THE
PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, Vol. 134, No. 2B 27 JULY 1964

Decay of Os^{194}†

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(Received 12 March 1964)

Os^{194} has been produced by intense neutron irradiation of osmium metal. The species was conclusively identified from the growth of its 19-h Ir^{194} daughter. The measured half-life is 5.8±0.4 years. The Os^{193} neutron-capture cross section is estimated to be approximately 8 barns for reactor neutrons. The radiations accompanying the decay of chemically purified Os^{194} sources have been investigated by using scintillation and proportional-counter spectrometers. The total β-decay disintegration energy is 97±2 keV. Two electromagnetic transitions, both predominantly $M1$, accompany Os^{194} decay, with energies of 42.8±0.2 keV (abundance=27±5%) and 82.3±1.0 keV (abundance=4.8±1.0×10^{-2}%). It appears that these result from β-decay branches directly populating levels in Ir^{194} at these energies. The decay data indicate spins and parities of either 0- or 1- for these two states. The observed levels are compared with those to be expected for odd-odd nuclei in this mass region.

A LONG-LIVED activity in neutron-irradiated osmium metal has been reported by Lindner[1] which he identified as Os^{194} from its 19-h daughter, Ir^{194}. This result indicated a double neutron-capture process by Os^{192}, the heaviest and most abundant stable isotope of osmium. The radiations of Os^{194} were not characterized, but an approximate half-life of two years was given. The present work reports confirmation of the existence of long-lived Os^{194} and an investigation of its radiations and decay scheme.

EXPERIMENTAL

About 50 mg of natural osmium metal in a quartz ampoule were irradiated for 5 months in the MTR at a thermal neutron flux of 3×10^{14} n/cm^2 sec. Six years later, after the 90-day Os^{185} and other short-lived activities had decayed, the gamma spectrum was examined using a 3-×3-in. NaI(Tl) crystal and 400-channel analyzer. All features of the spectrum at energies over 100 keV could be ascribed to the 19-h Ir^{194} daughter of Os^{194} except for a trace of 5-yr Co^{60} impurity. The decay of the principal gamma peak, due to unresolved 294- and 238-keV gamma rays accompanying Ir^{194} decay, was followed for 10 months in order to estimate the Os^{194} half-life. Decay of a purified Os^{194} source is being followed further on a beta proportional counter.

The ampoule was next opened and portions of the osmium purified by dissolving the metal in concentrated nitric acid and distillation of the resulting volatile osmium tetroxide into 6N sodium hydroxide solution. The gamma spectrum of the purified osmium was recorded immediately after this distillation so that faint Os^{194} radiations were not obscured by the Ir^{194} daughter activity. By recording the growth of Ir^{194} in subsequent gamma spectra, the small contribution of this activity to the initial gamma spectrum could be determined and subtracted. Energy calibration was accomplished using a number of known sources (In^{114m}, Cs^{137}, Tb^{157}, Os^{185}, Au^{195}, and Hg^{203}).

For electron-spectrum measurements, solutions containing no solids other than osmium were desired. The distillate containing the osmium was warmed and OsS_2 precipitated with H_2S. This precipitate was dissolved in conc. nitric acid and the solution warmed with sodium peroxydisulfate to oxidize the osmium to the tetroxide. The solution was diluted with an equal volume

† This work was supported by the U. S. Atomic Energy Commission.
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[1] Manfred Lindner, Phys. Rev. 84, 240 (1951).

of water and the osmium tetroxide extracted into carbon tetrachloride. This organic phase was rinsed with several small portions of water to remove all acid and the osmium finally was back-extracted into conc. NH_3. The final separation of Ir^{194} was found to occur during the last water rinse.

