IDENTIFICATION AND DECAY OF ^{190}W, ^{196}Os, ^{230}Ra, AND ^{230}Ac†

S. Katcoff, J. Gilat*, P. E. Haustein, E.-M. Franz, N. A. Morcos**

Brookhaven National Laboratory, Upton, New York, USA

T. E. Ward, H. A. Smith, Jr.

Indiana University Cyclotron Facility, Bloomington, Indiana, USA

J. C. Hill

Iowa State University, Ames, Iowa, USA

R. F. Petry

University of Oklahoma, Norman, Oklahoma, USA

Abstract

Several new or poorly characterized neutron-rich nuclides have been produced and studied at the Brookhaven Medium Energy Intense Neutron facility, MEIN. ^{190}W decays with $T_{1/2}$ = 30.0 ± 1.5 min, E_{β} = 0.93 ± 0.07 MeV, and γ's of 157.6 and 162.1 keV. ^{196}Os decays with $T_{1/2}$ = 35.0 ± 0.4 min and γ's at 126.1, 200.7, 207.0, 257.0, 315.3, 407.6, 522.2, and 628.9 keV. ^{230}Ra decays with $T_{1/2}$ = 93 ± 2 min to 122 ± 3 sec ^{230}Ac; γ-ray energies and intensities are tabulated. A decay scheme is proposed for ^{190}W, and many of the transitions following the decay of ^{230}Ac are between known levels in ^{230}Th.

1. Introduction

A Medium Energy Intense Neutron facility[1], MEIN, has recently been installed at the 200-MeV Linac injector of the Brookhaven Alternating Gradient Synchrotron (AGS). Proton beams with mean currents up to 100 μA interact with a water cooled copper beam stop to generate secondary neutrons whose energy spectrum is reasonably flat in the region 30 to 160 MeV. The flux of neutrons (E ≥ 25 MeV) is ∿1.3 x 10^{11} n/cm^2-sec for 100 μA of protons; it is well suited for producing sources of neutron-rich isotopes by (n,2pxn) and (n,3pxn) reactions[1,2]. Targets, which are usually isotopically enriched, are irradiated in pneumatically operated rabbits and then rapidly transferred to the laboratory for chemical processing and nuclear spectroscopy. Although neutron-rich isotopes can also be made by irradiation with the primary proton beams[3,4] via reactions such as (p,3p), there is usually serious interference from neutron-deficient isotopic nuclei which are produced in much greater abundance. Neutron irradiation improves the ratios[1] of neutron-rich to interfering neutron-deficient isotopes by factors of 10 to 100.

Results on the new nuclide 68-sec ^{62}Fe have been reported previously[2]. Here we report on ^{190}W, ^{196}Os, ^{230}Ra, and ^{230}Ac produced by (n,2pn) reactions with effective cross sections of 0.1-0.5 mb.

2. Experimental

Targets of osmium metal enriched to 98% ^{192}Os were used to prepare sources of ^{190}W by the (n,2pn) reaction as well as by (p,3p). Separation of the W was by distillation of OsO_4, precipitation of tungstic acid, scavenging of Re and Ta as oxides, and finally precipitation of tungsten α-benzoin oxime. The ^{196}Os sources were made by (n,2pn) from 50-300 mg targets of $PtCl_4$ (96% ^{198}Pt). Purification was by distillation of OsO_4 into a NaOH solution, acidification to pH 5.5 with HCl, and then precipitation of OsS_4 with H_2S. Sources of ^{230}Ra (and its short-lived ^{230}Ac daughter) were made from 0.5-1.0 g targets of $^{232}ThO_2$ which were irradiated at the MEIN facility. The Ra^{+2} was first coprecipitated with 1 mg Ba^{+2} as the chloride from a cold concentrated HCl-ether solution. Further purification of the Ra and Ba was by scavenging with $Fe(OH)_3$ and two reprecipitations of $BaCl_2$. Finally the Ra was thoroughly separated from the Ba (including very large activity of fission product Ba) by cation exchange[5] on Dowex 50 (4X, 20-30 μm particles). The eluent was 0.1 M NH_4EDTA, 0.3 M $NH_4C_2H_3O_2$, adjusted to pH 5.5; Ac^{+3} was eluted in the first column volume, the Ba^{+2} followed after about 6 column volumes, and then Ra^{+2} started to appear at 10 column volumes.

