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NEUTRON-DEFICIENT IRIDIUM ISOTOPES

R. M. Diamond and J. M. Hollander

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Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

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ABSTRACT

A study has been made of the isotopes of iridium which are produced from natural rhenium by irradiations with alpha particles in the energy range 25 to 45 Mev. The known iridium isotopes of masses 190, 189, and 188 were observed, and in addition Ir^{187}, Ir^{186}, and Ir^{185} were identified by means of the threshold energy for their production. Analyses are based upon scintillation spectra and conversion electron spectra taken with high-resolution permanent-magnet spectrographs.

The interesting situation is encountered in which the three isotopes Ir^{187}, Ir^{186}, and Ir^{185} all have very similar half-lives; that of Ir^{187} is 13 hours, Ir^{186} 16 hours and Ir^{185} 15 hours. A preliminary study of the radiations from the odd mass isotopes has been made, and also some new transitions in Ir^{186}, Ir^{188}, and Ir^{190} are reported.

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INTRODUCTION

In the region of atomic number between 74 and 78 significant changes in the order and the nature of nuclear energy levels may be expected. The preceding elements, $64 < Z < 74$, have been characterized by their rotational structure,[1] and in elements as heavy as tungsten, $Z = 74$, well-defined rotational bands appear and have been extensively studied.[2] At $Z \geq 78$ the behavior of nuclear levels has been described best in terms of the single-particle shell model. It is of interest, therefore, to obtain experimental information about the levels of nuclei in the intermediate region, for which there is at present little theoretical description. The isotopes of osmium, $Z = 76$, would be good examples, and their levels can be populated by the electron-capture decay of light iridium nuclei or by the beta decay of heavy rhenium nuclei. We here report on the synthesis of two new neutron-deficient isotopes of iridium, Ir^{185} and Ir^{187}, and on the results of our preliminary study of the radiations from these isotopes and from Ir^{189}. Some additional data are given concerning transitions in Os^{186}, Os^{188}, and Os^{190} following electron capture decay of the corresponding iridium nuclei.

EXPERIMENTAL PROCEDURE

The light iridium isotopes were produced by (α,xn) reactions from irradiation in the Crocker 60-inch cyclotron of "thick" targets of rhenium metal foil of natural isotopic abundance (37% Re^{185}, 63% Re^{187}). Alpha particles of

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† On leave from Cornell University, Department of Chemistry, Ithaca, N. Y.

25 to 27, 33 to 34, and 45 Mev energy were used. Targets were either 5-mil foils bombarded singly or 1-mil foils bombarded in stack form. In this way the relative yields of the various isotopes could be varied and, in particular, the production of Ir^{185} by $Re^{185}(\alpha,4n)$ and of Ir^{186} by $Re^{185}(\alpha,3n)$ could be selectively eliminated.

After irradiation, carrier-free iridium fractions were obtained from the target foils by one of the two following procedures:

1. The rhenium foils were dissolved in concentrated nitric acid and the solution boiled to near-dryness to volatilize osmium as OsO_4. The small volume of residue was made about 4 M in HNO_3 and 1 M in HCl and adsorbed onto a Dowex-1 anion exchange column which had been pretreated with a solution 4 M in HNO_3 and 1 M in HCl. The column was then rinsed with more of this acid mixture in order to remove rhenium. Afterwards, the iridium was stripped from the column with a solution 4 M in HCl and 1 M in NH_2OH. Concentrated nitric acid was added and the mixture was boiled in order to destroy the hydroxylamine and to reduce the volume.

2. Same initial treatment to remove the osmium. After evaporation to a small volume, the solution was diluted to about 0.5 N HNO_3 and passed through a Dowex-50 cation exchange column; under these conditions iridium is adsorbed on the resin but rhenium can be washed through with several column volumes of 0.15 M HNO_3. The iridium was then eluted from the column with 8 M HNO_3 and boiled down to a small volume.