The specific activity (0.2 μCi/mg) was too low to permit study of the electron spectrum with the available magnetic spectrographs and spectrometers. Internal counting using a gas-flow proportional spectrometer was used instead. This counter had a diameter of 4 in. and a length of 12 in. End effects were reduced using "field tubes" as described elsewhere.[2,3] The counter had a removable thick aluminum lining consisting of three cylindrical segments of equal length. A one-inch hole had been made in the central lining segment which coincided with a conducting $\frac{1}{4}$-mil Mylar window located in the aluminum wall of the counter. In this way external x-ray sources (Fe^{55}, Y^{88}, In^{114m}) could be used for calibration. The counting gas employed was 90% argon -10% methane at atmospheric pressure.

The solution containing Os^{194} was either evaporated on the inside surface of a thin aluminum foil placed over the window or else was spread over the entire inside surface of the center lining segment. The latter technique minimizes source self-absorption but could not be used when scintillation measurements were to be made on the same source.

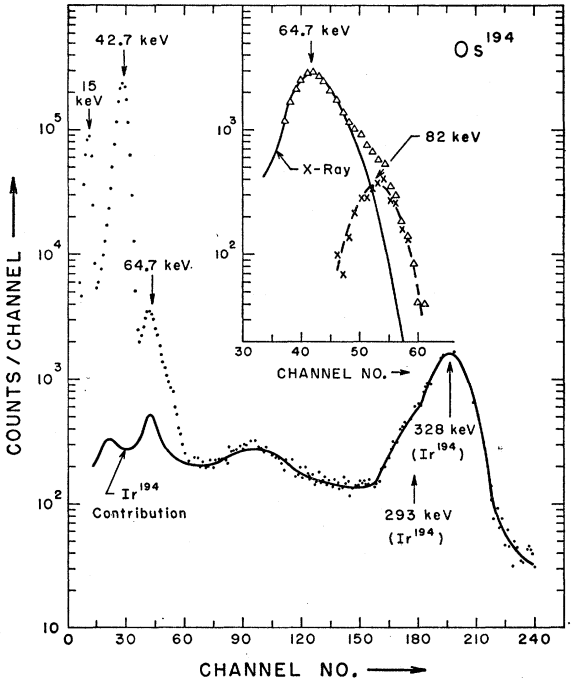

FIG. 1. Sodium iodide scintillation spectrum of freshly separated Os^{194}. The solid curve in the main figure gives the Ir^{194} contribution. Inset: the triangles give the Os^{194} spectrum between 60 and 95 keV; the solid curve gives the K x-ray contribution to the 64.7-keV peak; the crosses show the 82-keV peak (triangles minus the solid curve).

The electron spectrum was also examined with a Pilot B plastic scintillator. For these measurements, Os^{194} activity was deposited on Al foil and placed in direct contact with the plastic.

Beta-gamma coincidence measurements were made between the proportional counter and a 3-×3-in. NaI(Tl) crystal using a resolving time of 2 μsec. The effect of chance coincidences was estimated by delaying one of the detector outputs sufficiently to eliminate true coincidences. For both the singles spectra and the coincidence spectrum, the corrections required for background, Ir^{194} contribution, and chance coincidences were usually relatively small. Near the end points of the beta spectra the correction for Ir^{194} was relatively large.

Source purity was checked in several ways. The chemical purification procedure was improved and repeated without altering the scintillation spectrum. The scintillation spectrum recorded above 100 keV was identical to that of Ir^{194} prepared by the (n,γ) reaction on enriched (98.7%) Ir^{193}. No evidence for 90-day Os^{185} or 1-yr $Ru^{106}-Rh^{106}$ was found by comparison with known sources of these nuclides. Examination of the residue after the first distillation of osmium showed that the nonvolatile impurities, chiefly Co^{60}, were present in the original source in amounts far too small to affect our half-life determination.

RESULTS

In addition to the known transitions in Pt^{194}, the gamma scintillation spectrum of an equilibrium $Os^{194}-Ir^{194}$ source showed a prominent photopeak corresponding to a transition of 42.7±0.3 keV. The spectrum of a freshly distilled Os^{194} source showed this transition as well as a much weaker photopeak at 64.7±0.4 keV (Fig. 1). This energy is in good agreement with that expected for Ir K x rays (weighted average 64.4 keV). The intensity of this peak was approximately eight times the intensity of the Pt K x rays arising from the Ir^{194} present at this time.