Measurements of the γ-ray spectra were with 50 cm^3 Ge(Li) detectors (resolution of 1.7-1.9 keV at 1332 keV); data analysis was by means of INTRAL[6] and CLSQ[7] computer codes. X-radiation was studied with a thin Ge(li) detector, and β-radiation with a plastic scintillator. Beta-gamma coincidence measurements were also performed in which the Ge(Li) detector was used to gate on selected γ-rays and the plastic scintillator used to detect the spectrum of coincident β radiation.

3. Results and Discussion

3.1 Identification and Decay of ^{190}W

The well known[8] 3.1-min ^{190}Re was shown to grow into the purified W sources and then decay with a half-life of 29 ± 2 min (Fig. 1). Successive chemical milking of ^{190}Re from a tungsten fraction on a column of alumina confirmed the genetic relationship.

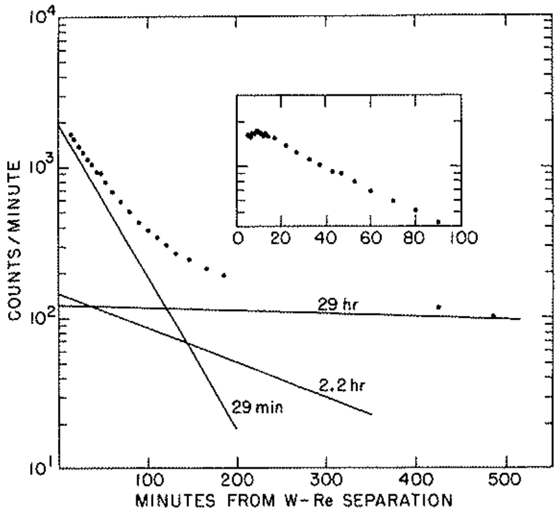

Fig. 1. Growth and decay of β-rays (E > 900 keV) following separation of 3.1-min ^{190}Re daughter activity from a ^{190}W source.

The γ-ray spectra showed the known radiations[8,9] of ^{190}Re decaying with $T_{1/2}$ = 30.0 ± 1.5 min (after secular equilibrium was attained). In addition, two new lines, at 157.6 and 162.1 keV, decayed with the same half-life and were attributed to ^{190}W (Fig. 2). Their intensities per decay of the ^{190}Re

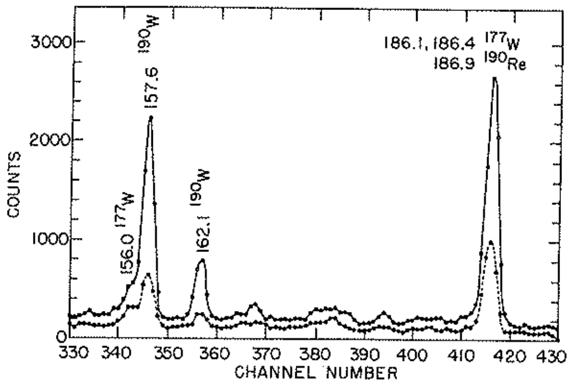

Fig. 2. γ-ray spectra in region 150-200 keV of source containing ^{190}W. Solid and dashed curves correspond to data taken at about 50 and 150 minutes after end of irradiation.

daughter are 0.39 and 0.11, respectively. The β-ray spectrum in coincidence with the 157.6-keV γ-ray had an end-point at 0.95 ± 0.07 MeV. Figure 3 shows a proposed decay scheme and a comparison with theoretical predictions. A $Q_β$ value of 1.27 ± 0.07 MeV is inferred which is close to 1.21 MeV given in the mass table of Viola, et al[10].