The following techniques were used to observe the radiations of the iridium fractions. Gross decay curves of the samples were obtained with proportional counters. Gamma-ray spectra were studied with a scintillation spectrometer employing a sodium iodide crystal, 1-1/2 in. in diameter by 1 in. high, coupled to a Penco 100-channel pulse-height analyzer. With this crystal the energy resolution of the 662 kev gamma ray of Cs^{137} was ~10%. Electron spectra, upon which much of the analysis is based, were obtained primarily with the high-resolution (~0.1%) permanent-magnet spectrographs of the type described by Smith and Hollander.[3] Use was made of six different spectrographs, with field strengths 50, 100, 125, 150, 200, and 340 gauss, respectively. Spectrograph sources were prepared by electrolytic deposition of iridium from

dilute NH_4HSO_4 solutions onto 10-mil platinum wires. Some electron spectra in the higher energy region were also obtained with a lower resolution (~1%) lens spectrometer.

EXPERIMENTAL RESULTS

Iridium-190 and Iridium-189

Two isomers of Ir^{190} are known, the 11 day ground state discovered by Goodman and Pool,[4] and the 3 hour isomer found by Chu.[5] On the basis of studies of their gamma radiation, Aten, deFeyfer, Sterk, and Wapstra,[6] have given a decay scheme for these isomers; that of Ir^{190m}, which decays to the 10 minute Os^{190m} isomer, has been elaborated further by Scharff-Goldhaber, Alburger, Harbottle, and McKeown.[7,8]

Ir^{189}, which has an 11-day half-life similar to that of Ir^{190}, was discovered by Smith and Hollander[9] as the daughter of 11-hour Pt^{189}. They observed from scintillation spectroscopy that its principal electromagnetic radiation is a 245-kev photon; higher energy photons are less intense by at least a factor of ten. Recently, Scharff-Goldhaber *et al.*[7] have shown that a 5.7 hour isomer of Os^{189} is produced by the decay of 11-day Ir^{189}.

In this work, we have attempted to distinguish the photons of 11-day Ir^{190} from those of 11-day Ir^{189} by comparisons of the relative photon intensities in the scintillation spectrum as a function of bombarding energy. At the lowest energy (25 to 27 Mev) the yield ratio Ir^{190}/Ir^{189} is expected to be higher by a factor of six or seven than at 33 to 34 Mev although the total cross sections will be lower for both products. We find, in fact, that the intensity ratio of the prominent 187 kev photon of Ir^{190} to that of the prominent 245 kev photon of Ir^{189} does increase by a factor of ~7 in the above bombarding energy interval.

The low-energy transitions (30 to 90 kev) cannot be assigned in the above manner because they are not resolved in the scintillation spectrum. In the conversion electron spectrum the lines of the low energy transitions are much more intense than those of the 245-kev transition, hence they are assigned to Ir^{189}; the decay of Ir^{190} is fairly well understood and it is not expected that intense low-energy transitions other than the 39 kev isomeric transition of Os^{190m} will be found in its decay.

In Table I we present a brief summary of the information we have obtained about the transitions of 11-day Ir^{190} and 11-day Ir^{189}. Transition energies are given, and multipolarity assignments are made from K/L ratios and L and M subshell conversion ratios. Figure 1 shows the levels of Os^{190} as presently known. The main level order is from the work of Aten et al.[6] and the isomeric state is from that of Scharff-Goldhaber et al;[8] the second 2+ state and the energies of the lower levels are from this work. The energy of the transition from the 4+ to the 2+ state in Os^{190} has been given as 356 kev by Aten et al.[6] and as 359 by Scharff-Goldhaber et al.[8] We observe this transition at 361.2 kev, and in addition a transition of 371.4 kev is seen in the electron spectrum. We suggest that the latter transition be assigned as the 2+ ——> 2+ transition and that the 558 kev transition is the crossover to the ground state. The sum 371.4 + 186.7 = 558.1 is in agreement with the energy of the crossover transition. Also, the energy of this second 2+ state agrees well with the value obtained by extrapolation from the Os^{186} and Os^{188} energies (see Fig. 3) and with the value of 0.57 Mev found from the Coulomb excitation[10] of Os^{190}.

