Preparation of Pu²³⁹ Sources

by

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INTER-OFFICE MEMORANDUM

SAVANNAH RIVER PLANT

Key WordsPu²³⁹

Source

Preparation ✓

DPSP-88-272-84

August 5, 1988

TO: A. GIBBS, 707-H

FROM: H. P. HOLCOMB, 772-F *HPH*PREPARATION OF Pu²³⁹ SOURCESINTRODUCTION AND SUMMARY

At your request, the Separations Technology Laboratory has prepared four sources to be used for calibrating a waste assay system (Passive/Active Neutron Assay) in Building 724-8G (Burial Ground).

The four sources contain 0.5, 0.1, 0.05, and 0.01 grams Pu²³⁹, respectively. The sources were prepared using aliquots from a single solution provided by the Quality Control (QC) group of Laboratories Department. The solution contained weapons-grade plutonium dissolved in nitric acid. Final solution acidity was 3M. Coulometry had been used to obtain a total plutonium content per unit volume. The weight percent of the plutonium isotopes present was obtained via mass spectrometry.

Pipettes, calibrated by the QC group, were used to prepare the sources. An average Pu²³⁹ content of each source was calculated from delivery volumes, together with the average Pu²³⁹ content of the solution from coulometry plus mass spec data. Pipette delivery precision and the precision of the total quantity of total plutonium from coulometry were used to calculate an error bar for each source. Mass spec data, being more precise, was not a limiting factor; so, it was not included in the precision calculation.

The resulting Pu²³⁹ contents, in grams, for the four sources were: 0.5011 ± 0.0023, 0.1001 ± 0.0004, 0.0501 ± 0.0002, and 0.0099 ± 0.0000, respectively.

The sources are primarily contained in 4-dram glass vials with the liquid immobilized by fine-particle alumina. The screw cap liner for the primary container is a polyethylene cone to ensure proper sealing. The secondary container is a 1" dia. X 3" high plastic vial with a polyethylene friction-fit cap, taped. These

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are contained in a clean-on-the-outside plastic bag, taped. Each plastic bag is placed in a one-half pint ice cream carton, the final container.

EXPERIMENTAL

Stock Plutonium Solution

Approximately 12 mL of a weapons-grade plutonium nitrate solution in 3M HNO₃ was obtained from the QC Group of Laboratories. This solution was placed in the Sep Tech Lab's glove box where the sources were prepared. Coulometric analysis of the solution yielded a total plutonium content of 85.423 ± 0.214 g ($\pm 0.25\%$) per liter. The precision of this measurement creates an error bar of ± 2 parts in 850, a significant contribution to the precision of the final plutonium value of each source.

Mass spectrometric analysis of this solution produced the following isotopic distribution:

	<u>wt %</u>
Pu ²³⁸ -	0.014
Pu ²³⁹ -	93.710
Pu ²⁴⁰ -	5.780
Pu ²⁴¹ -	0.438
Pu ²⁴² -	0.057

According to J. Satkowski of Labs, the precision of the Pu²³⁹ measurement is $\pm 0.06\%$, or an error bar of ± 6 parts in 9000. Because of its relatively low value, this number is not a limiting factor and, therefore, was not included in precision calculations.

The quantities of solution needed to prepare the sources were determined as follows. The source solution was considered to be 93.710 weight percent Pu²³⁹ and to contain 85.423 g total plutonium per liter. Therefore, each liter contained 80.05 g Pu²³⁹/L, on average; or each milliliter, 0.08005 g Pu²³⁹.

For a 0.5 g source, 6.25 mL of the stock solution is needed (0.5 g Pu²³⁹/0.08005 g Pu²³⁹ per milliliter). Likewise, for 0.1 g Pu²³⁹, 1.25 mL would be needed. For the sources one-tenth as large, 625 μ L and 125 μ L would be needed, respectively, for the 0.05 g and 0.01 g quantities.

