The Radioactive Decay of Cs^{134}, Os^{185}, Os^{191}, and Os^{193}

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(Received January 19, 1953)

From sources of cesium of high specific activity produced in the pile and studied in photographic magnetic spectrometers, nineteen electron conversion lines are observed. These are interpreted to show the existence of eleven gamma-rays, four of which have not been previously reported. Certain K/L ratios are measured and the resolution of the components of the beta-radiation presented. A plausible decay scheme consisting of seven levels in the resultant barium 134 nucleus is offered.

By irradiating osmium sources for various periods in the pile and obtaining successive exposures in magnetic spectrometers, it is possible to determine the energy and half-life of each electron conversion-line. Osmium 185 decays by K capture with a half-life of 96 days. Four gamma-rays are associated with this activity. Osmium 191 decays by a 14-hour isomeric transition followed by beta-emission with 15-day half-life. Two gamma-transitions in sequence follow the beta-decay. Osmium 193 is a beta-emitter with a 31-hour half-life. Some eighteen electron conversion lines are observed which indicate the presence of nine gamma-rays, only three of which had been previously observed.

I. CESIUM^{134}

SINCE the early observation[1] of a long-lived (2.3-year) radioactivity believed to be the result of Cs^{134}, many publications[2] reporting on its beta- and gamma-energies have appeared. As both $_{54}Xe^{134}$ and $_{56}Ba^{134}$ are stable isotopes, $_{55}Cs^{134}$ might be expected to decay both by beta-emission and by K capture or positron emission. Siegbahn and Deutsch[3] could find no evidence for the existence of the K x-rays of zenon and thus set an upper limit of 5 percent for K capture. The beta-spectrum is complex and several gamma-rays are present.

Specimens derived from the pile have been studied in magnetic spectrometers of high resolution, using both conversion and photoelectrons. The interpretation of the many conversion electron lines, whose energies are given in Table I, appears definite when both K and L lines are present. Where only a single electron line is visible, it is assumed to be a K line in barium. There thus appear to be at least eleven gamma-rays, as shown in Table II, for eight of which both K and L lines are present. Four of the gamma-rays had not been previously observed. The corresponding energies, as reported in two recent investigations,[4,5] are included in columns 2 and 3 in Table II.

TABLE I. Electron energies from Cs^{134} (kev).

Electron energy	Interpretation	Energy sum	Electron energy	Interpretation	Energy sum
165.1	K	202.5	765.2	K	802.6
438.1	K	475.5	790.3	L	796.3
468.5	L	474.5	795.1	M	796.4
525.6	K	563.0		or L	801.1
532.3	K	569.7	1001.5	K	1038.9
568.0	K	605.4	1033.5	L	1039.5
599.2	L	605.2	1130.7	K	1168
604.3	M	605.6	1162	L	1168
625.3	K	662.7	1331	K	1368
760.1	K	797.5	1362	L	1368

The K/L conversion ratios for two of the gamma-rays were measured by the densitometer method and the transitions interpreted as being electric quadrupole. Only a visual estimate was made of the ratios for the remaining gamma-rays as shown in column 5. It is possible to propose a relatively simple level scheme, as shown in Fig. 1, that will accommodate as transitions all but one of the observed gamma-energies. It had been suggested[4] that the two gamma-rays reported as 1.03 and 1.17 Mev, from single electron conversion lines, were either due to impurities or more likely were associated with K capture. From coincidence measurements[3] it had appeared that the three strong gamma-rays, 560, 602, and 799, were in sequence and the energy values of the two new lines did not fit as cross-over transitions. However, with the present revisions in the values of the energies and from the observed K-L differences, the probability of the gamma-rays being either caused by impurities or K capture appears unlikely. The electron energies more nearly satisfy the work functions for barium (i.e., beta-emission), although the certainty in measurement is not such as to preclude completely the possibility of K capture. Moreover, with the additional new gamma-energies the existence of many mathematical relationships is apparent. For example, it can hardly be regarded as fortuitous that 475 kev and 563 kev add to yield 1038 kev and similarly 563 kev and 605 kev give 1168 kev, both sums being observed as gamma-energies. The energy levels in the proposed scheme have values of 0, 605, 1168, 1402, 1643, 1846, and 1973 kev.