Transitions of 407, ~525, 568, and ~610 kev are assigned to Ir^{190} decay, but their positions in the level scheme remain unknown.

We have assigned most of the low-energy transitions to the decay of Ir^{189}. Because of the preliminary character of the data, we have not attempted to construct a level scheme for Os^{189}. However, certain sum relationships are suggestive and are given in Table II. The 30.8 kev transition seen here from Ir^{189} decay is assigned on the basis of its high L_{III} and M_{III} conversion as an M3; this is without doubt the same transition as that observed by Scharff-Goldhaber et al.[8] in the decay of the 5.7 hour Os^{189} isomer. The transition is relatively weak in the Ir^{189} spectrum, so that it appears that the isomeric state is not heavily populated in Ir^{189} decay. The most intense transition is the one at 69.5 kev, assigned as a mixed M1-E2; this probably proceeds to the ground state and hence defines the second excited state of Os^{189} to be 69.5 kev. From the sum relationships other states may be postulated to lie at 219.4, 255.7, 275.8, and 314.9 kev; however the transition orders must be verified by intensity and coincidence studies before these states can be considered as definite.

Table I

Transitions from Decay of Ir^{189} and Ir^{190}

Transition energy (kev)	Electron lines seen[a]	Photon Observed (scint spect)	Multipole Order	Mass Assignment
30.8	$L_{III}(22)$ M_I $M_{III}(6)$ N		M3	189
33.3?	L_{II} L_{III} M		E2	
36.2	$L_I(35)$ L_{II} $M_I(13)$ N_I		M1	189
56.4 or 117.3	L_I M_I or K L_{II} L_{III}			
59.0	$L_I(55)$ $L_{II}(5)$ M_I		M1	189
69.5	$L_I(\geq 100)$ $L_{II}(\geq 100)$ $L_{III}(\geq 100)$ M_I $M_{II}(35)$ $M_{III}(35)$ N(25) O		M1-E2	189
95.2	K $L_I(3)$ L_{II} L_{III} M N		M1	189
135.8	K $L_I(2)$ L_{II}		M1	
147.1	K			
149.9	K			
164.0	K			
185.9	K			
186.7	K L_{II} L_{III} M_{II} M_{III} N	yes	E2	190
188.6	K			
206.2	K			
216.6	K L_I L_{II}			
219.3	K L_I			
233.6	K L_I L_{II}			
245.0	K(15) L_I $L_{II}(6)$ $L_{III}(6)$ M_{II} M_{III} N	yes	E2	189

(continued)

Table I (continued)

Transition energy (kev)	Electron lines seen[a]	Photon Observed (scint spect)	Multipole Order	Mass Assignment
275.8	K L_I			
361.2	K			
371.4	K	yes		190
407	K			190
520-530				190
558	K	yes		190
568	K			190
600-620		yes		190
~700		yes		
760	K			
~800		yes		
~1000		yes		

[a]Approximate intensity values of the stronger electron lines, given in parentheses following the shell designations, are for identification only and not for purposes of analysis.

