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Preparation of Working Reference Materials: Calcined Waste Recovery Products Containing Uranium or Plutonium

University of California

LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos, New Mexico 87545

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J. E. Rein
G. R. Waterbury

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FOREWORD

This report provides guidance for preparing working reference materials of calcined waste recovery products containing uranium or plutonium. These materials are used to calibrate and maintain quality control surveillance of chemical methods of analysis for uranium and plutonium content and isotopic distribution measurements. Similar reports have been issued for plutonium nitrate solution (LA-NUREG-6348, NUREG-0118), uranium nitrate solution (NUREG-0253), plutonium oxide (LA-NUREG-7260, NUREG/CR-0061), and mixed oxide (LA-7322, NUREG/CR-0139), and a report is in progress for uranium oxide.

CONTENTS

PREPARATION OF WORKING REFERENCE MATERIALS: CALCINED WASTE RECOVERY PRODUCTS CONTAINING URANIUM OR PLUTONIUM

by

J. E. Rein and G. R. Waterbury

ABSTRACT

Procedures are presented for preparing calcined waste recovery products that have assigned values of uranium and plutonium contents and isotopic distributions. These working reference materials are used to calibrate and maintain measurement control surveillance of chemical methods for analyzing plant process materials. Statistical treatments are discussed that provide a measure of the reliability of working reference materials in applications to nuclear material accountability and safeguards.

1. INTRODUCTION

Use of calcination processes is increasing to recover uranium and plutonium from nuclear fuel-cycle scrap materials. Accountability and safeguards require accurate and precise determinations of the uranium and plutonium contents and isotopic distributions in the process products. Therefore, well-characterized materials must be used for calibration and measurement control surveillance of the analysis methods.

This report is part of a series,* prepared at the request of the Office of Standards Development of the Nuclear Regulatory Commission (NRC), that describes the preparation of materials defined as working reference materials (WRM), which are used to calibrate chemical analysis methods that determine uranium and plutonium contents and isotopic distributions. We discuss procedures designed to produce calcined waste recovery product WRMs, establishment of their uranium and plutonium contents and isotopic distribution values, and their packaging.

2. GENERAL CONSIDERATIONS GOVERNING PREPARATION OF A SOLID URANIUM OR PLUTONIUM WRM

A WRM must be prepared so that its assigned values are based on a certified reference material (CRM). This provides measurement compatibility through the national or international measurement system. Definitions of reference materials are given in Table I. Other important characteristics of a WRM are stability, chemical similarity to a process material, and reliability of the assigned content and isotopic values consistent with the reliabilities required of the analysis methods to which it pertains.

An ideal WRM would be an appropriate CRM, provided its chemical composition, including metal and nonmetal impurities, matched that of the special nuclear material in process and its availability in large quantities at reasonable cost was assured. Neither provision is met by available CRMs. Furthermore, CRMs are too valuable a resource to be expended for routine applications. An exception is their application to uranium and

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*The Idaho Office of Exxon Nuclear and the Los Alamos Scientific Laboratory are preparing reports for uranium-containing and plutonium-containing WRMs (Refs. 1-5).

