Title

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ABSTRACT

The new neutron-deficient iridium isotopes Ir^{182}, Ir^{183}, and Ir^{184} have been produced by irradiations with the Berkeley heavy-ion linear accelerator. By means of timed chemical separations, NaI scintillation spectrometers, and proportional counters, half lives have been determined as follows: Ir^{182}, 15 ± 1 min.; Ir^{183}, 55 ± 7 min.; and Ir^{184}, 3.2 ± 0.2 hr. The gamma-ray spectra of Ir^{182} and Ir^{184} are very complex, each extending above 4 Mev. Positron branches are also seen in both isotopes. Studies of the gamma-ray spectra indicate that the energies of the first and second excited states of Os^{182} are slightly higher than the corresponding states in Os^{184}. These data suggest that there is a maximum in the moment of inertia at Os^{184}.

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I. INTRODUCTION

The nuclear energy levels of the osmium isotopes are of particular interest in nuclear spectroscopy because these isotopes span the transition region between two broad groups of nuclei whose properties have been characterized in terms, respectively, of axially symmetric ellipsoidal shapes and of spherical shapes. A knowledge of the level structures of these isotopes can aid the development and testing of nuclear models applicable to this intermediate region--for example, the asymmetric rotor model of Davydov and Filipov. The levels of Os^{188}, Os^{190}, and Os^{192} are known primarily from decay-scheme studies of the corresponding iridium and rhenium isotopes.[1-6] In addition some Coulomb excitation work has been reported.[6] Diamond and Hollander have studied the decays of the neutron-deficient isotopes Ir^{185}, Ir^{186}, and Ir^{187},[3] and the decay of Ir^{186} has been investigated by Scharff-Goldhaber et al.[7]

In the interval from mass number 186 to 192, two interesting features are noted in the levels of even-mass osmium isotopes: (a) the ratio of energies of the first 4+ to the first 2+ state falls from 3.17 in Os^{186} to 2.94 in Os^{190}, and (b) the energy of the second 2+ state decreases sharply from 768 kev in Os^{186} to 489 kev in Os^{192} while the first 2+ state increases

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from 137 kev in Os^{186} to 206 kev in Os^{192}. The decrease in the ratio E (4+)/ E (2+) and the increase in energy of the first excited 2+ state are attributable to a decrease in the equilibrium nuclear deformation between Os^{186} and Os^{192}. The lowering in energy of the second 2+ state may be interpreted as indicating that the heavier osmium isotopes become "softer" toward the gamma-type of near-harmonic collective oscillations.

In the interest of extending our knowledge of the level structures of osmium isotopes, we have sought to synthesize and identify iridium isotopes of mass numbers less than 185, by heavy-ion irradiations of suitably chosen targets. In this paper the syntheses of Ir^{182}, Ir^{183}, and Ir^{184} are reported.

II. EXPERIMENTAL METHOD

Heavy-ion reactions offer a useful method of preparing highly neutron-deficient isotopes relatively free of activities lying nearer to the line of stability. For example, the compound nucleus formed from the irradiation of thulium ($_{69}Tm^{169}$) with oxygen ions is $_{77}Ir^{185}$, already six mass units lighter than the lightest stable isotope, Ir^{191}. By this method, Ir^{182} has been produced by the Tm^{169} (O^{16}, 3n) reaction. Production of Ir^{183} and Ir^{184} has been achieved by means of the Lu^{175}(C^{12}, 4n) and (C^{12}, 3n) reactions. Thick metallic targets of thulium and lutetium were irradiated with 160-Mev O^{16} ions or 120-Mev C^{12} ions in the Berkeley heavy-ion linear accelerator (Hilac). Bombardment times varied from 20 min. to 6 hr, according to the isotope being studied.

After chemical purification of the iridium fractions, their decay was followed by using end-window, flowing-methane, proportional counters. Photon spectra were examined with two scintillation spectrometers. One consisted of a 1-1/2-by-1-in. NaI crystal mounted on an RCA 6655A photomultiplier tube, and the other a 3-by 3-in. crystal mounted on a Dumont 6363

tube. Gamma-gamma coincidences were measured with a conventional fast-slow coincidence circuit and with a two-dimensional analyzer.[8] Where possible, half lives and the genetic relationships to known osmium and rhenium isotopes were established by means of timed chemical separations of daughter activities.

III. CHEMICAL PROCEDURES

Iridium was separated from rare-earth metal targets by dissolving the target in 6N HNO_3, and boiling the solution almost to dryness in aqua regia to expel OsO_4. The solution was made 6 N in HCl and passed through a Dowex A-1 column to adsorb iridium (plus rhenium). After thorough washing of the column with 3N HCl, the resin was removed and equilibrated with a 10% hydroxylamine hydrochloride solution at ~100°C to reduce iridium to the trivalent state, which is desorbed. A further purification of iridium was effected by precipitation of IrO_2, by standard techniques.