The shape of this photopeak, when compared with the shape of the Pt K x-ray photopeak (weighted average 66.3 keV), showed the presence of a high-energy shoulder corresponding to photons of energy 82.3±1.0 keV. The intensity ratios of the 42.7-, 64.7-, and 82.3-keV radiations remained constant within experimental error (100:1.72±0.05:0.183±0.011) when the counting geometry or the source strength were changed by factors of about three. These peaks therefore correspond to actual photon energies, and are not due to chance or coincidence summing effects.

The simplest interpretation of the 64.7- and 82.3-keV radiations is that both result from the same transition, the former being Ir K x-radiation following K con-

[2] D. West, Progr. Nucl. Phys. 3, 18 (1953).
[3] A. L. Cockroft and S. C. Curran, Rev. Sci. Instr. 22, 37 (1951).

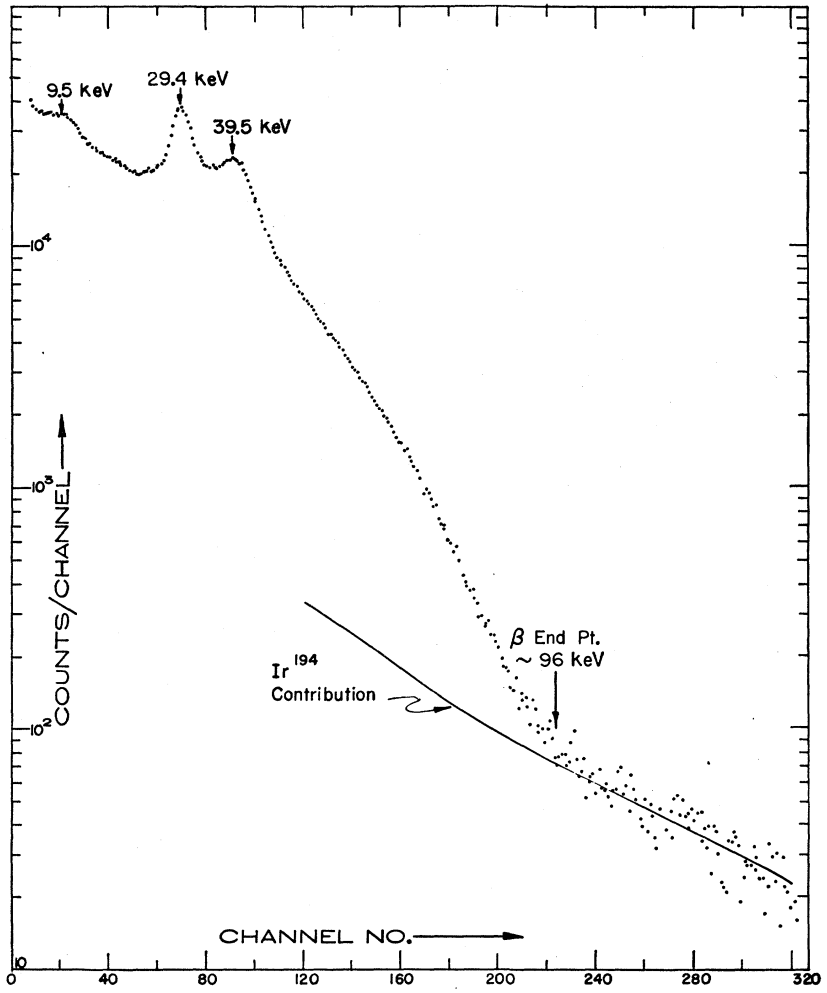

FIG. 2. Proportional counter spectrum of freshly separated Os^{194}. The solid curve gives the Ir^{194} contribution.

version. If so, the intensity ratio gives $\alpha_K = 9.4 \pm 0.5$, in excellent agreement with the theoretical $M1$ value of 9.1 (Rose[4]) or 8.9 (Sliv[5]). Any other magnetic multipole would give α_K at least one order of magnitude higher, while any electric transition would give α_K smaller by amost as great a factor. In particular, for an $E2$ transition, α_K is less than 1.0. The 82.3-keV transition thus appears to be predominantly $M1$, with $E2/M1 < 0.1$.

