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DOE STANDARD

Criteria for Preparing and Packaging Plutonium Metals and Oxides for Long-Term Storage

U.S. Department of Energy
Washington, DC 20585

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Foreword

This Department of Energy (DOE) Standard supersedes DOE-STD-3013-94 and is approved for use by all DOE components and their contractors.

Plutonium storage practices at DOE facilities evolved over decades during which the objectives of Department programs were to support nuclear weapons development and production. These practices reflected a desire to maintain plutonium in metal form for prompt recycling into weapon components. Prevailing procedures and safety requirements addressed only short-term storage considerations. The end of the Cold War and the new arms control agreements are leading to the retirement of large numbers of nuclear weapons resulting in inventories of plutonium that exceed defense needs and will necessitate longer term storage.

This Standard establishes safety criteria for packaging of plutonium metals and stabilized plutonium oxides for long-term storage at DOE facilities. Plutonium packaged according to these criteria should not need subsequent repackaging to ensure safe storage for at least 50 years.

Beneficial comments (recommendations, additions, deletions) and any pertinent data that may improve this document should be sent to the Office of the Associate Deputy Assistant Secretary for Technical and Environmental Support (DP-45), U.S. Department of Energy, Washington, DC 20585, by letter or by using the self-addressed Document Improvement Proposal (DOE F 1300.3) appearing at the end of this document.

DOE technical standards, such as this one, do not by themselves establish mandatory requirements. However, all or part of the provisions in a technical standard can become requirements under the following circumstances:

(a) they are explicitly stated to be requirements in a DOE requirements document; or

(b) the organization makes a commitment to meet a standard in a contract or in a plan or program required by a DOE requirements document.

Throughout this Standard, the word "shall" is used to denote actions which must be performed if this Standard is to be met. If the provisions in this technical Standard are made mandatory through one of the two ways discussed above, then the "shall" statements become requirements.

1. SCOPE

1.1 Purpose. This Standard provides criteria for packaging of plutonium metals and stabilized oxides for storage periods of at least 50 years. To meet the criteria, plutonium-bearing materials must be in stable forms and be packaged in containers designed to maintain their integrity both under normal storage conditions and during anticipated handling accidents.

1.2 Applicability. This Standard applies to packaging for safe storage of plutonium-bearing metals, alloys, and oxides that contain a minimum of 50 mass-percent plutonium. It does not apply to packaging of plutonium-bearing liquids, process residues, waste, sealed weapon components, irradiated fuels, or materials containing more than three mass-percent plutonium-238.

Storage facility design, safeguards and security interfaces, and transportation requirements are addressed in detail by other DOE directives (e.g., rules, orders) and other agencies' regulations. Such requirements are not repeated in this Standard. However, users of this Standard are advised to consult and assure adherence with other applicable directives while implementing these criteria.

2. REFERENCES

2.1 Federal Regulations. The following Federal regulations are referenced in this Standard.

10 CFR 830.120, Nuclear Safety Management, Quality Assurance Requirements.
10 CFR 835 [Appendix D], Occupational Radiation Protection, Surface Radioactivity Values.
49 CFR 178, Specifications for Packagings.

Copies of Federal regulations are available from the Government Printing Office (GPO), Superintendent of Documents, Mail Stop: SSOP, Washington, DC 20402-9329

2.2 Department of Energy Orders and Reports. The following DOE orders and reports are referenced in this Standard.

DOE O 420.1 Facility Safety, October 13, 1995.
DOE 5633.3B, Control and Accountability of Nuclear Materials, September 7, 1994.
DOE 5660.1B, Management of Nuclear Materials, May 26, 1994.
DOE/DP-0123T, Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities, January 1994.

Copies of DOE orders and reports are available from U.S. Department of Energy, Distribution and Staging, Washington, DC 20585, (202)586-9642, or on the Internet at the following world-wide web address:
http://www.explorer.doe.gov:1776/htmls/directives.html

2.3 Non-Federal References. The following non-government documents are referenced in this Standard.

ANSI N14.5-1987, "Standard for Radioactive Materials - Leakage Tests on Packages for Shipment," American National Standards Institute, Inc., New York, NY.

LA-12999-MS, Haschke, J. M., Ricketts, T. E., "Plutonium Dioxide Storage: Conditions for Preparation and Handling," August 1995. Los Alamos National Laboratory, Los Alamos, NM.

3. DEFINITIONS

Alloy - A condensed substance composed of two or more metallic elements formed by physicochemical process and intercomponent dissolution.

Alpha Plutonium - The stable metallurgical phase of pure plutonium at temperatures below 115 °C (239 °F).