In order to "milk" osmium from irradiated rare-earth targets the nitric acid solution of the target metal, containing carriers of rhenium, osmium, and iridium, was boiled almost to dryness to expel OsO_4, and then was transferred to a standard "ruthenium" distillation apparatus. For each collection of osmium daughter activity, 5 mg of osmium carrier was added to the still, together with 10 ml of concentrated HNO_3 and 1 ml of concentrated HCl. The OsO_4 was then distilled in a stream of air into 100 ml of 6N NaOH solution, which was mounted directly for scintillation analyses.

Rhenium granddaughter activities were isolated essentially by the removal of osmium as osmium sulfide from the 6N NaOH solutions of the osmium milked fractions, and the subsequent precipitation of rhenium sulfide from the acidified supernatants.

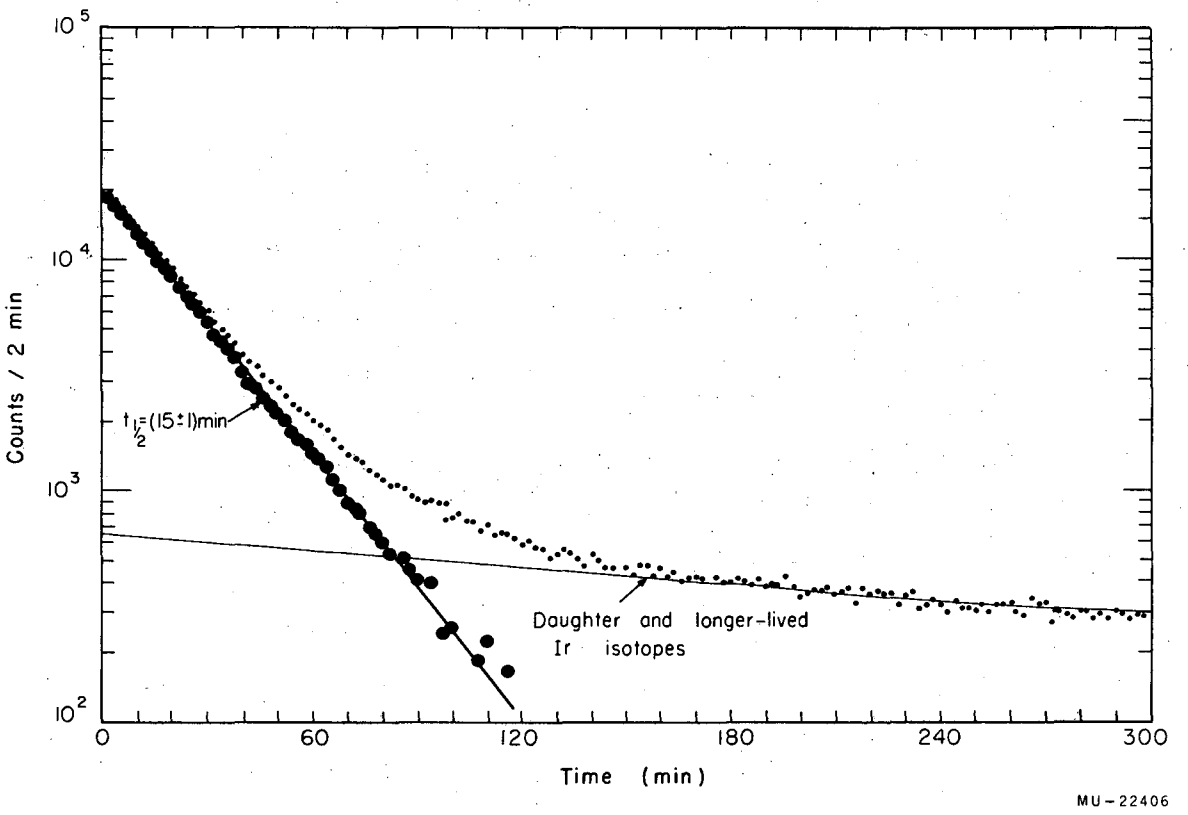

Fig. 1. Decay curve of iridium fraction from Tm(O^{16}, xn)Ir (1/16-in. aluminum between source and counter).

IV. EXPERIMENTAL RESULTS

Iridium - 182

The irradiation of Tm^{169} with 160-Mev O^{16} ions yields isotopes of iridium with mass number lower than 185. It is expected (and found) that the yield of the (O^{16}, n) reaction is very low because of the high excitation of the compound nucleus; hence one does not observe significatn amounts of Ir^{184}. Because the number of neutrons that can be evaporated at these energies may be quite large (>10), many iridium isotopes can be produced. However, in these experiments approximately 45 minutes was required to perform the initial iridium purifications, so that half lives of $\lesssim 10$ minutes probably would have wscaped detection. Thus, the lower limit on observable mass numbers is governed primarily by the speed of chemical separation.