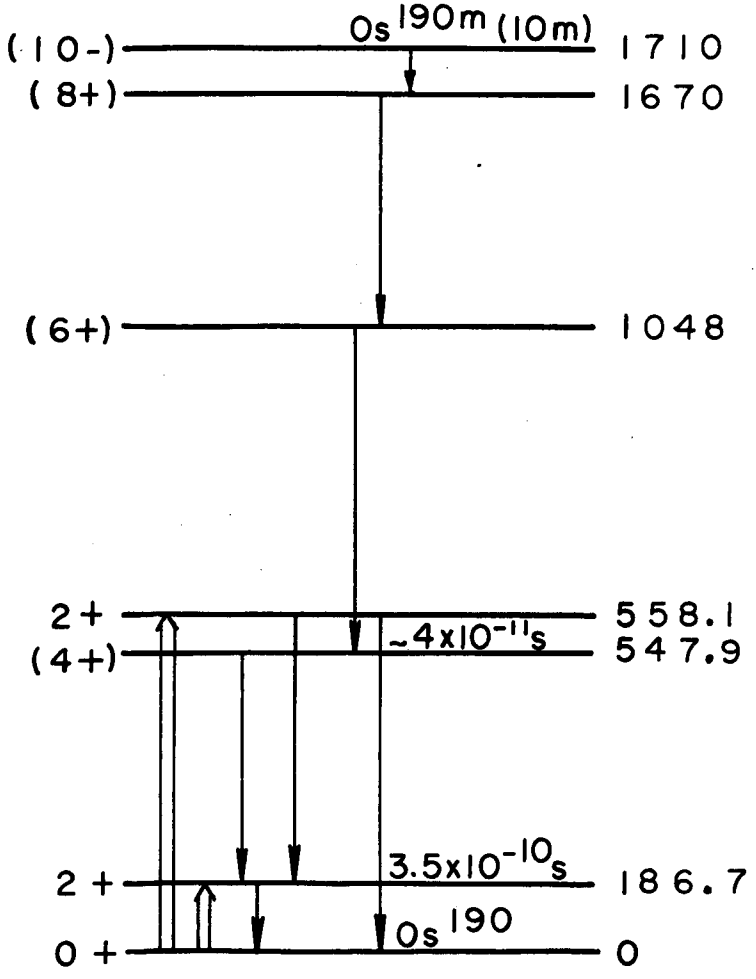

Fig. 1. Os^{190} level scheme.

Table II

Ir^{189} Transition Sums

(a)	30.8 + 188.6	=	219.4
	69.5 + 149.9	=	219.4
	Crossover	=	219.3
(b)	95.2 + 149.9	=	245.1
	59.0 + 185.9	=	244.9
	Crossover	=	245.0
(c)	36.2 + 149.9	=	186.1
	Crossover	=	185.9
(d)	69.5 + 206.2	=	275.7
	Crossover	=	275.8
(e)	56.4 + 149.9	=	206.3
	Crossover	=	206.2
(f)	36.2 + 59.0	=	95.2
	Crossover	=	95.2
(g)	245.0 + 69.5	=	314.5
	219.3 + 95.2	=	314.5

Iridium-188

A 41 hour activity in iridium was assigned to mass 188 by Chu[5] on the basis of its production by an α,3n reaction on enriched Re^{187}. Smith and Hollander confirmed this assignment[9] and observed gamma rays from Ir^{188} at approximately 150, 475, and 625 kev; these transitions appear to be the same as the lowest energy transitions found[11] in the beta-decay of Re^{188}.

The 41 hour half-life of Ir^{188} is sufficiently different from that of the other iridium isotopes produced in this work to allow its prominent gamma transitions to be identified from spectra taken at timed intervals on the scintillation spectrometer and with the electron spectrographs. These are listed in Table III. The principal transitions observed at 155.0, 478.5, and 633.5 kev are those mentioned above. The 825- (or 830-) kev gamma ray is possibly the 828 kev transition observed from the β^- decay of Re^{188}.

The 323.0 and 1212 kev transitions, observed here in both the scintillation and electron spectra, have not been reported from Re^{188} decay. It is tempting to postulate that the 323 kev transition is the one between the 4+ and 2+ states of the ground-state rotational band because the energy so obtained for the 4+ state lies reasonably between the corresponding 4+ states of Os^{186} and Os^{190}. The plausibility of this assignment seems to be marred by the fact that the postulated 478-kev level has bot been seen from Re^{188} decay. It is possible, however, that the spin of Re^{188} is low while that of Ir^{188} is high. Note that the higher energy gamma rays observed in this study of Ir^{188} do not correspond to those observed from Re^{188} decay, which is an indication that there are other significant differences in the final states populated by the decay of these two nuclides.

Figure 2 shows the lower levels of Os^{188}, and Fig. 3 gives a correlation diagram of the energies of the low-lying states in the osmium isotopes. (We have listed the 691-kev state of Os^{192} as 4+ even though there is some experimental evidence against this assignment.[12]) As shown in the correlation diagram the energies of the rotational states (2+, 4+, and 6+) are increasing with mass number because these nuclei lie just beyond the "rotational" region and the moments of inertia are decreasing rapidly as the next closed shell is approached. But at the same time the energy of the vibrational (Goldhaber-Weneser) 2+ state is dropping as the nucleus becomes "softer" toward this type of excitation.