The beta-spectrum had originally been reported[2] as consisting of two components of about 90 and 650 kev. Considerable uncertainty must attend the resolution of

[1] D. Kalbfell and R. Cooley, Phys. Rev. 58, 91 (1940).
[2] K. Way, "Nuclear Data" Circular No. 499, Natl. Bur. Standards, 1950.
[3] K. Siegbahn and M. Deutsch, Phys. Rev. 73, 410 (1948); see also W. Mims and H. Halban, Proc. Roy. Soc. (London) A64, 311 (1951).
[4] Waggoner, Moon, and Roberts, Phys. Rev. 80, 420 (1950).
[5] F. Schmidt and G. Keister, Phys. Rev. 86, 632 (1952).

a composite beta-spectrum when three or four components are present, particularly for the lower energies. A careful survey of the beta-spectrum of Cs^{134} has been carried out in this laboratory by Mr. A. E. Stoddard, using the double-focusing magnetic spectrometer. No indication could be found for two high energy components differing by about 50 kev, as previously reported,[5] but rather there was a single upper limit at 657±5 kev. A second beta-component exists at 410±5 kev. At lower energies, observations are not incompatible with a weak component at about 210 kev, with a stronger low energy beta of upper limit about 80 kev. These energies are in agreement with the transition values required to satisfy the scheme shown in Fig. 1. The relative percentages for the number of betas in the order of decreasing energies are about 81, 6, 3, and 10, respectively. The weak 662-kev gamma-ray for which only a single (assumed K) electron line was observed seems not to fit in the proposed level plan, and it may be subject to other interpretation.

TABLE II. Gamma-energies (kev).

Arbitrary number	WMR[a]	SK[b]	Present	K/L ratio
1			202.5	
2			475.0	~5
3	560	561.5	563.0	~10
4		566.5	569.7	
5	602	601.2	605.4	6.4±0.8, *E*2
6			662.7	
7	799	793.1	796.8	7.3±0.8, *E*2
8			802.6	
9	1030	1037.2	1039	~10
10	1170	1164.4	1168	~10
11	1363	1365.7	1368	~10

[a] See reference 4.
[b] See reference 5.

Barium 134, being an even-even nucleus, should have a ground state of zero spin and even parity. The log*ft* for the 657-kev beta is about 8.8, indicating a first forbidden transition with a spin change of zero or one, together with a change in parity.

II. OSMIUM

The capture of neutrons in the seven stable isotopes of osmium has been shown to produce three radioactive emitters, whose half-lives have been reported to be about 96 days, 16 days, and 31 hours. The longest-lived activity has been assigned to the mass 185, formed from the mass 184, whose normal abundance is only 0.02 percent. Considerable confusion has existed in the assignment of the other two activities. Early experiments by Seaborg and Friedlander[6] in which the osmium was exposed first to slow and then to high energy neutrons indicated that the $(n,2n)$ process favored the production of the 31-hour activity over the 16-day yield by an appreciable factor. This would have

[6] G. Seaborg and G. Friedlander, Phys. Rev. **59**, 400 (1941).

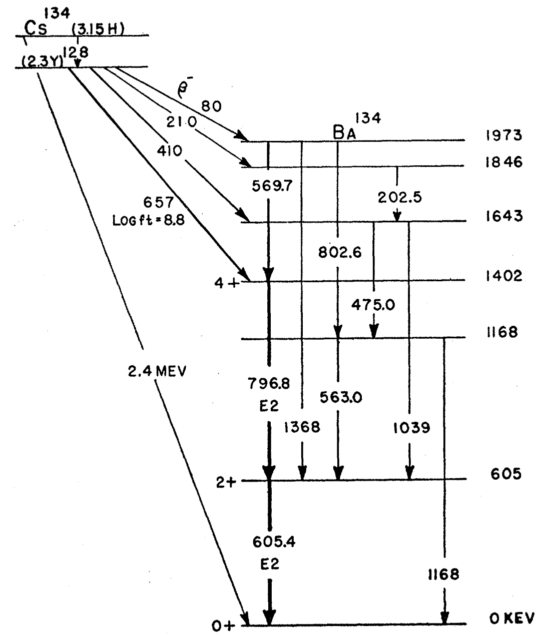

FIG. 1. Nuclear levels in barium 134 following β-emission from cesium 134.

indicated that the 16-day activity was associated with the isotope of mass 193. Similarly, Goodman and Pool[7] could find no evidence for the production of the 16-day emitter when osmium was bombarded by fast (9-Mev) neutrons, and they did observe the 31-hour activity. More recently, Swann and Hill have found that the (γ,n) reaction osmium with 22-Mev gamma-radiation results in the formation of the 15-day activity with no trace of the 31-hour product. It can thus be concluded that the 31-hour activity is in osmium 193, and perhaps the neutron energy in the earlier experiments was not adequate to induce the expected $(n,2n)$ reaction.

In the present investigation, specimens of osmium have been irradiated in the Argonne heavy water pile for periods ranging from 10 hours to 2 weeks and

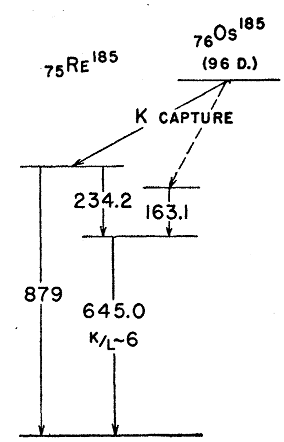

FIG. 2. A possible decay scheme for osmium 185.