Iridium - 182 has been identified both from irradiations of lutetium with carbon ions, and of thulium with oxygen ions. The decay curve of the iridium fraction from the Ir (O^{16}, xn) reaction is shown in Fig. 1. The half life of the shortest component, which we assign to Ir^{182}, is 15 ± 1 min. The curve shown was taken with a ~1.5-mm Al absorber interposed between source and detector; the use of an absorber enhanced the counting efficiency of the 15-min component relative to the longer-lived activities.

After an irradiation of lutetium with carbon ions, the genetic relationship of the 15-min Ir^{182} to its descendents, Os^{182} and Re^{182}, was established by a series of timed chemical separations ("milkings") in which OsO_4 was volatilized out of the iridium fraction every 15 min. In the early milkings, Os^{182} was clearly recognizable by its prominent 510-kev gamma ray[9] (not annihilation radiation) which showed the characteristic 21 hr half life of Os^{182} (see Fig. 2). The yield of this 510-kev photon in each milking as a function of the time of the milking is shown in Fig. 3A; from this curve a

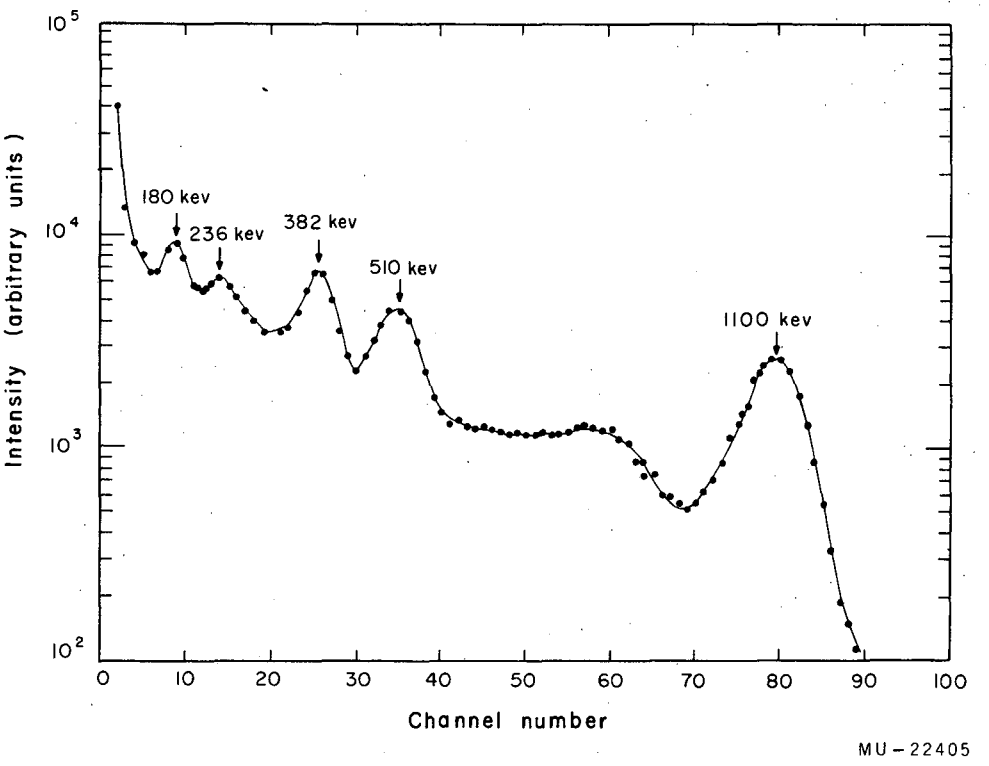

Fig. 2. Gamma-ray spectrum of first Os fraction milked from Lu (C^{12}, xn) Ir. The presence of 12-hr Os^{183} is indicated by the prominent photon at 382 kev, 9.9-hr Os^{183m} by the 1100-kev photon, and 21-hr Os^{182} by the 510-kev photon (reference 9).

