RECEIVED 29 Jan 15

ACCEPTED 03 Jul 15

Seaborg's plutonium? A case study in nuclear forensics

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Abstract

Passive X-ray and gamma–ray analysis was performed on a curious sample from UC Berkeley's Hazardous Material Facility inventory, and the object was found to contain ^{239}Pu. No other radioactive isotopes were observed. The mass of ^{239}Pu contained in this object was determined to be 2.0 ± 0.3 μg. These observations are consistent with the identification of this object as containing the 2.77-μg PuO_2 (2.44 μg ^{239}Pu) sample produced in 1942 and described by Glenn Seaborg and his collaborators as the first sample of ^{239}Pu that was large enough to be weighed.

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I. INTRODUCTION

In early 1941, in Room 307 Gilman Hall on the University of California's Berkeley campus, Glenn Seaborg and his collaborators Arthur Wahl and Joseph Kennedy chemically separated element 94 from uranium that had been bombarded with 16-MeV deuterons at Berkeley's 60-inch cyclotron.[1,2] Not long after, Seaborg's group proposed the name "plutonium" for this new element. For the next year-and-a-half, Seaborg's group performed numerous studies of both the nuclear and chemical properties of plutonium using only trace amounts of this new material. In order to study plutonium in its pure form, an effort was, therefore, made to produce a "macroscopic" amount of plutonium by irradiating many kilograms of natural uranium with neutrons produced by bombarding beryllium with accelerated deuterons. Cyclotrons at both Washington University and Berkeley were utilized for this production. After chemically extracting the plutonium from the uranium and fission products, on September 10, 1942, at the Metallurgical Laboratory of the University of Chicago, Boris Cunningham and Lewis Werner succeeded in producing the first sample of pure PuO_2 that was large enough to be weighed on a newly developed microbalance.[2,3] The mass of the PuO_2 was determined to be 2.77-μg. Thus the ^{239}Pu mass contained in this sample would have been 2.44 microμgrams. This sample was preserved by sealing the platinum boat and oxide deposit inside a glass tube.[2] For more information on the discovery, production, and the use of plutonium during the Manhattan project, see Chapter 13 of "*The Making of the Atomic Bomb*" by Richard Rhodes.[4]

We are not aware of any written records as to what was then done with this first macroscopic sample of plutonium. However, as reported by Richard Strickert,[5] it was on display for a number of years at the

Lawrence Hall of Science in Berkeley, CA. In 2007, the Lawrence Hall of Science requested that the sample be removed by UC Berkeley's Environment, Health & Safety (EH&S) department to make way for more interactive exhibits. The small plastic box that EH&S removed from the Lawrence Hall had a label on it which read "First sample of Pu weighed. 2.7 μg." The sample was placed into safe and secure storage at the Hazardous Material Facility on the University of Berkeley's campus and was assigned the sample number of S338. In 2008, during a routine inventory, UC Berkeley Health Physicist, Phil Broughton, recognized the significance of the sample and contacted several museums to see if they were interested in having it on display. The Smithsonian expressed concern over the authenticity of the sample, which was required before they would consider the sample for display. The paper trail documenting this sample's history had been lost and so the question was what could be done to establish its authenticity as Seaborg's plutonium?

II. EXPERIMENTAL PROCEDURE

In June ~~of~~ 2014, the Nuclear Engineering Department at UC Berkeley became aware of the sample and requested the opportunity to study it, in order to determine its origins. In an effort to preserve the possible historical significance of the sample, it was quickly decided that the box should not be opened and that only non-destructive testing should be performed on it. After examining and photographing the sample in its plastic box (see Fig. 1), it was placed approximately 6.3 mm away from the front face of a 36-mm diameter by 13-mm thick planar germanium detector. This detector is equipped with a thin beryllium window, allowing for detection of low-energy gamma rays and X-rays. The detector was shielded with 1.27 cm of copper and 5–10 cm of lead. (The detector and its shielding are also shown in Fig. 1.) The plastic box containing the sample of interest was counted for 21.2 h~~ours~~. The box was then removed and a background spectrum was collected for 21.3 h~~ours~~.

~~———~~Figures 2~~,~~ ~~3, and~~ 4 illustrate the relevant portions of the background-subtracted spectrum we obtained from the sample. (All energies are in keV.)~~.~~ All of the gamma rays observed can be attributed to the decay of ^{239}Pu (Ref. 6). The 38.7-, 51.6-, 56.8-, and 129.3-keV lines are the most intense gamma rays emitted in the decay of ^{239}Pu and are the only ones that would be expected with our planar Ge detector. In order to search

for higher energy gamma rays from other radionuclides, the box containing the sample was placed up against the front face of a shielded 85% relative efficiency coaxial germanium detector and counted for approximately 24 h~~ours~~. No evidence of any additional gamma rays up to 3 MeV was observed.