ANSI - American National Standards Institute.

Barrier - A confinement that prevents the dispersion of stored material. A sealed barrier may also serve to isolate stored material from atmospheres that could react with the stored material.

CFR - Code of Federal Regulations.

DOE - United States Department of Energy.

Loss-on-Ignition (LOI) - Mass loss measured when a representative thermally stabilized sample of plutonium-bearing oxide is heated to confirm the elimination of residual moisture and other volatile species from the thermally stabilized material.

MC&A - Materials Control and Accountability.

NDA - Non-Destructive Assay. A procedure (e.g., calorimetric or radiometric measurement) for determining the amount of plutonium in a container without sampling the material.

Non-Volatile Material - A chemical compound that has a vapor pressure less than $1x10^{-5}$ Pa ($1x10^{-10}$ atm.) at 1,000 °C (1,832 °F).

Package - A packaging plus its contents.

Packaging - A receptacle and any other components or materials necessary for the receptacle to perform its containment function.

Quality Assurance (QA) - All planned and systematic actions necessary to provide adequate confidence that a structure, system, or component will perform satisfactorily in service.

Quality Control (QC) - Those quality assurance activities that provide a means to direct and measure the characteristics of a structure, system, or component to established requirements.

Residue - Process-generated solid plutonium-bearing materials not classified as storable metal or stabilized oxide.

Shall, Should and May - "Shall" denotes that something is required. "Should" denotes that something is recommended. "May" denotes that something is permitted.

Storage - The method for safely maintaining items in a retrievable form for subsequent use or disposition.

Storage Facility - The building structure and other confinement systems that house the storage containers.

Thermal Stabilization - A process that exposes plutonium-bearing material in air to an elevated temperature for the duration required to convert reactive constituents present to oxides and to remove adsorbed moisture and other volatile species.

4. PACKAGING/STORAGE CRITERIA

The following criteria are established to control hazards to workers, the public, and the environment associated with the packaging/storage of plutonium metals and oxides. Bases for the criteria are provided in Appendix A, and are discussed in DOE/DP-123T, "Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities," January 1994. In addition to these criteria, stored materials must comply with other applicable directives such as those contained in DOE regulations, orders, and policies. For example, stored materials must comply with materials control and accountability and radiation protection requirements.

4.1 Material Form. The packaged materials shall be solid metals or oxides containing a minimum of 50 mass-percent plutonium.

4.1.1 Material quantities / physical properties. The quantity of material per storage package shall have a thermal output of less than 30 Watts.

4.1.2 Metals. Storage packages of metals shall meet the following additional requirements:

a. The quantity of material per container should be as close as practical to, but shall not exceed, 4.40 kg (9.68 lb.).

b. The steady-state temperature of stored alpha plutonium metal shall not exceed 100 °C (212 °F).

c. Stored metal pieces shall have thicknesses greater than 1.0 mm (0.04 in.) and have specific surface areas less than 1.0 cm^2/g (71 $in^2/lb.$) to reduce potential pyrophoric tendencies. Metal pieces with greater specific surface areas shall be recast or converted to stabilized oxide prior to packaging. (See Appendix A)

d. Stored metals shall be free of loose oxide.

4.1.3 Oxides. Storage packages of oxides shall meet the following additional requirements:

a. The quantity of material per container should be as close as practical to, but shall not exceed, 5.00 kg (10.97 lb.).

b. Oxides shall be thermally stabilized by heating in air or an oxidizing atmosphere to 950 ˚C (1,742 °F) or higher for at least two hours.

c. Thermally stabilized oxides shall exhibit less than 0.5 mass percent loss-on-ignition (LOI) and shall retain this characteristic through final packaging. The LOI test shall be based on heating a representative sample of the stabilized oxide in air to 1,000 ˚C (1,832 °F) or higher for at least one hour.

4.2 Packaging.

4.2.1 General requirements. The package shall include a minimum of two nested sealed containers to isolate the stored materials from the environment and to prevent contamination release. The package (outer container including inner container and stored material) shall have at least one container that remains leak-tight as defined by ANSI N14.5 after a free drop from a 9-meter (30 ft.) height onto a flat, essentially unyielding, horizontal surface. The drop test shall follow the test procedures specified in 49 CFR 178.603, and shall be conducted using prototype containers loaded with non-radioactive surrogate material that simulates the planned plutonium loading for the package. Containers used for tests shall not be recycled or reused for storage.