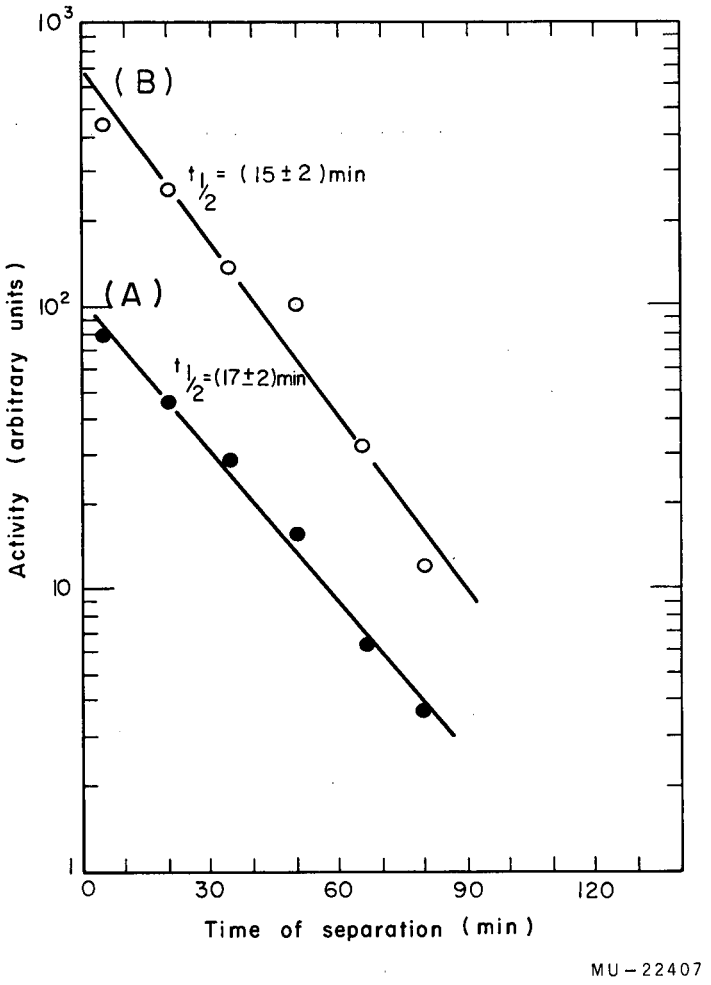

Fig. 3. (A) Yields of 510-kev gamma ray of 21-hr Os^{182} as a function of time of separation of Os from parent Ir.
(B) Yields of 1-Mev group of 13-hr Re^{182} in Re fractions separated from Os fractions of (A).

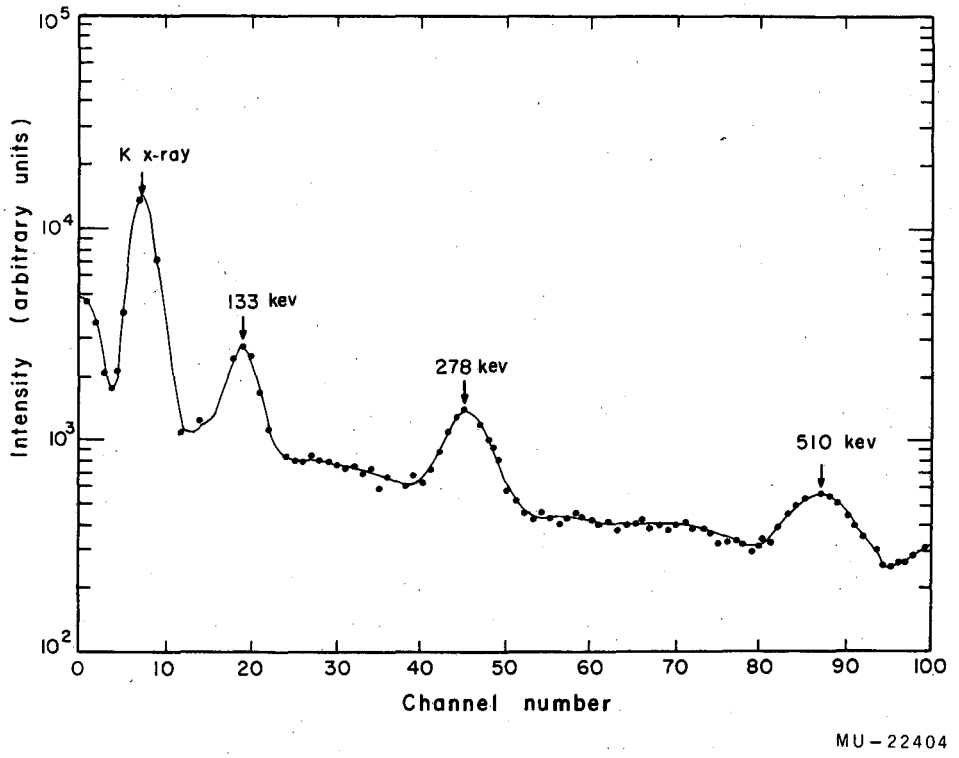

Fig. 4. Low-energy gamma-ray spectrum of 15-min Ir from Tm (O^{16}, xn) Ir.

half life of ~17 min. is obtained for the parent of Os^{182}. As a further check on this assignment, rhenium was separated from the first few osmium fractions 24 hr after their isolation. The gamma-ray spectrum of the rhenium fraction contained composite peaks at 1 and 2 Mev similar to those found by Gallagher et al.[10] in 13-hr Re^{182}. Figure 3B shows a plot of the 1000-to-1300-kev composite peak versus the time of chemical separation of the osmium parent from the iridium grandparent. From this curve we obtain ~16 min. as the half life of Ir^{182}. From the direct decay of the photon peaks in the Ir^{182} gamma spectrum, the measured half life is 15.5 ± 1 min. A consideration of these data leads to the value 15 ± 1 min. as the "best value" of the Ir^{182} half life.