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Pipettes Used

Three Rainin "Pipetman" pipettes that use disposable tips were calibrated by Labs' QC group. These pipettes were used in preparing the sources. Calibration data, furnished by Labs, is as follows:

<u>Pipette #</u>	<u>Indicated Delivery Volume, μL</u>	<u>Bias, %</u>	<u>Actual Delivery Volume, μL</u>	<u>Repetitive Delivery Precision, $\pm$%</u>
17016	125	-0.63	124	0.16
11129	625	+0.22	626	0.09
10656	1000	+0.19	1002	0.03

The 1000- μ L pipette had excellent precision for a single delivery to each of multiple containers. For the 0.5 g Pu²³⁹ source, however, it was used 6 times to deliver to a single container. So, the error bar, due to use of the 1000- μ L pipette only, for the 0.5 g source becomes $\pm 0.18\%$, a limiting factor. A calibrated 5000- μ L pipette was unusable because the liquid leaked due to its density.

Preparation of Sources

Cold testing was performed in the laboratory to determine the best containment for the sources because they were initially liquid. Several solids were tested as immobilizing agents for 3M HNO₃, the source solution matrix. These agents included Oil Dri (ground clay), alumina (adsorption, 80-200 mesh), and Drierite® (CaSO₄). The best agent for sorbing the liquid was alumina.

The initial container for the plutonium nitrate solution and alumina was a 4-dram glass vial with a screw cap containing a polyethylene cone liner. To begin with, the vial was filled approximately one-quarter full with alumina for each source.

For the 0.5 g Pu²³⁹ source, six successive pipettings with the 1000- μ L pipette were made to a vial followed by two pipettings with the 125- μ L pipette. Then, alumina was slowly added, in portions, to immobilize the liquid.

For the 0.1 g Pu²³⁹ source, one pipetting with the 1000- μ L pipette was made plus two with the 125- μ L pipette.

For the 0.05 g Pu²³⁹ source, one pipetting with the 625- μ L pipette was made.

For the 0.01 g Pu²³⁹ source, one pipetting with the 125- μ L pipette was made.

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The vials were then loosely capped and allowed to stand for 24 hours to allow liquid sorption by the alumina. Finally, each vial was filled to its shoulder with alumina and capped tightly. So, each vial contained approximately the same quantity of alumina, but different quantities of liquid.

Packaging of Sources

Each tightly-capped glass vial was placed in a 1" dia. X 3" high plastic vial that was capped with a polyethylene press-fit cap that fit snugly against the cap of the glass vial. The plastic cap was taped securely to the vial with plastic tape. The doubly-contained sources were then removed from the glove box by putting each in a plastic bag, kept "clean" on the outside. The bag top was twisted and taped securely.

Each containment package was placed on its side in the approximate center of the bottom of a $\frac{1}{2}$ -pint ice cream carton. Four "Kleenex" tissues were used in each carton to minimize package shifting. The carton top was then put in place and securely taped. A broad-tipped felt pen was used to inscribe each package's contents. Finally, the packages were given to 772-F HP for survey. Their radiation survey tag is on the bottom of each package. The largest source, 0.5 g Pu^{239} , radiated 5 mrad/5 mr per hour at contact.

CALCULATION OF Pu^{239} CONTENT OF SOURCES AND ERROR BARS

0.5 g Source

Six pipettings with the 1000- μL pipette produced a total of (6×1002) or 6012 μL with an error bar of $(6 \times 0.03\%)$ or $\pm 0.18\%$ (or $\pm 11 \mu\text{L}$). This yields $6012 \pm 11 \mu\text{L}$ from the six 1000- μL pipettings.

Two pipettings with the 125- μL produced a total of (2×124) or 248 μL with an error bar of (2×0.16) or $\pm 0.32\%$, (or $\pm 1 \mu\text{L}$). This yields $248 \pm 1 \mu\text{L}$ from the two 125- μL pipettings.