The following requirements apply to both the inner and outer containers:

a. Shall be fabricated of materials that are resistant to corrosion in the anticipated storage environment.

b. Shall contain an atmosphere that will preclude a need for restabilizing the material contents. Also, the atmosphere within the containers shall not adversely react with the container nor preclude leak-testing.

c. Shall be tested for leak-tightness as defined in ANSI N14.5.

d. Shall allow for NDA, material verification, and inspection and surveillance.

e. Shall have permanent identification markings such as by etching, or engraving.

f. Design shall be hydrostatically proof-tested to 1.5 times the calculated theoretical maximum pressure (See Appendix A, Section 4.2.1 F) and shall remain leak-tight, as defined in ANSI N14.5, after the proof-test.

g. Shall not include any combustible or organic material.

4.2.2 Inner container. The inner container shall meet the following requirements:

a. Shall be sized to fit into an outer container with clearance for welding the outer container.

b. The inner container shall allow for a non-destructive indication of a buildup of internal pressure at less than 690 kPa (100 psig).

c. The exterior surface shall be free of contamination as defined by 10 CFR 835, Appendix D.

4.2.3 Outer container. The outer container shall have a cylindrical geometry and meet the following requirements:

a. The dimensional requirements for the outer container shall be as follows:

1. Maximum outside diameter 12.5 cm (4.9 in.).

2. Maximum external height of 25.4 cm (10 in.).

b. The interior and exterior surfaces shall be free of contamination as defined by 10 CFR 835, Appendix D.

4.2.4 Optional container(s). The use of additional containers, sometimes referred to as "material" or "convenience" containers is optional. If used, the containers shall not include any organic materials such as plastics, elastomeric gaskets, and organic coatings; and shall be made of materials that will not lead to degradation of the stored plutonium or other containers. The container shall be sized to fit in the inner container with adequate clearance for welding.

4.3 Inspection and Surveillance for Safety.

a. Inspection and surveillance procedures shall be site-specific and shall identify:

1. Prerequisites.

2. Acceptance criteria.

3. Specific instructions to ensure that items not meeting acceptance criteria are addressed in accordance with approved procedures and DOE reporting requirements.

4. Frequency for surveillance.

4.3.2 Documentation of inspection and surveillance methods. Formal methods and responsibilities shall be documented and maintained for independent review and evaluation.

4.3.3 Surveillance plan. The surveillance plan shall include provision for:

a. Initial baseline package inspections within 30 days of closure.

b. Periodic surveillances throughout the storage period. Surveillance frequency, sample population, and package selection shall be established by a statistical approach.

c. Any indication of container deformation shall be evaluated.

4.3.4 Safety inspections. Safety inspections should be integrated with other required inspections (e.g., Materials Control and Accountability) and may be intrusive or non-intrusive as appropriate.

4.3.5 Schedule for surveillance. A schedule shall be maintained for continuing safety and accountability surveillance testing.

4.4 Documentation.

4.4.1 Data base. A data base shall be maintained to serve as a source of relevant information about stored materials and packages. For completeness, MC&A documentation should be coordinated with the data base.

4.4.2 Data base requirements. The data base shall include, as a minimum:

a. Identification of the following material characteristics:
   1. Chemical form (e.g., plutonium metal, plutonium oxide).
   2. Element and isotopic distribution (e.g., % ^{238}Pu, % ^{239}Pu, % ^{240}Pu, % ^{241}Pu, % ^{242}Pu).
   3. Quantity (mass).
   4. LOI test results and conditions of tests.
   5. Source of stored material (facility that generated the material).
   6. When plutonium was last separated - americium removed (if information is available).
   7. Specific processing condition(s).
   8. Other information relative to the contents.

b. Identification of the following package characteristics:
   1. Fill gas composition on sealing.
   2. Package configuration - number of containers in package.
   3. Date of packaging.
   4. Initial radiation field (gamma and neutron at contact and 30 cm. (12 in.)).
   5. Surveillance results.
   6. Baseline package weight and dimensions.

c. Records of the inspection tests performed, names of individuals performing inspections, and the dates of inspections,

d. Locations of stored materials.

4.5 Quality Assurance/Control Requirements.

4.5.1 All personnel participating in essential processes and procedures shall be trained and qualified for their areas of involvement.

4.5.2 Materials used in the fabrication and sealing of containers shall satisfy all specifications necessary to comply with the requirements of these criteria.

4.5.3 Procedures and processes that are essential for assuring compliance with these criteria shall be subject to Quality Assurance (QA) per 10 CFR 830.120 and Quality Control (QC) Procedures.