In the Tm(O^{16}, xn) bombardment, a fairly pure gamma-ray spectrum of Ir^{182} could be observed briefly in the iridium fraction without interference from the spectrum of the daughter Os^{182} activity. One of the earliest spectra is shown in Fig. 4. Clearly evident are K x-rays and photons of 133 ± 5 kev and 278 ± 5 kev. These photons are interpreted as transitions that de-excite the first and second rotational states of Os^{182}, respectively. The Ir^{182} spectrum above 500 kev is extremely complex, and extends higher in energy than 4 Mev.

Iridium - 183

Iridium-183 and 184 were produced from carbon-ion irradiation of lutetium metal by the reactions Lu^{175} (C^{12}, 4n) and (C^{12}, 3n). The proportional-counter decay curve showed the presence of activities of ~20 min. (Ir^{182}), ~1 hr, ~3 hr, and longer half lives.

Iridium-183 was identified principally from several series of milking experiments done at 20 and 40-min. intervals, for periods of up to 6 hr. The milked osmium fractions were examined in liquid form (~100 cc) under

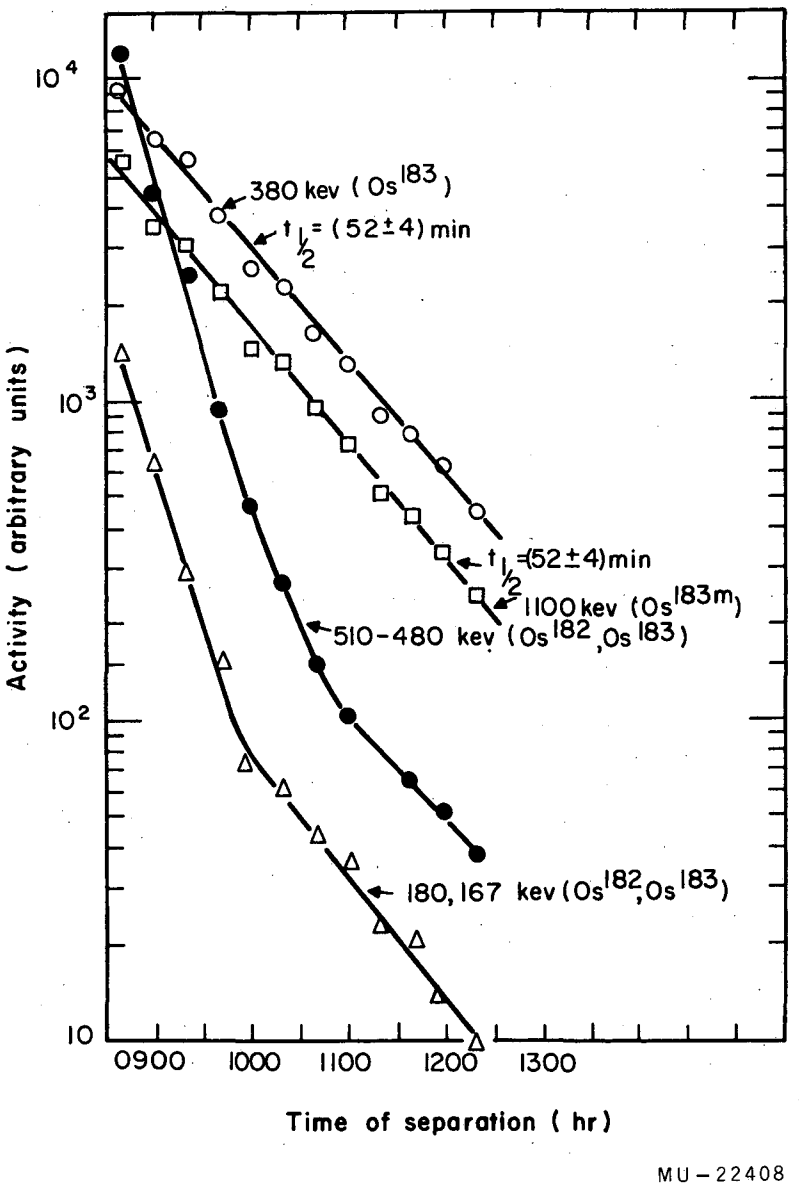

Fig. 5. Yields of gamma rays in Os milked from Ir produced by Lu (C^{12}, xn) Ir.

identical conditions in the scintillation spectrometer. Figure 2 shows the gamma spectrum of the first osmium milking measured immediately after chemical separation. The presence of 12 hr Os^{183} is indicated by the prominent photon at 382 kev, while the 1100-kev photon arises from the decay of 9.9 hr Os^{183m}, as reported by Newton.[9] Since both the 382-and 1100-kev photons (in the osmium milkings) are detected in comparable amounts immediately after the chemical separation, it is evident that the isomeric states of osmium 183 are populated simultaneously in the decay of Ir^{183}. In the first osmium fraction, the presence of Os^{182} which arises from the decay of 15-min. Ir^{182} is indicated by the 510-kev photon and by the subsequent growth of Re^{182}. The later osmium fractions show evidence neither of Os^{182} nor of Re^{182}.