TABLE I

TYPES OF REFERENCE MATERIALS

Type	Definition	Example
Reference Material (RM)	A material or substance one or more properties of which are sufficiently well established to be used for the calibration of an apparatus or for the verification of a measurement method [ISO-Guide 6-1977(E)]. A generic class of characterized, homogeneous materials, produced in quantity and having one or more physical or chemical properties experimentally determined within stated measurement uncertainties.	Any or all of the materials listed below.
Certified Reference Material (CRM)	An RM accompanied by, or traceable to, a certificate stating the property value(s) concerned (and its associated uncertainty), issued by an organization, public or private, which is accepted as technically competent [ISO-Guide 6-1977(E)].	Any PCRM or SCRM (see below).
Primary Certified Reference Material (PCRM)	A CRM of high purity, possessing chemical stability or reproducible stoichiometry, and generally used for the development/evaluation of reference methods and for the calibration of RMs. PCRMs are certified using the most accurate and reliable measurement methods available, consistent with end-use requirements for the RM	Standard Reference Materials of the National Bureau of Standards (NBS SRMs), reference materials of the International Atomic Energy Agency (IAEA) bearing the IAEA classification "S," and any other PCRM.
Secondary Certified Reference Material (SCRM)	An RM characterized relative to a primary certified reference material, generally used for development/evaluation of field measurement methods or for day-to-day intralaboratory comparison programs. SCRMs may be less pure or less stable than PCRMs, depending on their intended use. Accuracy required of the certifying measurements also depends on intended use.	Some reference materials available from the US DOE, New Brunswick Laboratory. IAEA reference materials classification "R."

TABLE I (cont)

Type	Definition	Example
Working Reference Material (WRM)	An RM characterized relative to PCRM or SCRM, usually for use within a single laboratory or organization. WRMs are generally used to assess the level of performance of measurements on a frequent (e.g., daily) basis. WRMs are usually prepared from material typical of a given process [previously known as Working Calibration and Test Materials (WCTMS)].	Process stream materials and any RM prepared according to applicable NUREG documents (Refs. 1-5).

plutonium isotopic distribution measurements by thermal ionization mass spectrometry, which requires very small samples. Many laboratories find it practical to use separate WRMs for content and isotopic distribution measurements.

Several options exist to establish the elemental content(s) and/or isotopic composition(s), depending on the starting material and the preparation process. Two independent measurements that provide the same average value within stated uncertainties are required for elemental content. In lieu of one of the measurements, a calculated "makeup" value can be used when the WRM is prepared using a weighed quantity of a characterized starting material under complete recovery conditions. However, even the uranium or plutonium content of a WRM prepared from a CRM under complete recovery conditions must be confirmed by analysis to ensure that the preparation uncertainty is within the uncertainty limits of the CRM. When the makeup value cannot be calculated, the elemental content is to be established using two different analytical methods by one laboratory, or by two laboratories, preferably using different methods. All operations of both methods, including dissolution of the material and any chemical separation treatments, should be completely independent.

When an isotopic distribution CRM is used as a starting material for a WRM, the certificate values can serve as makeup values. The values should be verified by mass spectrometric analysis unless stringent precautions are taken to prevent contamination of the WRM.

As stated previously, WRMs must be prepared so that their uranium and plutonium contents and isotopic distribution values are related directly to CRMs. This is achieved by using CRMs to calibrate the methods used to analyze starting materials and WRMs. These calibrations are to be concurrent with the analysis of the starting material or WRM; the number of replicate measurements of the CRM and of the starting material or WRM are to be equal. The CRM for elemental content characterization normally is a solution prepared on a weight basis using weights traceable to NBS (National Bureau of Standards) mass references. Its chemical composition is to simulate that of the starting material or WRM with respect to uranium or plutonium content and impurity element concentrations. However, because adding impurity elements to a CRM solution can adversely affect its stability, a portion of impurity solution can be added to each individually delivered CRM portion at the time of the analysis.

The selection of methods used to determine uranium and plutonium contents of starting materials and WRMs is based on high-precision capability and freedom from impurity effects. The first qualification is important because the required number of replicate measurements decreases as the method's precision improves. Freedom from impurity effects increases accuracy. When two methods are used for the characterization, the impurity effects of the methods should be different to decrease inaccuracies that can be caused by undetected impurities and by slight differences between the impurity element composition of the CRM and that of a starting material or WRM.

A major operation in a WRM preparation is the statistical treatment of the characterization analysis results. The statistical treatments vary depending on the starting material reliability and the characterization analysis options (Sec. 6).

Radioactive decay of plutonium isotopes and growth of americium and uranium daughters affect the uranium and plutonium elemental contents and isotopic distributions as discussed in Sec. 7.