Table III

Transition Energy (kev)	Electron Lines Seen[a]	Photon Observed (Scint spect)
155.0	K(~350) L_I(70) L_{II}(320) L_{III}(250) M_{II} M_{III} N O	yes
323.0	K(~12) L	yes
478.5	K(~30) L M	yes
633.5	K L	yes
825	K	yes
830	K	
1212	K	yes
1330		yes
1450		yes (complex)
1600		yes
1710		yes (complex)
1940		yes
2050		yes
2180		yes

[a] See note "a" of Table I

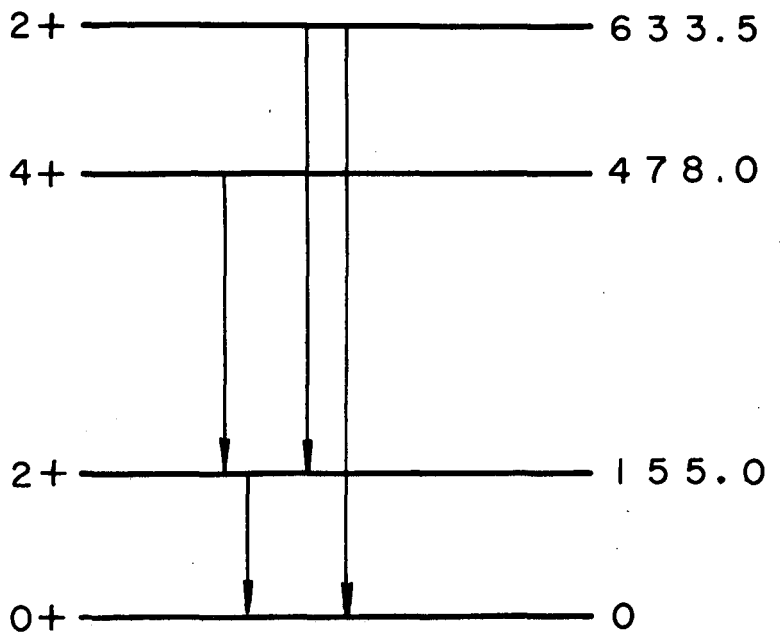

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Fig. 2. Os^{188} level scheme.