The half life of Ir^{183} was obtained by plotting the relative yield of the 382-kev Os^{183} photon and that of the 1100-kev Os^{183m} photon as a function of the time of separation of osmium from the parent iridium fraction. Figure 5 shows one such yield curve as well as the yield curves of the 180- and 510-kev photons. The short components of these decays represent the 180- and 510-kev gamma rays in the decay of Os^{182} (which grew from Ir^{182}), while the long components arise from the 168- and ~480-kev gamma rays in the decay of Os^{183}. From several such experiments the best value of the Ir^{183} half life appears to be 55 ± 7 min. A sample of Ir^{183} sufficiently free of Ir^{182} and Ir^{184} to permit the observation of a distinct Ir^{183} gamma-ray spectrum could not be prepared. However, in the iridium fraction isolated from lutetium bombarded with carbon ions, a photon of energy about 238 kev was observed to decay with a 58-min. half life.

It should be noted that the 117-, 236-, and ~510-kev photon peaks in the osmium milkings showed an initial short-lived decay (about 110 min.) as

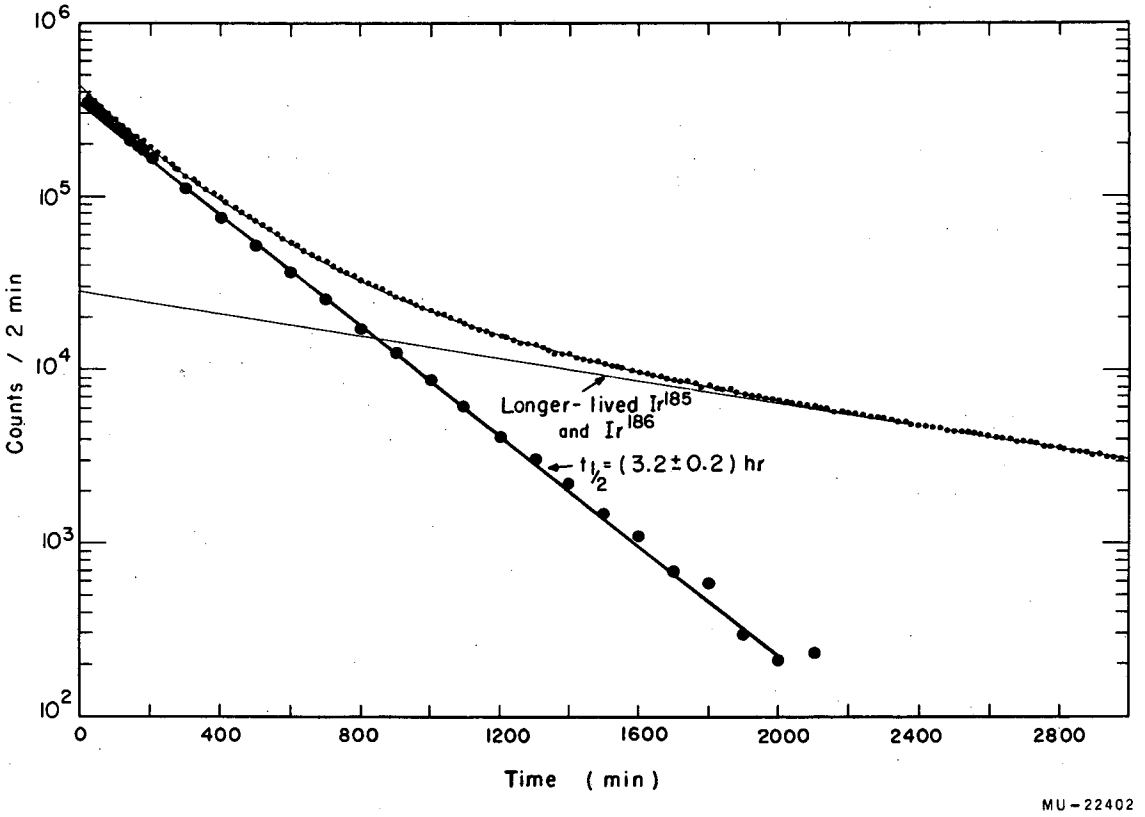

Fig. 6. Decay curve of Ir fraction from Lu (C^{12}, xn) Ir chemically separated seven hours after irradiation.

well as the expected longer periods of the Os^{183} isomers. That these gamma rays, with energies similar to those of some Os^{183} gamma rays, show such a short component while the 180-kev photon (known to occur in the decay of Os^{182}) does not might suggest the possibility of a third isomeric state in Os^{183}. It is further observed, however, that the prominent 382-kev gamma ray (of Os^{183}) does not show the short-lived component. Therefore, if the 236- and 510-kev photons seen here really originate from Os^{183} decay, they cannot be identified with the transitions of similar energies in the decay scheme of Os^{183} reported by Newton,[9] since this would require the simultaneous observation of a (short-lived) 380-kev gamma ray.