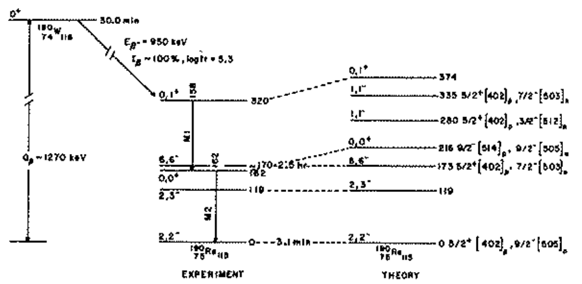

Fig. 3. Proposed decay scheme for ^{190}W and theoretical level structure of ^{190}Re.

3.2 Identification and Decay of ^{196}Os

The γ-ray spectrum from the osmium fraction, separated from irradiated ^{198}Pt, was found to contain seven lines characteristic[11] of 52-sec ^{196}Ir. These decayed with $T_{1/2}$ = 35.0 ± 0.4 min. Ten more lines (Table 1) decayed with the same half life and were assigned to decay of ^{196}Os. The most precise lifetime measurement was from decay of the ^{196}Ir β-rays with E > 1.5 MeV (Fig. 4). Beta-rays from ^{196}Os with maximum energy of 440 ± 50 keV were determined to be in coincidence with the 407.6-keV γ-rays. Some progress has been made toward elucidation of a decay scheme.

3.3 Characterization of ^{230}Ra and ^{230}Ac

The discovery of ^{230}Ra was reported[12] by Jenkins and Seaborg who observed beta rays decaying

Table 1. Energies (keV) and intensities (per 100 disintegrations) from decay of 35.0-min ^{196}Os; preliminary values.

$E_γ$	$I_γ$	$E_γ$	$I_γ$
126.1	4.9	315.3	2.6
200.7	0.6	407.6	6.1
207.0	2.5	522.2	0.8
257.0	2.4	∿586	0.6
∿308	∿0.4	628.9	1.6

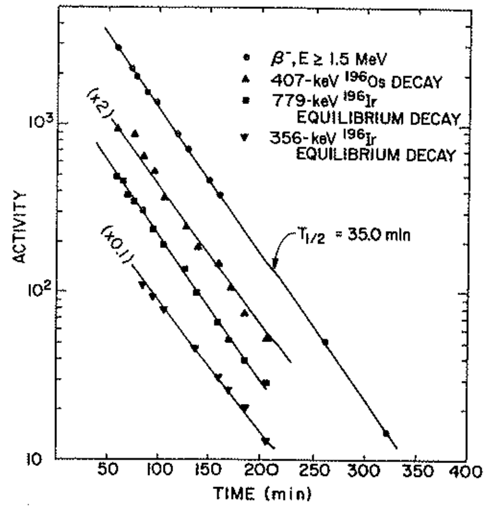

Fig. 4. Decay of various radiations from ^{196}Os and its daughter ^{196}Ir in secular equilibrium. Lines are least squares fits to the data points.

with a half-life of one hour in a radium fraction separated from thorium bombarded with 180-MeV deuterons. They set an upper limit of one minute on the half-life of the ^{230}Ac daughter which was presumed to be present in secular equilibrium. Chayawattanangkur, Herrmann, and Trautmann[13] produced ^{230}Ac directly by irradiation of Th with 150-MeV brehmstrahlung. After rapid chemical separation of Ac they found two γ-rays, at 455 and 508 keV, and these were reported[13] to decay with $T_{1/2}$ = 80 ± 10 sec. These γ-rays correspond to transitions from the well characterized[14] 508-keV level in ^{230}Th.

In the present work numerous γ-rays were found from decay of both ^{230}Ra and ^{230}Ac. The parent half-life was measured as 93 ± 2 min and that of the daughter as 122 ± 3 sec. Figure 5 shows the results of a chemical milking experiment in which Ac was eluted at 20 minute intervals from a Dowex-50 column which retained the Ra. The radiations from the Ac fractions were measured with a NaI well detector. The genetic relationship and the respective half-lives of ^{230}Ra and ^{230}Ac are clearly established. A minor interference is shown by the 10.6-h component which results from the presence of ^{224}Ra and its decay products in the separated Ra fraction.

