon was substituted for the argon. This increased the counting efficiency of the quanta other than the hardest by a factor averaging 3.5.

Preliminary absorption and decay measurements showed that the radiation of the mixed isotopes consisted of four main components: soft beta-radiation, quantum radiation corresponding to L x-rays of elements in the vicinity of osmium, quantum radiation corresponding to K x-rays of the same region, and a gamma-ray of somewhat less than 1-Mev energy. Differential decay measurements showed the beta-radiation to decay most rapidly, the gamma-ray to decay least rapidly, and the intermediate quantum radiation to decay with intermediate rates. This preliminary information immediately gave the picture of a beta-emitter also emitting L x-rays and either a soft gamma with K x-rays or a gamma-ray just too soft to convert in the K shell, and a second isotope emitting a hard gamma-ray together with K and L x-rays. If the long-lived isotope were due to an impurity, such as ruthenium, a harder beta-particle and softer gamma-ray would be found,[4] but a more direct reason for considering the activity due to a K-capture with gamma-radiation is the high ratio of soft quantum to hard quantum counts. Since Os^{185} would be expected to be either a positron emitter or K-capture isotope, it seemed probable that we were not dealing with an impurity.

The decay rate of the soft beta-activity was determined differentially, using an ionization chamber fitted with an FP-54 electrometer and sealed with a window of mica of 5-mg/cm² surface density. A pair of measurements was made at each observation, one of the bare sample and one with 30 mg/cm² of polythene sheet interposed. The difference between the two readings represents the ionization contribution of the soft particles alone. Absorption experiments with additional polythene showed essentially zero absorption of the quantum radiation by the material. The results of these measurements, illustrated in Fig. 1, give a half-life of 15.0 days for Os^{193}. The slight tailing at the end is due to soft particles, conversion electrons, or secondary radiation accompanying the longer-lived isotope.

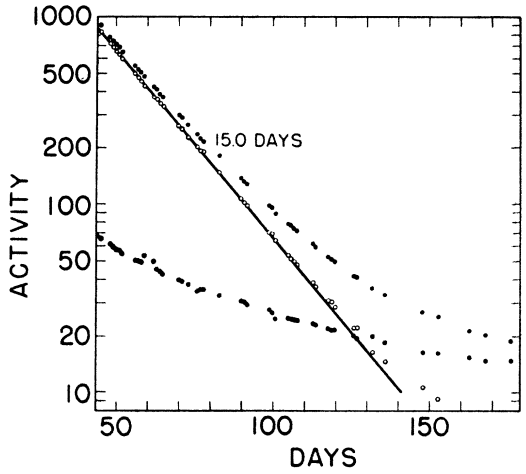

FIG. 1. Half-life determination of Os^{193}. Upper full points, FP-54 values with no added absorber; lower full points, FP-54 values with 30 mg/cm² of polythene over sample; open points, difference values.

Coincidence absorption measurements were also made with a sample of the 7-day irradiation material. These measurements showed a quantum of half-thickness 25–30 mg Al per cm² to be in coincidence with the beta-particle. The half-thickness value corresponds well with that to be expected from the prominent L x-rays of iridium.[5]

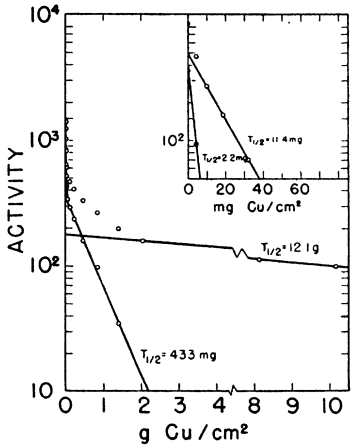

FIG. 2. Copper absorption curve, radiations of Os^{185}. Inset—L x-ray components. Curve taken through 470 mg/cm² of beryllium.

[4] Plutonium Project, J. Am. Chem. Soc. 68, 2411 (1946).

[5] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand and Company, Inc., New York, 1935).