One concern about the authenticity of the sample is the possibility that it was produced in a reactor. We checked this by gauging the amount of ^{241}Pu in the sample. Spent nuclear fuel discharged from power reactors typically contains plutonium with ^{241}Pu/^{239}Pu isotopic ratios in the range of 0.045–0.23 (Ref. 7). Even weapons-grade plutonium contains ^{241}Pu/^{239}Pu ~ 0.005 (Ref. 8). Because the half-life of ^{241}Pu is 14.4 y~~ea~~r~~s~~ and its decay product is ^{241}Am, the abundance of which grows over time, plutonium produced in a reactor should have measurable amounts of ^{241}Am. However, from Fig. 3, it can be seen that we observed no evidence of the 59.5-keV gamma ray produced by the decay of ^{241}Am. Our data allow~~s~~ us to set an upper limit on the isotopic abundance ratio of ^{241}Am/^{239}Pu to be less than 2.3 ×~~x~~ 10^{-7}. Thus, the plutonium contained in this sample was most likely *not* produced in a reactor, but in a low-neutron-fluence environment. This is consistent with Seaborg's description of producing the plutonium by irradiating many kilograms of natural uranium with neutrons produced by deuteron bombardments of beryllium.[3]

In order to determine the mass of ^{239}Pu contained in this sample, we measured the efficiency of our detector using calibrated sources of ^{57}Co, ^{137}Cs, and ^{241}Am. These sources provide X-ray and gamma-ray lines at 26, 32, 36, 59, 122, and 136 keV, thus covering the energy range of interest. (All necessary nuclear data were~~as~~ taken from Ref. 6.) We carefully measured the distance from the sample attached to the end of the plastic rod in the sample, to the front face of our detector and then placed our calibration sources at this same distance. Gamma rays emitted from the sample had to pass through the 0.63-cm thick wall of the plastic box in which it is contained. In order to account for the attenuation this produced, we placed a 0.63-cm thick block of polyethylene between our sources and the detector. We extracted the peak areas of the 38.7-, 51.6-, and 129.3-keV lines from the spectrum shown in Fig. 2, and then determined the sample mass from each line. The results were then averaged to establish the mass of ^{239}Pu contained in the sample to be 2.0 ± 0.3 μg. Seaborg and Cunningham and Werner stated that the first weighed sample contained 2.77-μg of PuO_2 with no uncertainty given. This would imply a ^{239}Pu mass of 2.44-μg. These authors also stated that the first weighed sample of

plutonium was evaporated and then converted to an oxide on a platinum boat.[2,3] The low energy portion of our spectrum shown in Fig. 4 contains a peak at 9.4 keV that is consistent with being the L_α X-ray from platinum. Other still lower energy peaks are most likely germanium K X-ray escape peaks produced by interactions of higher energy X-rays near the surface of our detector.

III. CONCLUSIONS

Passive X-ray and gamma-ray analyses were performed on UC Berkeley's Environmental Health and Safety sample S338. The object was found to contain ^{239}Pu. No other radioactive isotopes were observed. The mass of ^{239}Pu contained in this object was determined to be 2.0 ± 0.3-μg. Evidence of plutonium L X-rays was also seen. While not 100% conclusive, these observations are consistent with the identification of this object being the 2.77-μg PuO_2 sample (2.44-μg ^{239}Pu) described by ~~Glenn~~ Seaborg and his collaborators as the first sample of ^{239}Pu that was large enough to be weighed. More detailed measurements such as mass spectrometry or chemical testing could be performed but that would require opening the plastic box and sacrificing some of the plutonium-containing material. It is hoped that in the near future a commemorative display will be set up on the campus or in Seaborg's old office in Gilman Hall where the discovery of plutonium was made. This sample would be a fitting item to showcase in such a setting.

ACKNOWLEDG~~E~~EMENT~~S~~

This work~~ed~~ was supported in part by the U.~~-~~S. Department of Energy National Nuclear Security Administration under Award N<u>o.</u>~~umber~~ DE-NA0000979.

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[B]Please provide last page number in Ref. 5.

~~Figure Captions~~

Fig. 1. (a) Outside of sample box with labels; (b) side view showing sample attached to plastic rod; (c) planar Ge detector with copper and lead shielding.

Fig. 2. Background-subtracted spectrum observed from sample S338. All of the labeled peaks are X-rays and gamma-rays attributable to the decay of ^{239}Pu.

Fig. 3. Expanded region of the spectrum observed around 50 keV. All of the labeled peaks are produced by the decay of ^{239}Pu. No evidence of the 59.5-keV gamma-ray produced by the decay of ^{241}Am was observed.

Fig. 4. Low-energy portion of the spectrum showing uranium L X-rays produced by the decay of ^{239}Pu. The small peak at 9.4 keV is consistent with being an L_α X-ray of platinum. The peaks at lower energies are likely to be Ge escape peaks produced by the higher energy X-rays.