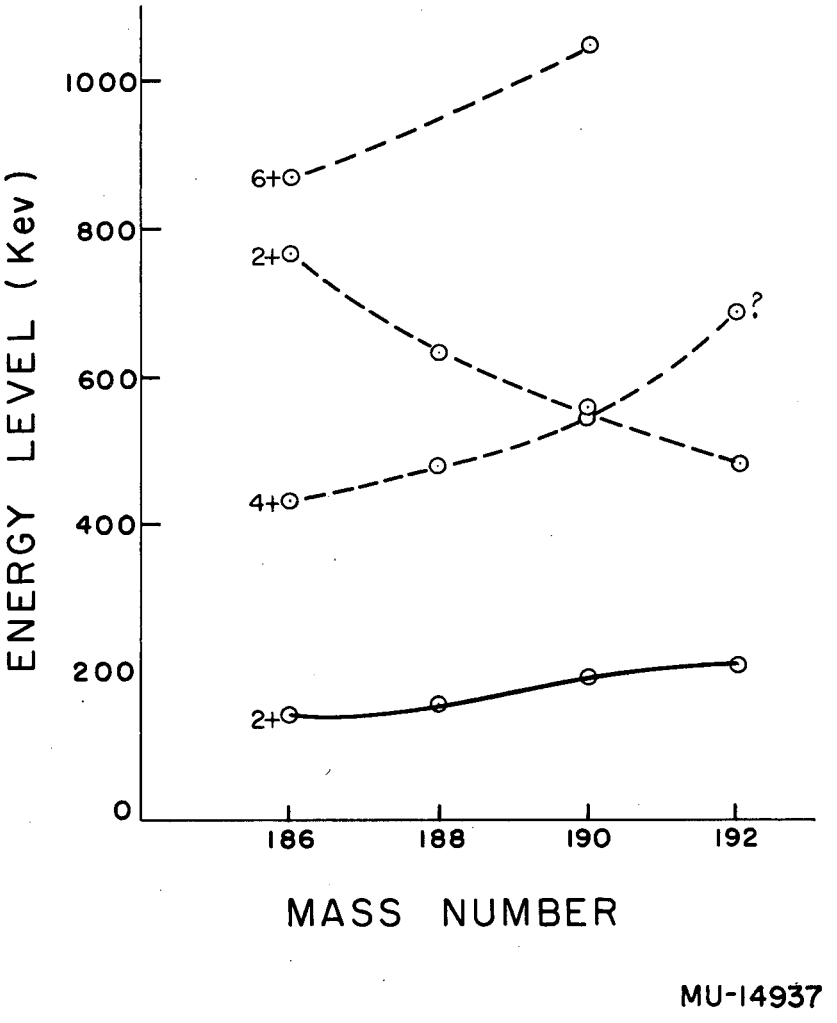

Fig. 3. Correlation diagram of energy levels of even-mass osmium nuclei.

Iridium-187

In irradiations of rhenium foils at an initial alpha particle energy of 25 to 27 Mev, i.e., below the threshold for the production of Ir^{185} and Ir^{186}, several short-lived conversion electron lines and gamma rays were observed in the separated iridium fraction. By comparing the intensities of the more prominent of these electron lines in a series of timed exposures in the electron spectrographs with lines of known intensities in a group of standard plates, we obtained a value of 13 ± 3 hours for the half-life of this new activity. Actually, the magnitude of the half-life relative to those of Ir^{185} and Ir^{186} determined in the same manner can be measured more accurately than the absolute value, and the half-life is definitely shorter by 2 to 3 hours than those of Ir^{185} and Ir^{186}.

The new activity might possibly be due to an isomer in Ir^{188}, Ir^{189}, Ir^{190}, or even in Os^{188}, Os^{189}, Os^{190}, but has been assigned to Ir^{187} for the following reasons. It cannot be assigned to an osmium isomer because the half-life is shorter than that of the parent iridium isotope and yet no growth and subsequent decay with the parent half-life was observed. It is unlikely to be an isomer in Ir^{188} or Ir^{190} because the resulting even-even Os^{188} or Os^{190} nuclei formed would not be expected to have many low-lying levels (and no such levels have been seen in Ir^{188} or Ir^{190} decay, see above), whereas the most intense transitions of the decay under consideration are of low energy. Furthermore, no known transitions in Os^{188} or Os^{190} are observed as would be expected, and as are observed in the decay of the 3 hour Ir^{190m}. The number of intense low energy transitions strongly suggest an odd mass iridium isotope, but an isomer of Ir^{189} is unlikely since its decay into excited levels in Os^{189} would almost surely cascade into at least one of the several low-lying levels observed from the decay of 11-day Ir^{189}, and such transitions are not observed. Thus Ir^{187} is the only remaining assignment for this 13-hour activity. Consistent also with this assignment were the variations in the relative yields of prominent transitions of Ir^{186}, Ir^{188}, and the 13-hour activity as a function of bombarding alpha-particle energy, as compared with the results obtained by John[13] on α,xn cross-sections of lead (Z = 82).

The transitions assigned to Ir^{187} decay, as well as those of Ir^{186} and Ir^{185}, are listed in Table IV, as are also some weak and some higher energy transitions which it was not possible to assign any more definitely than to these shorter lived isotopes.