4.5.4 Essential procedures and processes covered by QA and QC requirements shall include (but will not be limited to):

a. Thermal stabilization procedure.

b. LOI procedure used for oxide stabilization certification.

c. Sealing (e.g., welding) procedure used in container fabrication and closure.

d. Leak testing procedure used for certifying that containers are leak-tight.

e. Surveillance procedure(s).

f. Data base recording procedure and characterization parameters addressed in Section 4.4.B.

APPENDIX A

BASES AND ADDITIONAL INFORMATION
FOR PLUTONIUM PACKAGING/STORAGE CRITERIA

This Appendix briefly provides the bases for the paragraphs in the body of this Standard. The section numbers in this Appendix correspond to the sections in the body of the Standard.

1. SCOPE

This Standard establishes criteria for packaging of plutonium metals and plutonium oxides containing more than 50 mass-percent plutonium for storage at DOE facilities. Materials packaged to meet these criteria should not need subsequent repackaging for at least 50 years.

2. REFERENCES

No Basis Provided

3. DEFINITIONS

Adopted from relevant titles of the Code of Federal Regulations (CFR) and The Handbook of Acronyms, Abbreviations, Initialisms, Proper Names and Alphanumerics Encountered in Nuclear Safety Literature, March 1993.

4. PACKAGING/STORAGE CRITERIA

4.1 Material Form. This Standard applies to the packaging of solid plutonium metals, selected alloys, and stabilized oxides that contain a minimum of 50 mass-percent plutonium. It does not apply to plutonium-bearing liquids, residues, waste, sealed weapon components, irradiated fuels, or material containing more than three mass-percent plutonium-238.

Typical plutonium isotopic compositions (by mass-percent) for various grades of plutonium are as follows:

Isotope (mass %)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Weapons Grade	<0.05	93.6	6.0	0.4	<0.05
Fuel Grade	0.1	86.1	12	1.6	0.2
Power Grade	1.0	62	22	12	3.0

To support nondestructive assay needs, material should be packaged in a consistent manner with respect to: plutonium quantity, uniformity of the matrix, application of seals and identification markings.

4.1.1 Material quantities / physical properties. The permitted quantity of material per storage package depends on its heat output, radiation level, and criticality constraints. The thermal output is limited to a maximum of 30 Watts per package to assure compliance with limits at existing and planned storage facilities as well as for possible future shipment off-site. To promote material homogeneity for facilitating MC&A measurements, only similar materials should be combined and packaged for storage. For example, when repackaging for safe long-term storage, plutonium materials should not be mixed or diluted with uranium materials. Also, consideration should be given, consistent with the ALARA concept, to the impurities in plutonium oxide, for impurities that facilitate alpha-neutron reactions could be an additional source of radiation exposure.

4.1.2 Metals. The mass limit for plutonium metal is based on criticality safety limits and future off-site transportation considerations. The maximum fissile unit for an isolated sphere of pure plutonium-239 that is water-reflected and has a 0.05 margin in K_{eff} is calculated at 4.53 kg (9.97 lb.). The existing shipping packages that could potentially be used for off-site transportation have a 4.40 kg (9.68 lb.) limit for plutonium-239. The quantity of plutonium stored in a package should be as close as practical to the maximum allowable quantity to minimize the number of packages generated, but shall not exceed the 4.40 kg (9.68 lb.) limit.

The respective limits refer to the total masses of these materials, not to the amounts of plutonium metal present. This constraint will prevent a potential criticality incident including cases of stored impure fissile materials (i.e., U-233, U-235, Np-237, Am-241). Since the critical masses of these radioisotopes are greater than that of plutonium compromising criticality safety is precluded by the mass limits on the materials when they are assumed to be entirely plutonium-bearing materials.

Plutonium metal exhibits six distinct allotropic forms between room temperature and its melting point. The transformation temperature of plutonium metal from the alpha-phase to the beta-phase is approximately 117 °C (225 °F). The metal expands significantly (and not necessarily isotropically) during heating and phase transformation. A container must provide sufficient heat dissipation to maintain alpha-phase plutonium metal below 100 °C (212 °F). Relevant data for the six allotropic forms of plutonium are presented in the following table.

ALLOTROPE	RANGE OF STABILITY (°C)*	X-RAY DENSITY (g/cm^3)*	Linear Expansion Coefficient per °C $x10^6$ *
Alpha	<115	19.86	54.0
Beta	115 - 200	17.70	42.0
Gamma	200 - 310	17.14	34.6
Delta	310 - 452	15.92	-8.6
Delta-prime	452 - 480	16.00	-65.6
Epsilon	480 - 640	16.51	36.5

*Plutonium Handbook, A Guide to the Technology, ANS, 1980.