[7] L. Goodman and M. Pool, Phys. Rev. **71**, 288 (1946).

TABLE III. Electron energies from Os^{193} (31 hr).

Electron energy (kev)	Interpretation	Energy sum (kev)	Electron energy (kev)	Interpretation	Energy sum (kev)
30.4	K	106.4	175.2	K	251.3
59.7	L_1	73.1	204.7	K	280.8
60.3	L_2	73.1	245.1	K	321.2
63.1	K	139.2	267.4	L	280.8
93.0	L_1	106.4	277.6	M	280.8
103.1	M	106.3	307.5	L	320.9
105.5	N	106.2	328.0	K	404.0
125.2	L	139.0	384.4	K	460.5
135.5	M	138.7	481.7	K	557.8

quickly placed in magnetic photographic spectrometers. By successive exposures from a single source, it has been possible to determine the energy and the approximate half-life of each of the many electron lines. The osmium seems always to contain traces of iridium as an impurity, which, by virtue of its large capture cross section, gives many additional but well-known lines.

Osmium 185

Osmium 185 decays by K capture to rhenium 185 with the emission of gamma-radiation with a half-life of about 96 days. In one investigation[2] two gamma-rays of energy 648 and 878 kev were reported with a relative abundance of about 6 to 1. Swann and Hill observed[8] a gamma-ray of energy 653 kev in the long-lived osmium and noted two other gammas as questionable at 163 and 235 kev. In the present investigation electron lines are observed from the long-lived osmium with energies of 91.4, 162.8, 221.4, 573.3, 633, and 807 kev. Using the work functions for rhenium, these electron lines are interpreted to yield four gamma-rays whose energies are 163.1, 234.2, 645.0, and 879 kev. These values suggest that the 879-kev gamma is a crossover for the 234.2- and 645-kev transitions. The 645-kev gamma is much stronger than any of the others in conversion, and has a K/L ratio of about 6. This suggests that the radiation is either M1 or E1 or possibly E2 in type. A possible arrangement of the transitions satisfying the energies and the observed relative intensities is shown in Fig. 2.

TABLE IV. Gamma energies, Os^{193} (31 hr).

Arbitrary number	Energy (kev)	K/L ratio
1	73.1	$(L_1/L_2)\sim 1$
2	106.3	$(L/M)\sim 3$
3	139.0	~ 5
4	251.3	
5	280.8	~ 10
6	321.2	~ 8
7	404	
8	460.5	
9	557.8	

[8] J. Swann and R. Hill, Phys. Rev. **88**, 831 (1952).

Osmium 191

It has recently been shown[8] that a gamma-transition of half-life 14 hours and energy 74.2 kev exists in osmium 191, leading to the isomeric state whose half-life is 15 days. Following a low energy beta-emission, two gamma-rays of energy 41.7 and 129.1 kev are found to be emitted in sequence. The present investigation supports this proposed decay scheme except for minor adjustments in the expressed energies.

Osmium 193

Early absorption measurements had indicated[2] that the osmium radioactivity of half-life 31 hours decayed by the emission of a beta-ray of energy 1.1 Mev and a gamma-ray of 1.17 or 1.58 Mev. In another investigation[9] only a low energy gamma-ray of about 65 kev was observed, while a different report[10] stated that no evidence could be found to indicate the existence of any

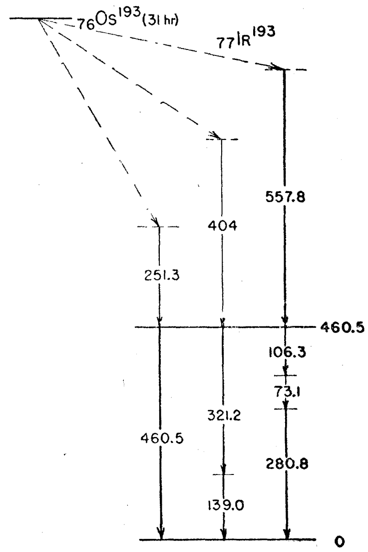

FIG. 3. Proposed nuclear levels in iridium 193 following β-emission from osmium 193.

accompanying gamma-radiation. Swann and Hill reported gamma-energies of 215, 323, and 460 kev. In addition several weak electron lines were observed with energies 55.2, 59.4, 60.0, and 67.2 kev. The 59.4- and 60.0-kev lines were assumed to be L_1 and L_2 conversion groups for a 72.4-kev gamma-transition.