Iridium -- 184

The prominent 3-hr component of the decay curve from the Lu (C^{12}, xn) bombardments was thought to be associated with Ir^{184}. For a study of this activity, the irradiated target was allowed to stand for about nine hours before chemical purification, to permit the shorter-lived iridium isotopes of mass number less than 184 to decay. Figure 6 shows the decay curve obtained from such a fraction by using a proportional counter. The longer-lived "tail" arises from the presence of Ir^{185} and possibly Ir^{186} (~15 hr) rather than from daughters. This was shown by a subsequent repurification of the iridium fraction and by the presence in the scintillation spectrum of photons known to occur in the decays of Ir^{185} and Ir^{186}.[3] The first few points in the resolved 3.2-hr component are high because of the presence of some 55-min. Ir^{183}. Because there was no indication of a 3-hr iridium parent in the osmium milking experiments and since the gamma-ray spectrum of the 3-hr activity was similar to that expected for a light even-even osmium isotope, we conclude that the activity is Ir^{184}, which decays to stable Os^{184}. Our "best value" for the half life of Ir^{184} is 3.2 ±0.2 hr.

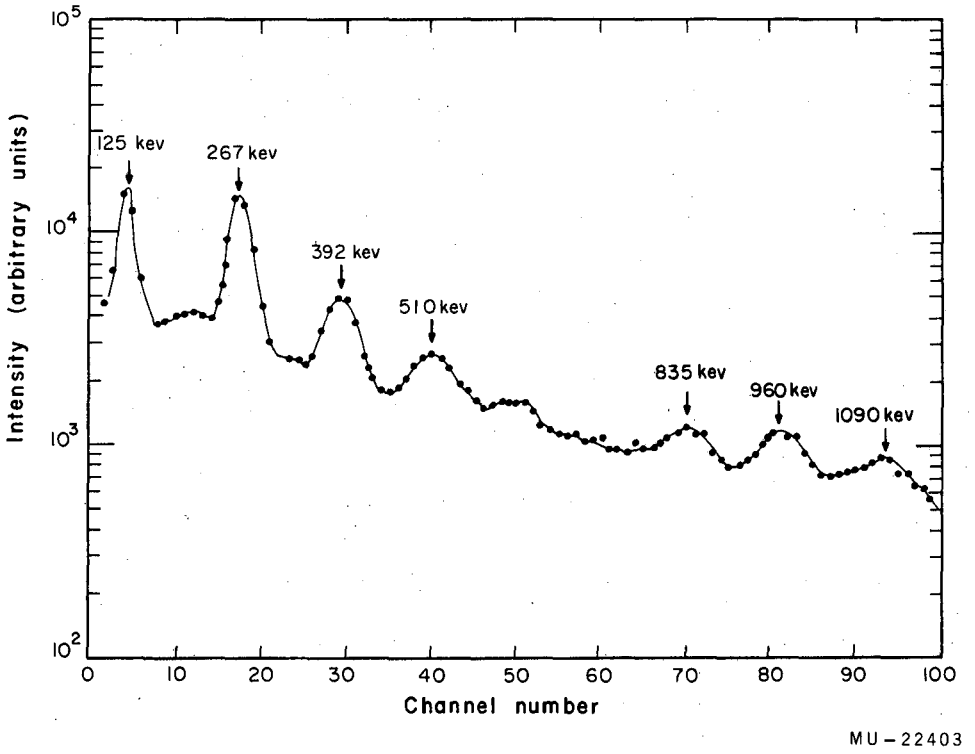

Fig. 7. Low-energy gamma-ray spectrum of Ir^{184}.

The low-energy spectrum of Ir^{184} is shown in Fig. 7. Besides the K x-rays (not shown), gamma rays are present with energies of 125 ± 5, 267 ± 5, 392 ± 5, and 511 ± 5 kev (probably annihilation radiation). The relative photon intensities are 100, 203 ± 20, 87 ± 9, and 57 ± 8, respectively. The gamma-ray spectrum above the 511-kev peak is quite complex, but additional gamma rays of about 835, 960, and 1090 kev are evident, as well as others which are not completely resolved. The end point of the gamma-ray spectrum is about 4.3 Mev. Gamma-gamma coincidence experiments show that the 125-, 267-, and 392-kev gamma rays are in cascade, indicating excited states in Os^{184} at 125, 392, and 784 kev, with possible spins of 2+, 4+, and 6+, respectively. A partial level scheme for Os^{184} is shown in Fig. 8.

V. DISCUSSION

In the work reported here, the new neutron-deficient iridium isotopes with mass numbers 182, 183, and 184 have been synthesized, and their gross decay properties characterized.

Much remains to be learned about these isotopes. In the case of the mass-183 chain, we see that the two Os^{183} isomers, with widely different spins, are populated to a similar extent in the decay of Ir^{183}. Thus Ir^{183} itself may possess two isomeric states of similar half lives, or the decay of Ir^{183} may lead to high-lying states of Os^{183} which decay to both isomeric levels. The possibliity of triple isomerism in Os^{183} also warrants investigation.