Potentially pyrophoric plutonium metal pieces are not acceptable storage forms. Metal fines, foils, and turnings are potentially pyrophoric and should be either converted to a stable metal form (bulk metal) or stabilized oxide prior to storage. The specified thickness and surface area limits are based on the Assessment Report, Section II.F, "Pyrophoricity," and Section V.B.1, "Chemical Reactivity." (See DOE/DP-0123T). Some loose oxides may also be pyrophoric. Prior to repackaging plutonium metal, loose oxide should be removed from the metal. This may be accomplished using a soft bristle brush. An adherent oxide layer on stored metal is beneficial because it tends to retard further oxidation. The Assessment Report, Section II.B, describes the reactions of plutonium metal with moisture, resulting in its degradation and creation of potentially pyrophoric hydrides (See Section II.C of the Assessment Report, DOE/DP-0123T).

4.1.3 Oxides. The quantity of oxide, 5.00 kg (10.97 lb.), is the plutonium dioxide equivalent of 4.40 kg (9.68 lb.) of plutonium metal. Actual quantity may be further limited due to facility-specific considerations including criticality, radiation, and heat output constraints. The respective limits refer to the total masses of these materials, not to the amounts of plutonium oxide present.

This constraint will prevent a potential criticality incident including cases of stored impure fissile materials (i.e., U-233, U-235, Np-237, Am-241). Since the critical masses of these radioisotopes are greater than that of plutonium compromising criticality safety is precluded by the mass limits on the materials when they are assumed to be entirely plutonium-bearing materials.

Oxides with residuals that could over-pressurize the inner storage container over a 50-year period are not acceptable for storage. Plutonium oxide powder can have a high surface area per unit weight depending on preparation conditions. Such powder could adsorb up to 8% of its weight as moisture. The storage hazard associated with adsorbed moisture is the potential over-pressurization of a sealed oxide container over a prolonged period through radiolytic and chemical processes. The quantity of oxide, 5.00 kg (10.97 lb.), is the plutonium dioxide equivalent of 4.40 kg (9.68 lb.) of plutonium metal. Actual quantity may be further limited due to facility-specific considerations including criticality, radiation, and heat output constraints. The respective limits refer to the total masses of these materials, not to the amounts of plutonium oxide present. This constraint will prevent a potential criticality incident including cases of stored impure fissile materials (i.e., U-233, U-235, Np-237, Am-241). Since the critical masses of these radioisotopes are greater than that of plutonium compromising criticality safety is precluded by the mass limits on the materials when they are assumed to be entirely plutonium-bearing materials.

To assure stability of oxide, it is subject to a calcination process wherein the material is exposed to an oxidizing environment at an elevated temperature. Moisture and other adsorbates are effectively removed by heating the oxide in air at 950 °C (1,742 °F) or higher for at least two hours[1]/ . Additional benefits of the calcination process are that it makes the powder less dispersible, by reducing its specific surface area, and inhibits readsorption of moisture.

The loss-on-ignition (LOI) test is the standard procedure for confirming the thermal stabilization of plutonium oxide. It is determined by measuring the weight loss resulting from heating a representative weighed sample of

[1]Haschke, John M., "Plutonium Storage: Conditions for Preparation and Handling," LA-12999-MS, August 1995.

calcined oxide in air to above the thermal stabilization temperature to drive off any remaining moisture and volatile compounds. This Standard requires calcined oxide to exhibit a less than 0.5 mass percent LOI after heating in air at 1,000 °C (1,832 °F) or higher for at least one hour. This measurement requirement ensures that a batch of oxide has been sufficiently stabilized so that the storage package will not over-pressurize from gases generated during long-term storage.

Oxides should be sufficiently blended to allow for nondestructive assay. Alpha-neutron compounds (e.g., plutonium fluoride, or other low atomic number elements mixed with plutonium) in stabilized oxides should be limited to levels that would not impact non-destructive assay (NDA) analysis. Known information regarding the chemical form and quantity of an alpha-neutron compound included in the stored material should be recorded (see Section 4.4B).

4.2 Packaging. The design goals for the storage package are that it should be maintenance free, and that it could be shipped in qualified shipping containers without further reprocessing or repackaging.