3. PREPARATION OF CALCINED WASTE RECOVERY PRODUCT WRMs

Three methods are discussed. The first uses a plant product as the starting material. The second uses a blend of solid components, and the third uses a blend of solid and solution components. Because of the wide variety of calcined waste recovery products, only general guidelines for preparing WRMs are given.

The final operations include grinding, mixing, and sieving to promote homogeneity, and heating at 900°C to produce oxides of uranium, plutonium, and matrix components that are quite stable and reheatable to the same stoichiometric form. The analyses of the WRMs to establish their uranium or plutonium contents and their use thereafter are made on 900°C-heated portions.

3.1 Plant Product as Starting Material

The batch should be typical of the plant product, and its weight should be maintained after heating at 900°C. The selected quantity of material is in a tared, preferably quartz or platinum, container and is heated at 900°C in an air-atmosphere muffle furnace. After cooling, the product is weighed. Cycles of heating and weighing are repeated until a constant weight is obtained that has an uncertainty consistent with the objective uncertainty of the WRM (Sec. 5.3). Then, the material is ground and sieved through a US standard series 200-mesh screen. The unsieved fraction is reground and resieved until only ungrindable components remain. The sieved fraction is mixed in a shaker mill or a V-blender until it is homogeneous.

3.2 Blend of Solid Components

A makeup value can be calculated for this WRM preparation when a uranium or plutonium oxide that has a characterized elemental content at 900°C is used as one of the starting materials, complete recovery is maintained in the mixing operations, and weights of all materials and all heated products are measured.

Usually, a fine powder mixture of components that simulates the plant material, termed matrix material, is prepared and uranium or plutonium oxide is blended into it. To create homogeneous blends that contain low levels (several percent or less) of uranium and/or plutonium, an intermediate blend is prepared with a concentration of uranium or plutonium 10 to 20 times that of the final blend. The intermediate blend, a relatively small quantity, can be prepared by manually grinding the matrix material and uranium oxide or plutonium oxide in a mortar to produce a very homogeneous powder mixture. The final blend is prepared by mixing portions of the intermediate blend and more matrix material in a shaker mill or a V-blender until the mixture is homogeneous. The final blend is in a tared, preferably quartz or platinum, container and is heated at 900°C in an air-atmosphere muffle furnace. After cooling, the product is weighed, and the heating and weighing cycle is repeated to obtain a constant weight that has an uncertainty consistent with the objective uncertainty of the WRM (Sec. 5.3).

3.3 Blend of Solid and Solution Components

A makeup value can be calculated for this WRM preparation when the solution phase contains a characterized concentration of uranium or plutonium, complete recovery is maintained in the mixing operations, weights of all materials and all heated products are measured, and the filtrate is analyzed for uranium or plutonium.

This method is patterned after that used at the New Brunswick Laboratory for preparing calcined ash WRMs.[6] An acidic solution of soluble components that includes uranium or plutonium is added to an acidic suspension of insoluble components including cellulose and the mixture is stirred vigorously. (The amounts of insoluble and soluble components are selected to simulate the composition of the plant material.) While vigorously mixing, a twofold excess of ammonium hydroxide is added and the mixture is digested on a steam bath for 2 hours. After cooling, the mixture is filtered and the filter cake is washed with dilute ammonium hydroxide. It is transferred to a tared quartz or platinum container and dried at 110°C for 18 hours. The container and dried material are placed in a cold, air-atmosphere muffle furnace, heated gradually to 900°C, and maintained at this temperature for about 16 hours. After cooling, the product is weighed, and the heating and cooling cycle is repeated until a constant weight is obtained that has an uncertainty consistent with the objective uncertainty of the WRM (Sec. 5.3). The material is ground and sieved through a US standard series 200-mesh screen until all the material is sieved. It is mixed in a shaker mill or a V-blender until the mixture is homogeneous.