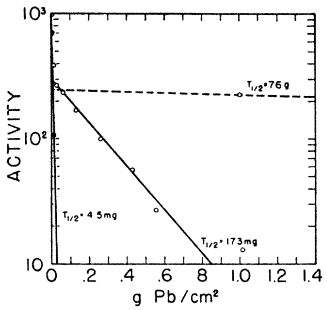

FIG. 3. Lead absorption curve, radiations of Os^{185}. The hard gamma-ray has been resolved out, as indicated by the dashed resolution line. Curve taken through 470 mg/cm² of beryllium.

Coincidences with some harder quanta were also observed, as well as some quantum-quantum coincidences.

A sample of osmium irradiated for 21 days was followed differentially with the Geiger counter. The gamma-radiation decayed with a half-life of 100 days, and the other radiations approached a similar slope. The gross decay and differential decay approximated each other after some 12 half-lives of the Os^{193}; a good average of the slopes is given by a half-life of 97 days. Absorption curves for the material when the short-lived isotope had died out are shown in Figs. 2 and 3. The resolution of the L x-ray components into two by the copper is expected for x-rays of rhenium, with the absorption edge of copper at 1.377A cutting between the L_α and L_β components.[5] Failure to obtain any such resolution of significance in the early stages of decay, when the L x-rays of iridium predominate, is consistent with the rays being all harder than 1.377A. The energy of the ostensible K x-ray component agrees well with that for the K radiation of rhenium. The gamma-ray energies obtained from copper and lead absorption curves do not agree, an anomalously low absorption being found for the copper. Since this type of behavior seems usual, reliance is placed on the value with lead. The gamma-ray absorption half-thickness of 7.6 g/cm² obtained from the lead absorption curves corresponds to an energy value of about 0.75 Mev.[6] The counting ratios (argon) are 1:0.34:1.3 at zero absorber for the gamma, K x-rays, and L x-rays, respectively.

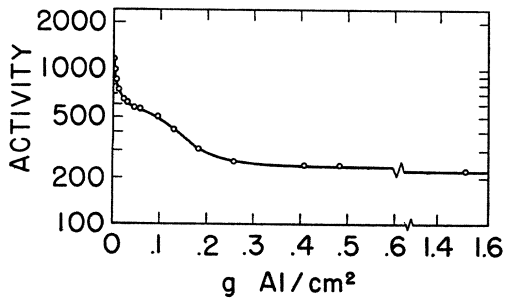

FIG. 4. Aluminum absorption curve of Os^{185}. The change of curvature due to secondary particles is clearly shown.

The meaning of the aluminum curve in the soft region (beryllium gives similar results) is not completely clear (Fig. 4). It seems fairly certain that the flat portion of the particle curve represents coming into equilibrium with secondary radiation due to passage of the gamma-ray through the absorber.[7] The initial rapidly dropping portion of the curve may represent secondary radiation of the softer quanta. The range corresponds to energies too high for auger electrons, and not high enough for conversion electrons of the gamma-ray. The possibility that an impurity such as Ru^{103} might be involved was eliminated by producing Os^{185} through cyclotron bombardment of rhenium with deuterons, using the Washington University cyclotron. The Os^{185} activity so produced was apparently identical with that from pile-irradiated material. Measurements with a Geiger counter in a magnetic field make it certain that the soft radiation in question is particulate.

A picture of the radiations of Os^{193} alone can be obtained by subtracting from an absorption curve for the mixed radiations a profile of the absorption curve for the Os^{185}, with zero-absorber count appropriate to the time the absorption curve of the mixed radiation is taken. This procedure is most accurate in the early stages of the decay, where Os^{193} is most abundant. A curve of this type is illustrated in Fig. 5. Resolution of this curve shows clearly the soft beta-ray, L x-ray component of 28 mg/cm², and hard component. The half-thickness of the hard component (210 mg lead) is equivalent to either 72 kev or 130 kev,[5] since it cuts across an absorption edge. It was not possible to demonstrate critical absorp-

[6] W. Heitler, *The Quantum Theory of Radiation* (Clarendon Press, Oxford, England, 1936).
[7] L. Meitner, Phys. Rev. **63**, 73 (1943).