In the even-mass osmium isotopes, it is of interest to correlate the energies of the first 2+ and 4+ excited states. Figure 9 shows such a correlation diagram in which the energies of Os^{182} and Os^{184} have been

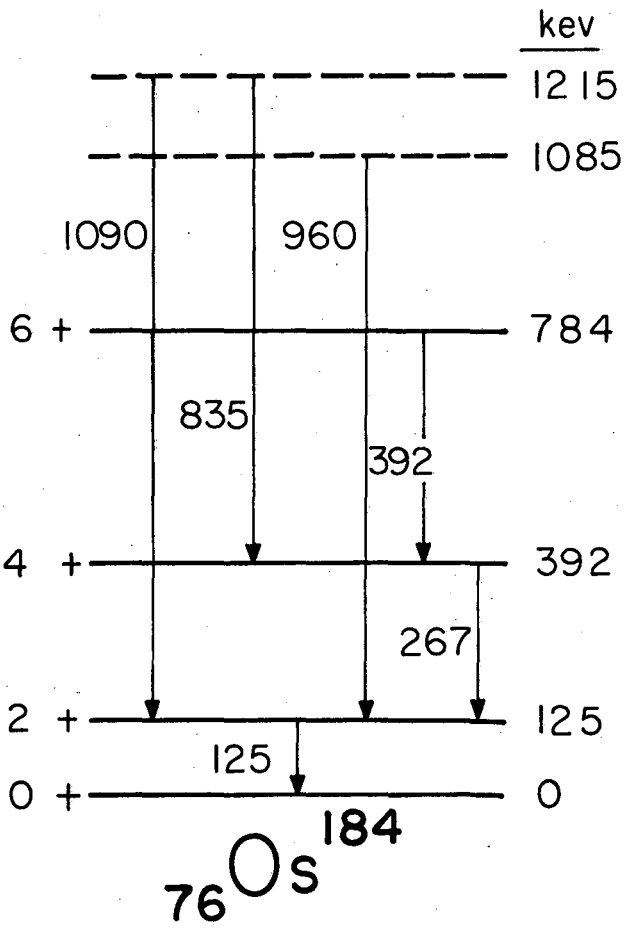

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Fig. 8. Partial level scheme of Os^{184}.

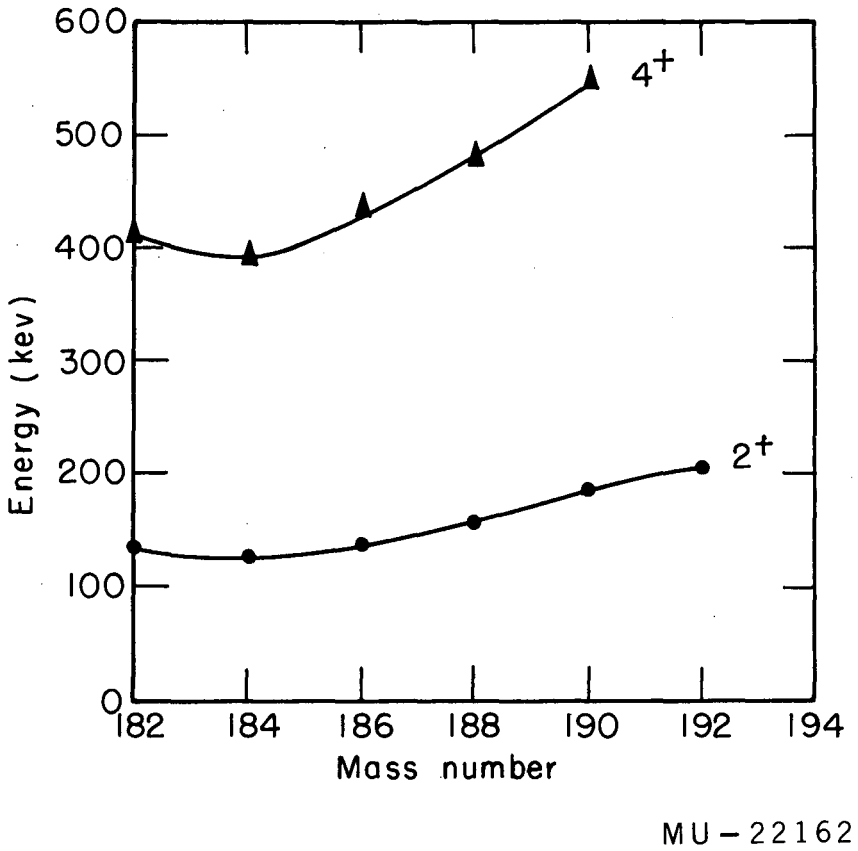

Fig. 9. Energies of first 2+ and 4+ levels in even-mass Os isotopes.

included. Note that there is a minimum in the energies of the 2+ and 4+ states at mass number 184. It thus appears that a maximum in the moments of inertia of the osmium isotopes is reached at Os^{184}, which has 108 neutrons. It is interesting to note that Nilsson has calculated theoretically that the position of this maximum is expected to be at neutron number 106.[11]

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