In order to distinguish γ-rays following decay of the ^{230}Ra parent from those following decay of the ^{230}Ac daughter, a continuous elution procedure was used. In one case the eluent from the cation exchange column was passed continuously through a flat 1 ml cell placed next to the Ge(Li) detector. Thus ^{230}Ac γ-ray spectra were obtained without interference

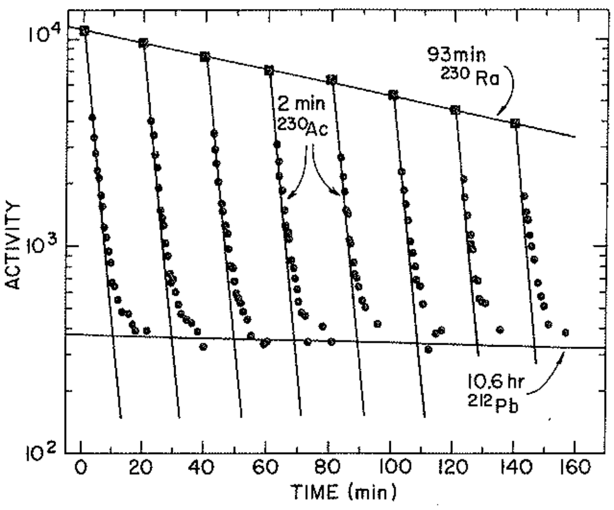

Fig. 5. Decay of Ac fractions milked from Ra parent at 20 minute intervals. Squares were obtained by least square fits to data points (circles) and extrapolations to the times of separation from the Ra.

from ^{230}Ra. In the other case the column itself was put next to the detector while the Ac was being swept out. Most experiments, however, were done with sources containing both parent and daughter. Tables 2 and 3 give the γ-rays and their intensities relative to 100 for the 454.9-keV γ of ^{230}Ac. Figure 6 shows decay of some of these γ-rays.

The level scheme of ^{230}Th has been previously determined[14)] from decay of 17.4-d ^{230}Pa and from inelastic scattering of deuterons[15)] on ^{230}Th. Of the 117 γ-rays associated with decay of ^{230}Ac (Table 3), 22 correspond to transitions between these known levels. About 45 additional transitions can be accommodated by adding 15 new levels between 1297.2 and 2282.5 keV. About 80% of the total γ-ray intensity is via these transitions. For the levels of ^{230}Ac itself no previous information is available, but analysis of the data from decay of ^{230}Ra should yield a tentative decay scheme. Beta-gamma coincidence measurements on decay of ^{230}Ra showed β-rays of $E_{max} \approx 500$ keV in coincidence with γ-rays of 63.0, 72.0, and 202.8 keV; thus $Q_\beta \geq 700$ keV. In decay of ^{230}Ac β-rays of $E_{max} \approx 1400$ keV were found to be coincident with the 1243.9-keV γ-rays and Q_β is probably ≈2700 keV.

Table 2. γ-rays from decay of 93-min ^{230}Ra. The energies are in keV and the intensities are normalized to 100 for the 454.9-keV γ-ray of ^{230}Ac. Values are preliminary.

E_γ	I_γ	E_γ	I	E_γ	I_γ
49	~1	178.3	2.3	296.1	1.0
63.0	37.1	184.1	11.7	316.4	0.8
72.0	99.0	189.2	17.0	412.8	0.9
101.0	15.2	192.7	1.7	448.9	15.0
110.7	3.4	198.2	5.2	457.9	19.0
134.3	4.1	202.8	31.5	469.7	29.7
147.9	5.4	211.8	11.7	478.7	24.1
151.5	1.9	251.5	8.4	484.4	2.0
162.9	2.7	274.6	1.5	509.5	7.5
167.7	1.2	285.2	18.0	537	1.3
174.8	1.8	292.9	3.6		

Table 3. γ-rays from decay of 122-sec ^{230}Ac; energies in keV, intensities relative to 100 at 454.9 keV. Values are preliminary.