tion with tantalum, platinum, or gold absorbers (edges from 67.5–81 kev), so the radiation is either not more energetic than 67.5 kev, or falls in the 130-kev range. If the latter is true, there may also be some K x-rays of iridium, *ca.* 65 kev. The difficulty is in theory resolvable by absorption measurements with copper. The behavior of the Os^{185} gamma-ray in copper mentioned above, however, makes copper subtraction curves analogous to Fig. 5 of low usefulness. There does seem to be in these curves a tendency for a considerable hardening of the "tail" as compared to the expected 0.6-gram half-thickness, slopes corresponding to 1–2 grams per square centimeter half-thicknesses being obtained (130 kev should have a 2.5-gram half-thickness), but the reliability is uncertain. An indication may be furnished by the high ratio of L x-ray counts to hard counts, 4:1. This is comparable to that found for the K-capture isotope, and might be evidence of a gamma-ray converting to a high degree in the K shell.[8]

Analysis of the range of the beta-particle by the method of Feather[9] is not satisfactory, due perhaps to conversion electrons; it is possible only to set an upper limit of 35-mg aluminum (168 kev). The apparent relative abundances of the beta-particles, L x-rays, and hard quanta are approximately 150:4:1 in terms of counting rates at zero absorber. No greater precision than 15 percent can be claimed for these values, with the beta-particle abundance being probably least reliable.

Gratitude is expressed to Professor F. N. D. Kurie of Washington University for arranging the cyclotron bombardment of rhenium, and to Mrs. Elaine Novey for technical assistance with some of the FP-54 measurements. Thanks are also due the pile operating crew for their cooperation in the osmium activations. Mr. Frank Wagner assisted with the magnetic measurements.

III. DISCUSSION

From the counting rates of the beta-particles and hard quanta for Os^{193} and the approximate efficiencies of the counting tubes involved, there is roughly one K x-ray quantum or gamma-quantum emitted at each disintegration. From the failure of the copper curves to give any clear resolution of a gamma-ray of the proper energy, it must be presumed that the ray is largely converted in the K shell. A low conversion would also fail to explain readily the high ratio of L to K x-ray counts, which is essentially the same as found for the K-capture isotope, Os^{185}. The total disintegration energy of Os^{193}, taking the beta-energy as 0.16 Mev, and the energy of the gamma-ray as 0.13 Mev, is 0.29 Mev.

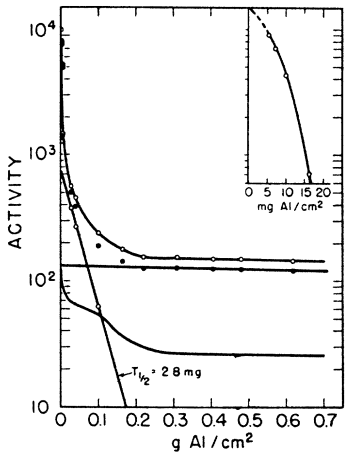

FIG. 5. Aluminum absorption curve of mixed Os^{185} and Os^{193} (upper open points), aluminum absorption curve due to Os^{185} content (bottom curve), and resolution of the Os^{193} curve obtained by difference (full points). Inset—curve for beta-particle component, extrapolated to zero air and counter window absorption.

Similarly, from the counting ratios for Os^{185} there seems to be emission of a gamma-ray of 0.75-Mev energy with each capture of an orbital electron, following a 97-day half-life.

The relative neutron capture cross sections of the parental isotopes, Os^{184}, Os^{190}, and Os^{192}, estimated from the intensities of the daughter activities, are, respectively, 80:3.5:1. The assumption was made of 100 percent counting yield for the beta-particles entering the sensitive volume of the counter, and 1 percent for the 0.75-Mev gamma-ray. The isotopic abundance data of Nier were used.[10]

[8] Since this report was prepared, Cork, Shreffler, and Fowler (Phys. Rev. **72**, 1209 (1947)) have given evidence for conversion electrons from a 129-kev gamma-ray in irradiated osmium.

[9] N. Feather, Proc. Camb. Phil. Soc. **34**, 599 (1938).

[10] A. O. Nier, Phys. Rev. **52**, 887 (1937).