4. PACKAGING

As soon as possible after preparation, the WRM should be distributed into containers to avoid contamination of the whole batch in one container. Randomly selected containers then provide the portions for characterization analyses. A recommended quantity per container is that required for 10 analyses. Separate packaging of a WRM characterized for uranium or plutonium elemental contents and for isotopic distributions is unnecessary; aliquots from one dissolved portion can serve for both measurements.

An inert atmosphere is unnecessary and various types of containers can be used, including a glass jar with a plastic poly-cone seal in a screw-on lid, a glass vial with a plastic insert or a plastic-covered cardboard insert in a screw-on lid, and a plastic (polyethylene) vial with a plastic slip-on, slip-in, or flip lid. A secondary container that is contamination-free on its outer surface is recommended for plutonium-containing WRMs to facilitate transfers among laboratory areas.

The primary container should be properly labeled, including its chemical composition or a source that provides chemical composition, date prepared, responsible person, assigned values, intended application, hazards and handling precautions, storage stability limit, description of packaging, recommended pretreatments, and details of preparation and characterization.

5. ESTABLISHMENT OF WRM VALUES

Calcined waste recovery products often contain refractory components—and this characteristic can be expected for WRMs prepared from plant materials. Heating in air at 900°C, recommended in all three methods given in Sec. 3, will produce PuO_2 that does not dissolve readily in acids, including mixtures of HNO_3 and HF. Refractory compounds may be produced from various matrix components in which the uranium and plutonium oxides may be occluded. Also, each portion taken for uranium- or plutonium-content analysis is reheated at 900°C to constant weight to produce a reproducible stoichiometric form.

The dissolution of 900°C-calcined plutonium oxide and of many other refractory element oxides in acids at ambient pressure is slow and often incomplete. Pressurized acid reactions at higher temperature, including use of a sealed quartz tube,[7] a sealed reflux tube,[8] and a Teflon-container metal shell,[9] promote complete dissolution. A mixture of H_2SO_4 and HNO_3 containing $(NH_4)_2SO_4$ to raise the boiling point has been recommended for dissolving high-fired plutonium oxide.[10] Fusions using molten salts such as sodium bisulfate[11] also are effective. A common treatment is elevated pressure, acid reactions followed by filtration and a molten salt fusion of the filter cake.

Each solution is inspected carefully for residue including fine suspension shown by the Tyndall effect. Options that apply when a residue or suspension is present are further dissolution treatment (and reinspection), filtration followed by measurement of the plutonium on the filter, or rejection of the WRM portion. The filtration must give complete recovery of the solution and the residue. A recommended filter is a 0.45-μm-pore size, acid-resistant plastic membrane, such as Gelman Vinyl Metricel.

5.1 Uranium or Plutonium Content

A WRM prepared using a CRM as the starting material (not recommended as discussed in Sec. 2), which is prepared under conditions designed to give complete recovery, may be characterized for its uranium or plutonium content by one method. Statistical tests compare the result to the makeup value. An optional but less desirable mode is to characterize the WRM using two different methods each for uranium or plutonium followed by statistical comparisons of the results. This foregoes use of makeup values, which is the justification for using a CRM as a starting material. The same options apply to a WRM prepared from starting materials other than CRMs, except their uranium or plutonium contents must be established by two different methods each if makeup values are computed. When ≥99.95% pure metal or compound is used, one of the two methods can be the determination of total metallic and nonmetallic impurities, then the uranium or plutonium content is computed by subtracting the sum of the impurities, on a percentage basis, from 100. There are no options for a WRM prepared from an uncharacterized starting material, such as plant material. The uranium or plutonium content values are to be determined by two different methods each, followed by appropriate statistical tests of comparison.

As discussed in Sec. 2, the selection of uranium and plutonium determination methods is based on high-precision capability and freedom from interference effects caused by impurities present in starting materials or WRM. When two methods are used, the effects of the impurities should be as different as is practical.