Table IV

Transitions from Decays of Ir^{185}, Ir^{186}, and Ir^{187}

Transition energy (kev)	Electron lines seen	Photon observed	Multipole order	Mass assignment
25.5	L_I M_I N		M1	187
37.4	L_I L_{II} L_{III} M_I N_I O		M1-E2	185
59.9	L_I L_{II} M_I N		M1	185
(64.5)	L_I			187
65.2	L_{II} L_{III} M_{II} M_{III} N O		E2	187
70.8	L_I L_{II} $M_{I,II}$ N			186 or 187
74.2	L_I L_{II} M_I N		M1	187
75.0	L_{II} L_{III} M_{II} M_{III} N		E2	187
90.5	K L_I L_{II}			187
97.3	L_I L_{II} L_{III} M_{II} M_{III} N O		E2-(M1)	185
100.8	K L_I L_{II} M_I		M1	185
104.4	K L_I			185
113.1	K L_I L_{II} L_{III} M		M1-E2	187
(115.6)	K			
137.2	K L_I L_{II} L_{III} $M_{I,II}$ M_{III} N O	yes	E2	186
149.9 or 89.0	K or L_I			Possibly 185
153.6 or 92.7	K or L_I M_I			185
(162.9)	K			186 or 187
(163.6)	K			186 or 187

(continued)

Table IV (continued)

Transition energy (kev)	Electron Lines Seen	Photon observed	Multipole order	Mass assignment
177.6	K L_I L_{II} L_{III}	yes	M1	187
187.5	K L_I L_{II} L_{III} M	yes	M1-E2	187
252.6	K			
254.4	K L_I			185
296.9	K L_I L_{II} L_{III} $M_{II,III}$ N		E2	186
299.9	K			probably 185 or 186
303.1	K			"
306.1	K			
309.7	K			
314.0	K $L_{II,III}$			187
351.8	K			probably 185 or 186
401.0	K $L_{I,II}$			187
421.6	K			185-186
427.2	K L_I	yes		187
434.8	K $L_{I,II}$ L_{III} M N	yes	E2	186
442.2	K			probably 187
493	K $L_{I,II}$	yes		probably 186 or 187
502	K	yes		probably 186 or 187
577	K L	yes		
586	K L	yes		
612	K L M	yes		probably 186 or 187
623	K	yes		
631	K	yes		probably 186

(continued)

Table IV (continued)

Transition energy (kev)	Electron lines seen	Photon observed	Multipole order	Mass assignment
769	K	yes		probably 186
775	K			
842	K			
914	K L	yes		probably 187
923	K			186 or 187
979	K	yes		probably 187
990	K			
1099	K			

Iridium-186

Chu,[5] in 1950, synthesized an 11.8 hour iridium activity which he assigned to Ir^{187} on the basis of excitation function experiments. A 14-hour iridium activity, obtained by Smith and Hollander[9] as the daughter of a 2.5 hour platinum activity, was assigned to either mass 186 or 187, with 187 preferred. The principal radiations were reported to be transitions of 135, 300, and 435 kev. Scharff-Goldhaber et al.[8] have made a definite assignment of this 14-hour iridium activity to mass 186 on the basis of the systematics of levels of the even-mass osmium isotopes and have observed transitions at 137, 297, 434, 625, and 773 kev, as well as several at higher energies. From K to L conversion ratios the multipolarities of the three lowest energy transitions were assigned as E2 and were interpreted as the cascade from the third rotational level (6+ ——> 4+ ——> 2+ ——> 0+); the energy ratios are close to those calculated from the rotational formula,

$$E_i = \frac{\hbar^2}{2\Im} I(I+1) .$$

In this work we have established the transitions that belong to mass 186 by an observation of the particular electron lines and photons that disappear from the complex spectrum when the alpha particle bombarding energy is lowered from 33-34 Mev (threshold for production of Ir^{185}) to 25-27 Mev (threshold for production of Ir^{186}); such transitions are assigned to Ir^{186}. The assignment in this manner of the 137.2, 296.9, and 434.8 kev gamma rays to Ir^{186} is in agreement with the work of Scharff-Goldhaber et al. These, together with the 631 and 769 kev transitions in Os^{186} previously known[14] from the β^- decay of Re^{186} and also observed in this work, allow the scheme shown in Fig. 4 to be constructed for the lower levels of Os^{186}. Again, by comparing the intensities of the most prominent of these conversion lines in a series of timed spectrograph exposures with lines of known intensity on standard plates, we have obtained a value for the half-life of Ir^{186} of 16 ± 3 hours. The conversion lines and gamma rays definitely assigned to Ir^{186} are listed in Table IV. Some of the higher-energy unassigned transitions given there may also belong to Ir^{186}.