In the present investigation some 18 conversion electron lines are observed to be associated with the 31-hour activity. These are shown in column 1, Table III, together with their interpretations. Of the nine gamma-rays concluded to exist, as shown in Table IV, four are based upon the observation of single K electron lines. The two electron lines at 59.7 and 60.3 kev, could be regarded as M Auger lines but no accompanying more intense L lines are present, as would be expected. They are therefore assumed to be L_1 and L_2 conversion lines.

[9] F. McGowan, Phys. Rev. **79**, 404 (1950).
[10] Bunker, Canada, and Mitchell, Phys. Rev. **79**, 610 (1950).

The gamma-ray reported by Swann and Hill at 215 kev must have been based on the assumption that the electron line at 135.5 kev is due to K conversion. It appears more probable that this is an M line for a 139-kev gamma-ray whose rather strong K and L lines are obscured in early exposure by the strong blackening due to the 14-hour activity. Subsequent plates show the lines in their proper ratio.

It is possible to observe some equivalent mathematical combinations of the gamma-energies and thus to suggest a nuclear level scheme as shown in Fig. 3. The three gamma-rays shown in the upper part of this figure could be arranged in alternate plans dependent upon the evaluation of the components of the accompanying beta-radiation. No measurement was made in this investigation of the beta-energies. The electron intensity ratios can be approximately expressed for five of the gamma-rays, as shown in column 3, Table IV. These numbers are based on visual estimates.

Some of the short exposures required to distinguish the electron lines associated with the 14-hour activity from those of 31-hour half-life were taken near the Argonne pile by Mr. W. C. Jordan. This investigation was made possible by the joint support of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.

PHYSICAL REVIEW VOLUME 90, NUMBER 3 MAY 1, 1953

Relative Abundances and Neutron Capture Cross Sections of the Neodymium Isotopes*

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(Received November 24, 1952)

A double-focusing mass spectrometer has been used to determine the relative isotopic abundances of naturally-occurring neodymium and the relative isotopic neutron capture cross sections. Allowance for isotope fractionation, an effect which is only noticeable under certain experimental conditions, has been accomplished by an averaging technique. The relative abundances are compared with the results previously published.

The relative cross sections are normalized to a value of 48 barns for the capture cross section of normal neodymium. The resulting isotopic cross sections are in satisfactory agreement with the recently published results of Pomerance.

THE neutron capture cross sections of the neodymium isotopes have been studied previously, both by activation methods[1] and with a mass spectrometer.[2] In view of the very high fluxes available with the Chalk River reactor, it was felt that the mass spectrometer studies should be repeated. This work has now been completed and the results, both capture cross sections and relative abundances, are reported below.

In a recent paper[3] Pomerance has reported on the neutron capture cross sections of the isotopes of twenty-four elements, including neodymium, by a pile oscillator method, using separated isotpes. The cross-section data obtained by the two methods are in fair agreement (See Table II).

The mass spectrometer method for determining neutron capture cross sections has been described previously.[2] A sample of neodymium was irradiated in the Chalk River pile for a period of time long enough to effect an appreciable change in the abundances of the isotopes. The relative abundances of the neodymium isotopes in the irradiated and unirradiated samples were measured with a Dempster-type double-focusing mass spectrometer[4] employing a heated crucible ion source.[5]

A comparison of the two sets of results permits the calculation of the relative capture cross sections of the individual isotopes. If the net change in the isotopic abundances is small, it is permissible to use a simple set of equations derived on the assumption that the rate of change in abundance is constant with time. If n, n_2,—n_s, and n_1', n_2',—n_s' are the initial and final abundances, respectively, of the 1st, 2nd, — and sth isotopes of an element, having capture cross sections σ_1, σ_2,—σ_s, then

$$\sigma_1 Nt = (n_1 - n_1')/\tfrac{1}{2}(n_1 + n_1'),$$
$$\sigma_2 Nt = [(n_1 - n_1') + (n_2 - n_2')]/\tfrac{1}{2}(n_2 + n_2'),$$

etc. Here N is the number of neutrons per cm^2 per sec, and t is the total irradiation time in seconds. These equations are sufficiently accurate that, even for Nd^{143}

* Submitted by one of the authors (W. H. W.) in partial fulfillment of the requirements for the degree of Master of Science at McMaster University.

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[1] W. Bothe, Z. Naturforsch. **1**, 179 (1946).

[2] D. C. Hess and M. G. Inghram, Phys. Rev. **76**, 300 (1949).

[3] H. Pomerance, Phys. Rev. **88**, 412 (1952).

[4] A. J. Dempster, Proc. Am. Phil. Soc. **75**, 755 (1935).

[5] A. E. Shaw, Atomic Energy Commission Report MDDC 308, (unpublished).