Each time a starting material or WRM is analyzed, the analytical method is to be calibrated using the same number of aliquots of a CRM that simulates the WRM concentrations of uranium, plutonium, and impurity elements. A suggested practice to attain simulation of impurity elements is to add them as a solution to the delivered aliquots of the CRM. The results for the starting material or WRM are computed relative to the concurrent results obtained for the CRM. This means that impurity contents of starting materials and WRMs must be known.

In general, because metal ions (rather than nonmetal ions) interfere with uranium and plutonium analytical methods, techniques that provide a multielemental metal analysis are used, such as emission spectrography, spark source mass spectrometry, and x-ray fluorescence.

Electrotitrimetric methods are used for determining uranium and plutonium. Recommended methods[11,12] for the determination of uranium content are controlled-potential coulometry at a mercury cathode with $0.5\underline{M}$ H_2SO_4 electrolyte and potentiometric titrimetry using Fe(II) reduction in phosphoric acid medium followed by Cr(VI) titration; and for the determination of plutonium content, controlled-potential coulometry at a platinum electrode with $0.5\underline{M}$ H_2SO_4 electrolyte and amperometric titrimetry with Fe(II) in a $2.5\underline{M}$ H_2SO_4 medium following Ag(II) oxidation.

5.2 Uranium or Plutonium Isotopic Distributions

Uranium and plutonium isotopic distributions are determined by thermal ionization mass spectrometry after separation from each other and from americium by anion exchange in hydrochloric acid medium. The separation method is reported in Refs. 11 and 13; Ref. 11 also details mass spectrometric measurements. However, low levels (<0.1%) of ^{238}Pu can be determined more accurately by alpha spectrometry.

An isotopic distribution CRM used as a WRM does not require verification provided stringent precautions are taken to prevent contamination. We recommend that the dissolved CRM be apportioned to a series of containers and that a container's use be limited to about a month. Uranium and plutonium isotopic distribution WRMs should not be combined because uranium daughters grown into the plutonium WRM will alter the uranium isotopic distribution so that the certified values no longer will apply.

A WRM prepared with a material other than an isotopic distribution CRM is to be characterized by analysis. The analytical method is to be calibrated at the time the WRM is analyzed, using uranium or plutonium CRMs with isotopic distributions closest to those of the uranium or plutonium in the WRM. Like the uranium- or plutonium-content method calibrations, the number of WRM and CRM portions analyzed are to be equal. All portions are to undergo the same chemical treatments immediately before the mass spectrometric measurements. Because the accuracy of a mass spectrometric measurement is affected almost solely by impurity elements that form charged ions in the uranium and plutonium mass range, low-mass impurity elements present in the WRM do not have to be added to the CRM portions. The recommended separation procedure, anion exchange in hydrochloric acid medium, provides uranium and plutonium fractions free of each other, americium, and most impurity elements that can cause unstable emission.

5.3 Criteria Governing Number of Portions Analyzed

The equal number of portions of the starting material or WRM and the concurrently analyzed CRM is selected to produce a desired limit of error (LE)* for the values of the uranium and plutonium contents and isotopic distributions assigned to a WRM. The LE is a function of the precision required of these values in the plant material to which the WRM applies.

The LE to be associated with the uranium or plutonium concentration or isotopic distribution of a WRM is selected to be ≤1/3 of the LE associated with the plant material if known. This permits a calculation of the number of portions to be analyzed.

The RLE associated with the uranium or plutonium content or isotopic distribution is

$$\mathrm{RLE} = 100\,\mathrm{LE}/M\,, \tag{1}$$

in which M is the measured average uranium or plutonium content or isotopic distribution

$$\mathrm{LE} = 2\,S_M \tag{2}$$

$$= 2\,S_1/\sqrt{n}, \tag{3}$$

where S_M is the standard deviation of the mean, S_1 is the standard deviation for a single measurement of the analytical method in use in the laboratory preparing the WRM, and n is the number of replicate aliquots analyzed concurrently for both the WRM and CRM. Combining Eqs. (1) and (3) gives