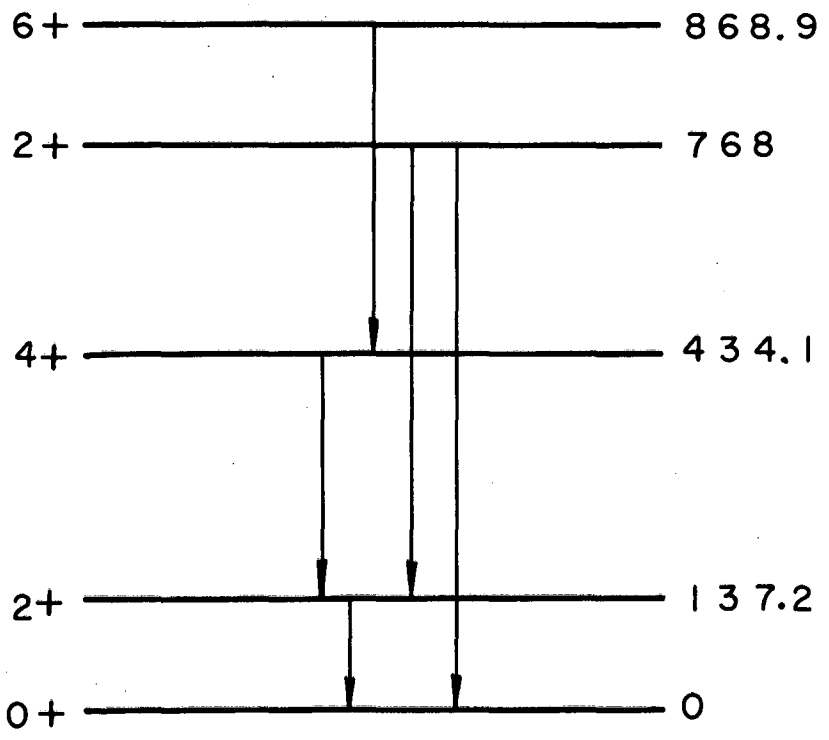

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Fig. 4. Os^{186} level scheme.

Iridium-185

With alpha particles of 45 Mev initial energy Ir^{185} is produced from rhenium by the reaction $Re^{185}(\alpha,4n)Ir^{185}$; there are simultaneously produced the heavier isotopes of iridium from lower order reactions. The Ir^{185} radiations can be distinguished, and the mass assignment made, by repeating the bombardment with alpha particles of initial energy lower than the threshold for the $(\alpha,4n)$ reaction, i.e., 33-34 Mev, and observing which conversion lines and photons are eliminated at the lower energy irradiations. The conversion lines and photons so found and assigned to Ir^{185} are listed in Table IV.

As for Ir^{186} and Ir^{187} we have determined the half life of Ir^{185} by visual comparisons of electron line intensities with those on standard plates; the value of the half-life so obtained is 15 ± 3 hours, about 1 hour less than that of Ir^{186} and 2 hours greater than that of Ir^{187}. All the lines assigned to Ir^{185} show a half-life of this magnitude.

Confirmation of the production of Ir^{185} in the 45 Mev alpha particle bombardments was obtained by volatilizing the 95 day Os^{185} daughter at timed intervals from a target foil dissolved in concentrated nitric acid. The value of the parent half-life obtained in this way, 14 to 20 hours, is quite rough but is consistent with the value obtained directly from decay of the conversion lines. The "milked" osmium fractions were examined with the scintillation spectrometer and it was verified that they had the characteristic half-life and gamma ray spectrum of Os^{185}. The prominent gamma rays of Os^{185} also appeared in the iridium fractions from all the 45 Mev bombardments but were not in evidence in the bombardments done at 33 Mev or lower energy.

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