E_γ	I_γ	E_γ	I_γ	E_γ	I_γ
53.3	(1.5)	987	---	1642.5	1.3
120.8	3.1	991.2	1.3	1675.4	1.2
388.5	1.4	999.1	2.7	1691.7	9.7
397.7	4.0	1009.7	3.1	1695.7	4.0
423.2	0.7	1026.3	2.1	1717.5	10.1
444.0	1.4	1043.2}	2.0	1721.9	10.4
448.9	2.9	1044 }		1757.5	14.1
454.9	100	1053.1	1.7	1775.3	17.9
508.2	61	1068.7	1.6	1797.2	1.1
518.5	3.3	1093.5	2.0	1800.4	0.8
571.1	0.7	1099.8	1.0	1810.7	2.8
581.8	5.4	1106.2	0.7	1939.6	1.5
624.4	1.8	1147.9	4.7	1869.0	2.4
628.8	2.6	1187.5	1.5	1896.7	8.6
652.0	(0.6)	1198.9	1.9	1902.7	10.7
677.6	2.0	1212.0	2.3	1913.8	9.3
728.0	5.2	1226.8	11.4	1920.2	7.1
750.4	1.0	1243.9	53.2	1949.8	21.0
772.6	2.9	1252.5	0.7	1956.9	7.3
781.5	4.6	1267.1}	7.9	1966.7	1.3
789.0	6.0	1268 }		1971 }	1.5
798.0	1.5	1302.6	6.0	1973 }	
816.7	3.8	1311.5	1.8	2000.9	4.7
839.9	2.2	1322.1	10.9	2010.1	1.3
867.1	5.2	1347.7	24.1	2024.6	0.8
878.0	1.0	1375.4	15.7	2069.5	4.9
892.7	8.0	1392 }	2.1	2084.9	4.4
898.5	2.8	1395 }		2098.6	9.3
913.7	1.1	1401.0	4.2	2122.8	10.3
918.6	3.2	1432.4	2.9	2150.9	1.1
927.9	1.1	1438	---	2187.7	1.2
939.2	0.9	1455.5	2.0	2203.0}	1.0
946.3	1.0	1524.6	0.7	2205 }	
952.0	9.7	1536.6	0.6	2229.6	1.4
956.5	5.8	1573.5	3.1	2233.5	~1.0
959.1	2.4	1585.4	3.2	2282.5	2.1
973.0	1.6	1612.4	1.1	2298.6	1.0
977.6	0.8	1625.1	2.2	2314.0	0.8
982	---	1636.8	1.8	2330.8	1.1

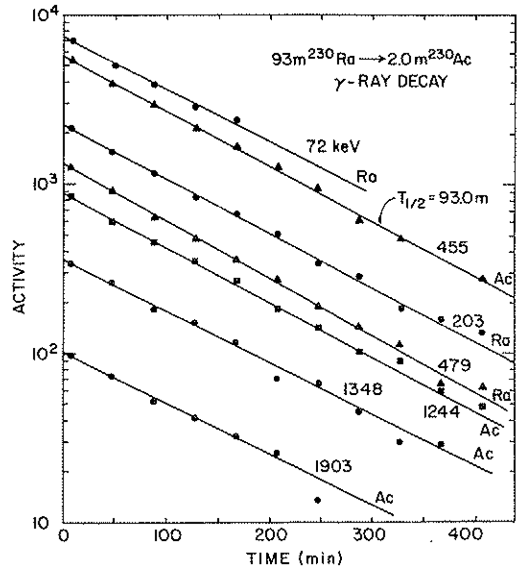

Fig. 6. Decay of representative γ-rays emitted by ^{230}Ra and ^{230}Ac in secular equilibrium. Lines are least squares fits to the data points.

References and Footnotes

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* On leave from Soreq Nuclear Research Center, Yavne, Israel.

** Present address: E. R. Squibb and Sons, Inc., Georges Road, New Brunswick, New Jersey, USA.

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