The pulse-height spectrum from the proportional counter (Fig. 2) shows, in addition to the strong beta continuum, a weak peak at about 9.5 keV, a stronger peak at 29.4 ± 0.2 keV, and a broadened peak at about 39.5 keV. These results are consistent with assignment of the first peak to Ir L x rays, the second peak to the L-conversion electrons of the 42.7-keV transition, and the third peak to M, N, etc., conversion electrons as well as to the sum peak arising from summing L x rays and L conversion electrons. An attempt to assign these peaks to more than one transition calls for additional peaks which are not observed. If the transition is $M1$, as is indicated below, conversion is primarily in the L_{I} shell, and the transition energy is then 42.8 ± 0.2 keV, in excellent agreement with the measured photon energy.

Quantitative interpretation of the intensities observed in the proportional-counter spectra is complicated by summing. Since the source geometry is essentially 50%, sum peaks for coincident radiations are comparable to the individual peaks. The resulting distortion of the beta spectrum makes it difficult to subtract the beta contribution from the conversion peak. In addition, for each event giving a count in the L x-ray peak, there is another in which both the x ray and the conversion electron are detected; this shifts events from the 29.4-keV peak to the 39.5-keV peak. Uncertainties in the corrections for these effects are probably the largest source of error involved in estimating conversion

[4] M. E. Rose, *Internal Conversion Coefficients* (Interscience Publishers, Inc., New York, 1958).

[5] L. A. Sliv and I. M. Band, Coefficients of Internal Conversion of Gamma Radiation: Part I, K Shell—Report 57 ICC K1; and Part II, L Shell—Report 58 ICC L1, Physics Department, University of Illinois, Urbana, Illionois (unpublished).

coefficients and the beta branching ratio for the decay to the 42.8-keV state.

In order to measure the total L-shell conversion coefficient for the 42.8-keV transition, the number of electrons converted in the L shell was estimated from internal counting in the proportional spectrometer, the Os^{194} source was then removed from the proportional counter, and the gamma-ray intensity determined by counting with the NaI(Tl) scintillation spectrometer in standard geometry. The result, $\alpha_L=9.6\pm1.5$, is in good agreement with the theoretical $M1$ values of 9.0 (Rose[4]) and 10.4 (Sliv[5]). The expected values of α_L for $E1$ and $E2$ transtions are 0.58 and 190, respectively. Thus the 42.8-keV transition is essentially pure $M1$, with $E2/M1<0.02$. The branching ratio for decay through the 42.8-keV transition was estimated by comparing the total beta intensity to the sum of the conversion electron intensities and the gamma intensity. The result is $27\pm5\%$. The abundance of the 82.3-keV transition is $(4.8\pm1.0)\times10^{-2}\%$, as determined from its intensity relative to the 42.7-keV gamma ray.

The proportional counter spectrum gave a beta end-point energy of 96 ± 3 keV for the ground-state transition. The Pilot B plastic scintillator spectrum gave 102 ± 5 keV. Agreement is probably satisfactory, in view of the very poor resolution of the latter detector at these low energies.

As expected, the proportional counter pulse-height spectrum in coincidence with the 42.7-keV gamma ray showed only a smooth beta spectrum with no structure, indicating that no other transition is in coincidence with the 42.7-keV gamma. The end-point energy of the coincident beta radiation, as determined from a Fermi-Kurie plot, was 54.5 ± 2 keV, giving a total decay energy of 97 ± 2 keV, in good agreement with the measured ground-state transition end point. The energy of the beta transition populating the 82.3-keV level is then 15 ± 3 keV.