4.2.1 General requirements. The purpose of the drop test requirement is to ensure that a storage package accidentally dropped from the maximum storage height would not release any material.

a. The requirement for corrosion resistant containers is based on operational experience and current technical evaluations for storing stable plutonium-bearing materials. Ductile, non-corrosive material should be used for the containers. A suggested container material is 304 L stainless steel which is ductile and has good resistance properties to stress corrosion.

b. The stored material condition should not change due to the container atmosphere. If material stabilization has to be repeated, there would be additional handling and unnecessary worker exposure.

c. ANSI N14.5, Leakage Tests on Packages for Shipment, specifies that the acceptable maximum leak rate is 1 x 10^{-7} std. cc/sec at one atmosphere. Full penetration weld closures and ports provide the highest integrity and longest life seals possible. They eliminate gaskets, which may degrade and leak, and mechanical devices such as bolts or screwed connections that are prone to wear, creep relaxation, seizure, or other mechanical failure.

d. Storage of plutonium-bearing material must comply with existing materials control and accountability (MC&A), safeguards and security, and audit and surveillance directives which rely on nondestructive assays as a technique for validations. The MC&A requirements call for routinely assaying stored materials for process, accountability, and inventory controls. Plutonium packaging and storage should not preclude adherence with these directives.

e. Identification markings are required by MC&A directives for maintaining an inventory data base and to facilitate management of stored accountable materials.

f. A maximum theoretical pressure in the inner container is calculated based on the sum of the initial pressure sealed in the container during packaging, the pressure that could be generated by chemical reactions involving residual adsorbates, and the pressure of helium generated by alpha decay of the contained plutonium during storage. This pressure (in psia) may be estimated using the following equation:

$$P = (T/T_0)P_i + [0.67(LOI)(m)(T)] / [V - (0.0873\ m)] + [1.3x10^{-4}\ (m)(t)(T)] / [V - (0.0873\ m)]$$

- T (in K) is the maximum average gas temperature anticipated during storage and is estimated to be 477 K (400 °F), unless the isotopic content and packaging configuration cause the value to be higher.

- T_0 (in K) is the maximum average temperature of gas in the container when the container is sealed.

- P_i (in psia) is the atmospheric pressure when the container is sealed.

- LOI (in %) is the maximum allowable loss on ignition value.

- m (in kg) is the mass of packaged oxide.

- V (in liter) is the free volume in the container after correction for volume displacement by an enclosed container or objects other than the oxide. The correction for volume displacement by enclosed containers (in liter) is given by [(mass of container in kg)/(density of the container material in kg/liter)]. The term 0.0873m in the equation corrects for the volume displacement attributed to stored oxide and is given by [(mass of oxide, kg)/(11.45 kg/liter)].

- t (in years) is the elapsed storage time after the package is sealed.

The second term is large compared to the first and third terms and is the primary determinant of the pressure increase. The basis for this term is the following reaction of oxide with residual adsorbed water to form hydrogen:

$$PuO_2\,(s) + xH_2O\,(adsorbed) = PuO_{2+x}\,(s) + xH_2\,(g).$$

Experimental measurements show that this reaction occurs at a measurable rate at 25 °C (77 °F) when the concentration of adsorbed water present on the oxide surface is high. The reaction rate increases as the oxide temperature increases and is expected to be much lower for LOI values less than 0.5%. Since current knowledge about the kinetics of reaction is inadequate to predict the extent of reaction for a given LOI at any point in time, the second term in the pressure equation is calculated for complete reaction of the total amount of water defined by the LOI. Therefore, the resulting pressure is the maximum theoretical value. (Note that the coefficient of the second term is only two-thirds of that for the pressure equation given in STD-3013-94. That relationship was alpha-induced radiolysis of water to produce one-half mole of oxygen in addition to the mole of hydrogen. Since experiments show that the above reaction occurs instead of the radiolytic process, the second term of the pressure equation has been reduced.)

The last term, which defines the build-up of helium pressure over time, is formulated for plutonium-239 and is applicable to oxides containing the nominal isotopic distribution of weapons-grade plutonium. If the isotopic composition of the oxide differs significantly from that of weapons-grade plutonium material, this term must be evaluated accordingly. An estimate for any known isotopic composition may be made using data in the following table. The fraction of each plutonium isotope that has decayed to a daughter nucleus plus an alpha particle (Helium atom) is presented for selected time periods up to 100 years. (For example, the moles of helium generated by 10 grams of plutonium-238 after ten years in storage is: [(10 g of ^{238}Pu)(0.076)/238 g per mole)].