The average Pu^{239} content of the 0.5 g source is 6.260 mL X 0.08005 g $\text{Pu}^{239}/\text{mL}$ or 0.5011 g, with an error bar from pipetting alone of $(\pm 11 \mu\text{L} + \pm 1 \mu\text{L})$ or $\pm 12 \mu\text{L}$, which is equivalent to 0.001 g Pu^{239} . Therefore, from pipetting, the 0.5 g source has a Pu^{239} content of 0.5011 ± 0.0010 g.

However, the original Pu stock solution had a precision of $\pm 0.25\%$ in the coulometric analysis of its total plutonium solution. This component must also be factored into all calculations of the sources' error bars. So, the 0.5011 g Pu^{239} has an error

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bar of $\pm 0.25\%$ (or 0.0013 g) from the uncertainty in the coulometric assay of the stock solution.

Thus, the 0.5 g Pu^{239} source contains $0.5011 \pm (0.0010 + 0.0013)$ or $0.5011 \pm 0.0023 \text{ g Pu}^{239}$.

0.1 g Source

For this source, the $1000\text{-}\mu\text{L}$ pipette was used once producing $1002 \mu\text{L} \pm 0.03\%$ or $1002 \mu\text{L} \pm 0.3 \mu\text{L}$.

The $125\text{-}\mu\text{L}$ pipette was used twice producing the same quantity as was produced by it for the 0.5 g source, $248 \pm 1 \mu\text{L}$.

So, from pipetting, the total quantity of Pu^{239} in the 0.1 g source was $1.250 \pm 0.0013 \text{ mL} \times (0.08005 \text{ g Pu}^{239}/\text{mL})$ or $0.1001 \pm 0.0001 \text{ g Pu}^{239}$.

From the $\pm 0.25\%$ uncertainty in the total Pu content from coulometry comes an error bar of $0.1001 \pm 0.25\%$, or $0.1001 \pm 0.00025 \text{ g Pu}^{239}$.

Therefore, the 0.1 g Pu^{239} source contains $0.1001 \pm (0.0001 + 0.00025)$ or $0.1001 \pm 0.0004 \text{ g Pu}^{239}$.

0.05 g Source

For this source, only one pipetting was made with the $625\text{-}\mu\text{L}$ pipette. This yielded $626 \mu\text{L} \pm 0.09\%$ or $626 \mu\text{L} \pm 1 \mu\text{L}$. From pipetting, the quantity of Pu^{239} in this source is $0.626 \pm 0.001 \text{ mL} \times (0.08005 \text{ g Pu}^{239}/\text{mL})$ or $0.0501 \pm 0.0001 \text{ g Pu}^{239}$.

From the coulometric uncertainty of $\pm 0.25\%$ in the total Pu content of the stock solution comes an error bar of $\pm 0.0001 \text{ g Pu}^{239}$.

Therefore, the 0.05 g Pu^{239} contains $0.0501 \pm (0.0001 + 0.0001) \text{ g Pu}^{239}$ or $0.0501 \pm 0.0002 \text{ g Pu}^{239}$.

0.01 g Source

For this source, only one pipetting was made with the $125\text{-}\mu\text{L}$ pipette. This yielded $124 \mu\text{L} \pm 0.16\%$ or $124 \mu\text{L} \pm 0.2 \mu\text{L}$. From pipetting, the quantity of Pu^{239} in this source is $0.124 \pm 0.000 \text{ mL} \times (0.08005 \text{ g Pu}^{239}/\text{mL})$ or $0.0099 \pm 0.0000 \text{ g Pu}^{239}$.

From the coulometric uncertainty of $\pm 0.25\%$ in the total Pu content of the stock solution comes an error bar of $\pm 0.0000 \text{ g Pu}^{239}$.

Therefore, the 0.01 g Pu^{239} contains $0.0099 \pm (0.0000 + 0.0000) \text{ g Pu}^{239}$ or $0.0099 \pm 0.0000 \text{ g Pu}^{239}$.

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HPH/h