The preliminary Os^{194} half-life measured is 5.8 ± 0.4 years, a value almost three times as great as previously reported.[1] From the Os^{193} and Os^{194} half-lives, the Os^{194} yield, the neutron irradiation parameters, and the Os^{192} neutron cross section, the Os^{193} "reactor neutron" cross section is estimated to be about 8 b. This is much smaller than the value of 200 b previously reported.[1]

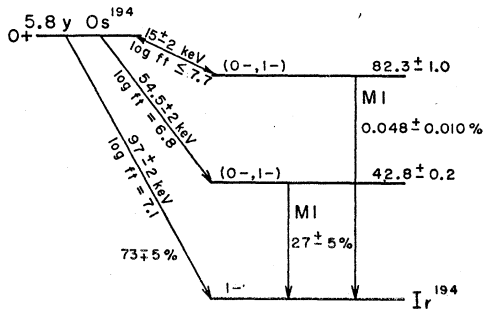

FIG. 3. Decay scheme of Os^{194}.

DISCUSSION

The 42.8-keV transition in iridium 194 must result from decay to the ground state of a level at 42.8 keV directly populated by beta decay (Fig. 3). This is shown by the absence of other electromagnetic transitions in coincidence with 42.8 keV gamma rays, by the fact that the singles spectra show no other transitions with comparable intensity, and by the fact that the sum of the coincident gamma and beta end-point energies equals the ground-state beta transition energy.

The situation is probably similar for the 82.3-keV transition, since the total decay energy is only 97-keV. There is no direct experimental evidence for this, due to the very low intensities involved. Also for this reason, cascade decay of the 82.3-keV state via the 42.8-keV level would not be detected unless such transitions were approximately 10 times as abundant as the observed cross-over transitions.

The spin of Ir^{194} has recently been measured as 1,[6] in agreement with the 1− spin and parity assignment made on the basis of the decay studies of this nuclide.[7] The $M1$ character of the electromagnetic transitions observed in these studies then indicates two excited states of Ir^{194} having spins 0−, 1−, or 2−.

From the measured half-life of Os^{194} and the beta branching ratios, the logft values are calculated to be 7.1, 6.8, and 7.7 for the 97-, 54.5-, and 15-keV beta transitions, respectively. The value for the transition to the 82.3-keV level will be only an upper limit, since the state may decay partially by cascade. Energetic factors alone would favor the cross-over transition by about 8:1. Unless there is some other factor greatly favoring cascade, the logft value obtained by ignoring the cascade should not introduce appreciable error.

The logft values of all three beta transitions fall in the range characteristic of first forbidden transitions. In addition, the values of $\log(W_0^2-1)ft$ for the 97-, 54.5-, and 15-keV transitions are, respectively, 6.7, 6.0, and 6.5. For first forbidden unique transitions, this quantity is generally within one unit of 10. The much lower values found here suggest that all three transitions are nonunique, and the levels involved are either 0− or 1−. It is not possible to distinguish between these two possibilities from the data obtained here.

Iridium 194 lies in the region of transition between the strongly deformed, spheroidal nuclei of the rare-earth elements and the spherical nuclei near lead 208. It is therefore of some interest to attempt a comparison of the properties of the iridium 194 levels observed in

[6] Walter M. Doyle, thesis, University of California Radiation Laboratory Report UCRL 10609, 1963 (unpublished).

[7] K. Way *et al.*, *Nuclear Data Sheets* (Printing and Publishing Offices, National Academy of Sciences—National Research Council, Washington, D. C., 1961.)

these studies with the low-lying nuclear states expected for odd-odd nuclei in this region.

In simple treatments, the ground states of odd-odd nuclei are often interpreted in terms of the coupling of two separate configurations for the neutrons and protons; the configurations used being those inferred from the study of the ground states of the adjacent odd mass nuclei containing the same number of neutrons or protons. The selection of the ground-state term is then usually made using either the Brennan-Bernstein[8,9] or Gallagher-Moszkowski[10] coupling rules for spherical or spheroidal nuclei.