$$\mathrm{RLE} = 100(2\,S_1)/M\sqrt{n}; \tag{4}$$

and solving for n,

$$n = 4(100\,S_1/M)^2/\mathrm{RLE}^2\,, \tag{5}$$

because 100 S_1/M is defined as the relative standard deviation in percentage for a single measurement (RSD_1),

$$n = 4\,\mathrm{RSD}_1^2/\mathrm{RLE}^2\,. \tag{6}$$

An example will demonstrate how n decreases with improving measurement precision. Assume that the assigned RLE for the plutonium content of the calcined waste recovery product in a material balance area is 0.45%. The RLE for the plutonium content of the WRM is ≤(1/3)(0.45%) = ≤0.15%. Two methods having predicted RSD_1 values of 0.1 and 0.3% are available to establish the plutonium content of the WRM. The computed values of n are for $\mathrm{RSD}_1 = 0.1\%$

$$n = (4)(0.1)^2/(0.15)^2 = 1.78\,.$$

For $\mathrm{RSD}_1 = 0.3\%$

$$n = (4)(0.3)^2/(0.15)^2 = 16\,.$$

The advantage of a more precise method is apparent. One restriction to the number of analyses is the minimum of n = 5. For this example, the plutonium content of the WRM could be established by analyzing five replicate aliquots each of the WRM and the concurrently analyzed CRM by the method with a precision of 0.1% RSD_1 contrasted to 16 replicate aliquots each by the less precise method.

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*The LE of an estimator T is defined (Ref. 14) as twice the standard deviation of T. The RLE, expressed as a percentage, is 100 LE/T.

6. STATISTICAL TESTS AND ASSIGNMENTS OF VALUES TO WRMs

The statistical tests applied to establish the uranium or plutonium contents and isotopic distributions and their associated uncertainties are presented in Refs. 1-5. Reference 4 for a mixed-oxide WRM is most appropriate because it deals with both uranium and plutonium in a solid material. The tests presented cover the use of two analysis methods and the use of a makeup value and one analysis method.

7. CORRECTION OF PLUTONIUM AND URANIUM CONTENTS AND ISOTOPIC DISTRIBUTIONS FOR DECAY OF PLUTONIUM ISOTOPES

The decay of plutonium isotopes changes the plutonium and uranium contents and isotopic distributions. All these values should be computed at selected intervals using currently accepted half-life values. At present, these values are 87.74 ± 0.04 y for ^{238}Pu, 24 119 ± 26 y for ^{239}Pu, 6 540 y for ^{240}Pu, 14.4 ± 0.05 y for ^{241}Pu, and 387 000 y for ^{242}Pu. Table II shows the decreased Pu content; grown-in quantities of ^{234}U, ^{235}U, ^{236}U, ^{238}U, and ^{241}Am; and changed Pu isotopic distribution for a typical NBS Pu metal SRM 949 at a 10-y decay time or about the elapsed time since the issuance of this reference material. For demonstration, the initial plutonium content is taken as 1 g Pu/g material (or 100% purity). It is noted the plutonium content decreases 0.05% from 100.00% to 99.95% and that 0.02% (or 40 relative percent of the 0.5% decrease) is caused by ^{241}Pu decay. The total uranium growth then is 0.03% of the plutonium. Reference 15 details the exact calculations and includes a FORTRAN IV program.

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TABLE II

DECAY OF PLUTONIUM ISOTOPES IN A TYPICAL NBS SRM 949 Pu METAL

Pu Isotope	Initial Atom Fraction	Final Atom Fraction	Pu Isotope Decay (μg/g)	Daughter Formed	
				Isotope	μg/g
238	0.00003	0.0000277	2.268	^{234}U	2.229
239	0.97617	0.976387	277.353	^{235}U	272.703
240	0.02324	0.023227	24.719	^{236}U	24.306
241	0.00054	0.000337	204.693	^{241}Am	204.693
242	0.00002	0.00002	0.0004	^{238}U	0.0004
			Σ=509.033		Σ=503.931

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