Elapsed Time (Years)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu*	^{242}Pu
0	0.000	0.000	0.000	0.000	0.000
10	0.076	0.000	0.001	0.006	0.000
20	0.146	0.000	0.002	0.020	0.000
30	0.211	0.000	0.003	0.036	0.000
40	0.217	0.001	0.004	0.035	0.000
50	0.326	0.001	0.005	0.070	0.000
100	0.546	0.003	0.011	0.147	0.000

* Each value is the fraction of the initial ^{241}Pu isotope that has beta-decayed to Am-241 and then alpha decayed to ^{237}Np.

g. Storage vault compartments are low fire hazard areas and may not contain fire suppression systems. Any combustible materials in vault compartments, such as packing materials, could jeopardize the accepted low fire hazard classification of storage vaults. DOE O 420.1 requires fire prevention procedures governing the use and storage of combustible and flammable materials to minimize the risk from fire.

The Assessment Report, Section II.D (See DOE/DP-123T), describes the radiolytic effects with plastics, hydrogenous compounds, and organic materials in the storage of plutonium. Prolonged plutonium storage necessitates exclusion of such materials from sealed containers. Radiolysis of organic material will produce combustible or corrosive gases and increase pressure within a sealed storage container. Radiation and heat would also change the composition of organic materials so that they would no longer be capable of performing the designed packaging function.

4.2.2 Inner container. The inner container serves as the primary barrier isolating the stored material from the environment. Maximum dimensional limits are based on the outer container design. Pressure indication, such as a pressure deflectable lid or bellows observable by radiography, will permit early detection of pressurizing packages prior to potential inner container failure, and ensure that the two-barrier requirement is being met. When the material and storage package criteria are met, an unusual pressure increase would indicate a defect in either material preparation (thermal stabilization), packaging, or both.

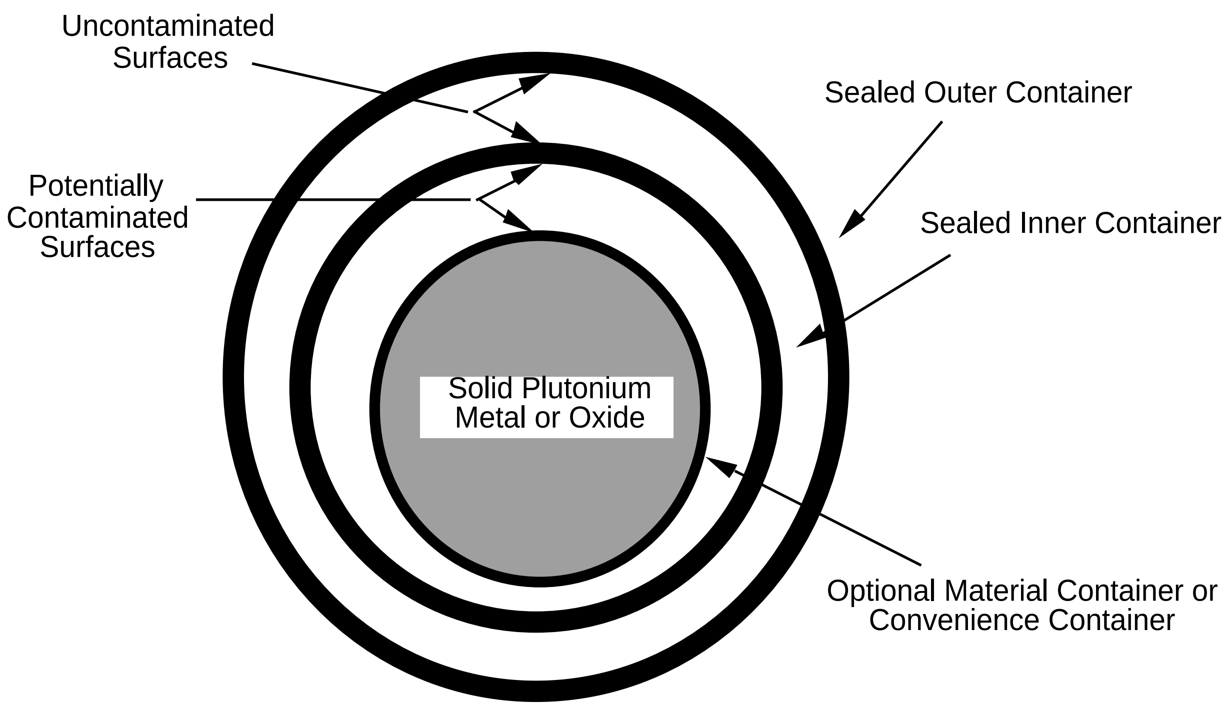

Figure 1. Top view of package for long-term storage of plutonium metals and stabilized oxides.

4.2.3 Outer container. The outer container is sized to fit into existing certified shipping containers. Its design will minimize future handling and avoid unnecessary additional personnel exposure, operational risk, and waste generation.