Three classes of excited states may be expected:

(a) intrinsic states resulting from the other terms obtained by recoupling the neutron and proton configurations used for the ground state,

(b) intrinsic states resulting from the coupling of neutron and proton configurations different from those of the ground state,

(c) collective excitations built upon the various intrinsic states.

The ground-state spin and inferred parity of the odd-A iridium isotopes is $\frac{3}{2}+$, while the ground-state spin and parity is $\frac{1}{2}-$ in odd-A isotones containing 117 neutrons (Pt^{195}, Hg^{197}). The ground state of Ir^{194} should then be either $1-$ or $2-$, the remaining term constituting an excited state.

If Ir^{194} is treated as a spherical nucleus and shell-model configurations assigned, these would be derived from the $d_{3/2}$ proton and $p_{1/2}$ neutron orbitals. The Brennan-Bernstein coupling rule $R2$ for spherical nuclei then predicts, incorrectly, that the ground state will be the $2-$ term. This is scarcely surprising, as the Brennan-Bernstein rules are being applied here in a mass region outside the range for which they were originally suggested.

If the Ir^{194} nucleus is treated as spheroidal, the most probable Nilsson orbitals are $[402]\frac{3}{2}+$ for the protons and $[510]\frac{1}{2}-$ for the neutrons.[11] The Gallagher-Moszkowski coupling rule for spheroidal nuclei then predicts, correctly, that the ground state is the $1-$ term.

The $2-$ state obtained by recoupling the ground-state configuration probably could not be observed here, since it could not be populated by the first forbidden, nonunique beta decay of Os^{194} For the same reason, one would not expect to observe any excited states describable as belonging to a rotational sequence based on the ground state. Collective excitations based on the ground state which have predominantly vibrational character and which involve quadrupolar distortions can lead to $1-$ excited states, but a comparison with neighboring even-even nuclei leads one to expect that these would lie at considerably higher energies than do the two excited states of Ir^{194} observed here. It is therefore likely that at least one of these excited states of Ir^{194} is an intrinsic state involving an excited neutron or proton configuration. The remaining excited state could be either a collective state based upon the first or yet another intrinsic state. It may be noted in this connection that the first excited states of Ir^{193} and Pt^{195} have been assigned spins and parities of $\frac{1}{2}+$ and $\frac{3}{2}-$ and are located at 73 and 99 keV.[7] These decay via transitions having considerable $M1$ character. It is possible that one or both of the corresponding configurations contribute to the Ir^{194} excited states observed in these studies.

Two comments should be made concerning approximations introduced in the previous discussion. It is well known that odd group coupling models for odd-odd nuclei, while often predicting correct ground-state spins and parities, cannot accurately reproduce more detailed nuclear properties, for example, the observed magnetic moments. Secondly, a distinction has been made above between intrinsic and collective states. The electromagnetic transitions occurring between the lowest-lying states of the odd A nuclei neighboring iridium 194 show enhanced electric quadrupole transitions attributable to the partially collective character of these states.[7,12] Thus a completely sharp distinction between collective and single-particle states cannot be made in this region.

ACKNOWLEDGMENTS

The authors wish to thank Dr. Ira F. Zartman and the staff of the Materials Testing Reactor for arranging the osmium irradiations. They also wish to thank Dr. Daniel Greenberg of National Distillers Company for providing samples of Ir^{194}.

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[9] A. de-Shalit and J. D. Walecka, Nucl. Phys. **22**, 184 (1961).

[10] C. J. Gallagher, Jr., and S. A. Moszkowski, Phys. Rev. **111**, 1282 (1958).

[11] B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Skrifter **1**, No. 8, 1 (1959).

[12] F. K. McGowan and P. H. Stelson, Phys. Rev. **116**, 154 (1959).