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

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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 $T_{1/2} = 30.0 \pm 1.5$ min, $Q_0 = 0.93 \pm 0.07$ MeV, and $\gamma$-rays of 157.6 and 162.1 keV. $^{196}$Os decays with $T_{1/2} = 35.0 \pm 0.1$ min and $\gamma$-rays at 126.1, 200.7, 207.0, 257.0, 315.3, 407.6, 522.2, and 628.9 keV. $^{230}$Ra decays with $T_{1/2} = 93 \pm 2$ min to $A = 122 \pm 3$ sec $^{230}$Ac; $\gamma$-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 (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 mA 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 ($E \geq 25$ MeV) is $3 \times 10^{11}$ n/cm²/sec for 100 mA of protons; it is well suited for producing sources of neutron-rich isotopes by $(n,2pn)$ and $(n,3pn)$ reactional-stage processes. 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 via reactions such as $(p,3p)$, there is usually serious interference from neutron-deficient isotopic nuclei that are produced in much greater abundance. Neutron irradiation improves the ratios$^4$ of neutron-rich to interfering neutron-deficient isotopes by factors of 10 to 100.

Results on the new nuclides $60$-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 96% $^{190}$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 OsO₄, precipitation of tungstic acid, scavenging of Re and Tc as oxides, and finally precipitation of tungsten α-benzoin oxide. The $^{196}$Os sources were made by $(n,2pn)$ from 50-300 mg targets of PtCl₄ (96% $^{198}$Pt). Purification was by distillation of OsO₄ into a NaOH solution, acidification to pH 5.5 with HCl, and then precipitation of OsO₄ with H₂S. Sources of $^{230}$Ra (and its short-lived $^{230}$Ac daughter) were made from 0.5-1.0 g targets of $^{232}$ThO₂ which were irradiated at the MEIN facility. The $^{232}$Ra was first coprecipitated with 1 mg BaO₂ 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 BaCl₂. 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 NH₄EDTA, 0.3 M NH₄NO₃, adjusted to pH 5.5; Ac³⁺ was eluted in the first column volume, the BaO₂ followed after about 6 column volumes, and then RaO₂ started to appear at 10 column volumes.

Measurements of the γ-ray spectra were with 50 cm³ Ge(Li) detectors (resolution of 1.7-1.9 keV at 1332 keV); data analysis was by means of INTRA$^6$ and CLSAN$^7$ 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 milling 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.](image-url)
The γ-ray spectra showed the known radiations9, of 190⁹⁸m decaying with $T_{1/2} = 30.0 \pm 1.5$ 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 190⁹⁸m (Fig. 2). Their intensities per decay of the 190⁹⁸m daughter are 0.39 and 0.11, respectively. The δ-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$_δ$ 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.80).

![Fig. 2. γ-ray spectra in region 150–200 keV of source containing 190⁹⁸m. Solid and dashed curves correspond to data taken at about 50 and 150 minutes after end of irradiation.](image1)

![Fig. 3. Proposed decay scheme for 190⁹⁸m and theoretical level structure of 190⁹⁸m.](image2)

### 3.2 Identification and Decay of 196⁹⁸m

The γ-ray spectrum from the osmium fraction, separated from irradiated 198⁹⁸Ir, was found to contain seven lines characteristic11) of 52-sec 196⁹⁸Ir. These decayed with $T_{1/2} = 35.0 \pm 0.4$ min. Ten more lines (Table 1) decayed with the same half life and were assigned to decay of 196⁹⁸m. The most precise lifetime measurement was from decay of the 196⁹⁸Ir δ-rays with $E = 1.5$ MeV (Fig. 4). Beta-rays from 196⁹⁸m with maximum energy of 440 ± 30 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.

![Fig. 4. Decay of various radiations from 196⁹⁸m and its daughter 196Ir in secular equilibrium. Lines are least squares fits to the data points.](image3)

### Table 1. Energies (keV) and intensities (per 100 disintegrations) from decay of 55.0-min 190⁹⁸m; preliminary values.

<table>
<thead>
<tr>
<th>$E_γ$</th>
<th>$I_γ$</th>
<th>$E_γ$</th>
<th>$I_γ$</th>
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<tr>
<td>126.1</td>
<td>4.9</td>
<td>315.3</td>
<td>2.6</td>
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<tr>
<td>200.7</td>
<td>0.6</td>
<td>407.6</td>
<td>6.1</td>
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<tr>
<td>207.0</td>
<td>2.5</td>
<td>522.2</td>
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<td>257.2</td>
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<td>308</td>
<td>0.4</td>
<td>628.9</td>
<td>1.6</td>
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### 3.3 Characterization of 230⁹⁸Ra and 230⁹⁸Ac

The discovery of 230⁹⁸Ra was reported12) by Jenkins and Seaborg who observed beta rays decaying 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 230⁹⁸Ac daughter which was presumed to be present in secular equilibrium. Chayawanathanurk, Herrmann, and Trautman13) produced 230⁹⁸Ac directly by irradiation of Th with 150-MeV bremsstrahlung. After rapid chemical separation of Ac they found two γ-rays, at 455 and 508 keV, and these were reported13) to decay with $T_{1/2} = 80 \pm 10$ sec. These γ-rays correspond to transitions from the well characterized14) 508-keV level in 230⁹⁸Th.

In the present work numerous γ-rays were found from decay of both 230⁹⁸Ra and 230⁹⁸Ac. The parent half-life was measured as 93 ± 2 min and that of the daughter as 122 ± 3 sec. Figure 9 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 230⁹⁸Ra and 230⁹⁸Ac are clearly established.

A minor interference is shown by the 10.6-keV component which results from the presence of 228Ra and its decay products in the separated Ra fraction.

In order to distinguish γ-rays following decay of the 230⁹⁸Ra parent from those following decay of the 230⁹⁸Ac 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 230⁹⁸Ac 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 230Ac. Figure 6 shows decay of some of these γ-rays.

The level scheme of 230Th has been previously determined from decay of 17.4-sec 230Ra and from inelastic scattering of deuterons 15 on 230Th. Of the 117 γ-rays associated with decay of 230Ac (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 230Ac itself no previous information is available, but analysis of the data from decay of 230Ra should yield a tentative decay scheme. Beta-gamma coincidence measurements on decay of 230Ra showed γ-rays of E_{max} ∼ 500 keV in coincidence with γ-rays of 63.0, 72.0, and 202.8 keV; thus Qγ ≥ 700 keV. In decay of 230Ac β-rays of E_{max} ∼ 1400 keV were found to be coincident with the 1243.9-keV γ-rays and Qβ is probably ≥700 keV.

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

<table>
<thead>
<tr>
<th>Eγ (keV)</th>
<th>Iγ (counts/min)</th>
<th>Eγ (keV)</th>
<th>Iγ (counts/min)</th>
<th>Eγ (keV)</th>
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References and Footnotes

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