4.2.4 Optional container (s). The material container or convenience container refers to a container that is sometimes used to transfer plutonium-bearing material. A material container is not required in packaging and is not considered as an isolation barrier.

4.3 Inspection and Surveillance for Safety Inspection and surveillance can be either intrusive or non-intrusive. Non-intrusive testing methods include the following:

a. Radiography to observe physical changes in the stored material (oxide growth on plutonium metal), and dimensional changes of the inner container (pressure change).

b. Acoustic resonance spectroscopy to detect pressure changes.

c. Weight measurement change, which would indicate a breach in the package.

Intrusive test methods involve destructive testing of sample containers to evaluate the long-term performance of the package design and its contents.

During the course of packaging and storage of plutonium metal and oxide, it is anticipated that some packages might become flawed or that packaged material might not perform as predicted. The function of the inspection and surveillance program is to identify errors and flaws in the initial packaging as well as to detect package failures and contents performance that might affect package integrity during storage.

Initial Inspection. Flaws in initial packaging are expected to be detected during inspection of every container within 30 days of packaging. This initial inspection should provide baseline information on the dimensions of the inner sealed container, the leak rate of both sealed containers, verification of contents through NDA measurements, and any other information deemed desirable and attainable through non-intrusive measurements such as radiography. This initial inspection may be part of the quality program for verifying package integrity.

Surveillance. Essentially all manufactured products exhibit failure hazard functions that graph as a "bathtub" shaped curve. This type of curve combines three general types of failures, corresponding to the three specific parts of the "bathtub" curve, that occur over the lifetime of the product. The relative magnitude of the three parts of the curve will vary depending on the type of product. For lack of long-term storage data on plutonium metal and oxide storage packages, it is reasonable to expect that failures would follow a pattern typified by the "bathtub" curve. Flaws in initial packaging are expected to be detected during inspection of every container within 30 days of packaging. This initial inspection provides baseline information on the dimensions of the inner sealed container, the leak rate of both sealed containers, verification of contents through NDA measurements, and any other information deemed desirable and attainable through non-intrusive measurements such as radiography. This initial inspection may be part of the quality program for verifying package integrity.

The first failure type occurs soon after packaging or very shortly after the package service life begins and is referred to as "infant mortality." Failures are caused by mistakes, missteps, or other problems in the packaging process that result in flawed or defective packages. These types of failures will occur less often as the non-viable products fail quickly and are removed from the in-storage population and corrected. During this phase, the failure rate begins high (on a relative bases) and decreases over time. Most flaws serious enough to cause loss of integrity of the package would be detected with the initial inspection. Additional "infant mortality" failures are more likely to occur with plutonium oxide packages and would be caused by improper stabilization or characterization resulting in pressurization of the package. To detect these types of failures, each package should be inspected during what must be determined to be the "infant mortality" time period. A case might be made to more closely monitor containers storing oxide during this time period.

The second type of failure occurs after packages with manufacturing of processing defects have been detected and corrected and the population reaches a period in which random failure occurs. Uniform changes in the population, such as a gradual pressure generation in oxide containers, are also expected to occur during this period.

Surveillance during this period could consist of statistical sampling to monitor the behavior of the population. The duration of this period is unknown and must be defined using surveillance data.

The third and final type of failure is attributed to the "wearing out" of the package. As this stage approaches, failure rates begin to increase and the surveillance frequency may need to be increased.

Surveillance programs should be developed to monitor for indications of all three types of failure during the storage period.

4.4 Documentation. Documentation requirements are based on sound records management practices within the nuclear industry and DOE orders in the 5600 series (e.g., DOE 5633.3B and 5660.1B).

Materials control and accountability directives require that information concerning package contents be available to identify any materials that could impact neutron or gamma-ray measurements (e.g., neutron-producing or radiation-shielding matrix materials, non-homogeneous contents).

4.5 Quality Assurance/Control Requirements. Sound quality assurance and quality control practices are defined by 10 CFR 830.120, Nuclear Safety Management, Quality Assurance Requirements.

CONCLUDING MATERIAL

Review Activity

DOE	**Field Office**
DP	Albuquerque Operations Office
EH	Chicago Operations Office
EM	Idaho Operations Office
MD	Nevada Operations Office
NN	Oakland Operations Office
NS	Oak Ridge Operations Office
RW	Rocky Flats Office
	Richland Operations Office
	Savannah River Operations Office

National Laboratories
Idaho National Engineering Laboratory
Los Alamos National Laboratory
Lawrence Livermore National Laboratory
Sandia National Laboratory

Power Authorities
WAPA

Preparing Activity:

DOE-DP-45

Project Number:
PACK-0009