This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy.

Stabilizing And Packaging Pu Materials Per 3013 At SRS

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ABSTRACT

The Savannah River Site (SRS) began packaging Pu metals into 3013 containers in April, 2003 and oxides in October, 2003. A total of 919 outer 3013 containers were made in the FB-Line at SRS on a very aggressive schedule (average of nearly 42 containers per month). Stabilization and packaging was completed in January, 2005. Experiences, lessons learned, and an overview of packaging activities are presented.

I. INTRODUCTION

The Department of Energy Standard DOE-STD-3013-2004 [1] provides stabilization and packaging criteria for long term (50 years) storage of plutonium bearing materials. The 3013 Standard originated in 1994 as a result of experiences with several plutonium package failures and concerns expressed by the Defense Nuclear Facilities Safety Board with long term plutonium storage across the DOE complex. The Standard provides stabilization guidance/criteria for both plutonium bearing metals and oxides. Additionally, the Standard provides guidance/criteria for the packaging containers.

A. Packaging Requirements

The packaging consists of two welded nested 300 series stainless steel containers. An optional convenience container may be used to confine the plutonium bearing materials. No plastic or other organic materials may be present in the packaging or the materials (e.g. plastic bags, gaskets, seals, oils/grease, etc.). Both containers are required to withstand a 4 ft. drop and be leak tested upon closure. The outer container has a design pressure of 699 psig, is designed in accordance with ASME Section VIII, and is sealed with a full penetration weld. Both containers are backfilled with helium. Currently, the DOE has decided to use only one outer 3013 design for the entire complex, however several inner 3013 container designs have been approved.

B. General Requirements

General 3013 Standard requirements for each 3013 container are heat generation of packaged materials shall be no more than 19 watts at any time during storage, no more than 4.4 kg of plutonium metal or 5.0 kg of plutonium oxide may be packaged, the outer surface of the outer container shall be less than 20 dpm/100 sq. cm transferable alpha contamination and the outer surface of the inner container shall be less than 2,000 dpm/100 sq. cm transferable alpha contamination. Lastly, the outer 3013 container must be weighed and fit within a cylinder 127 mm diameter by 255 mm tall.

C. Stabilization Of Metals

The stabilization requirements for plutonium metals includes removal of all non-adherent oxide, packaging no pieces with greater than 100 sq. mm/g specific surface area (the Standard recommends packaging no metal pieces less than 50 grams), and foils, turnings, or pressed briquettes are prohibited.

D. Stabilization Of Oxides

The stabilization requirements for plutonium bearing oxides include heating for no less than 2 hours to 950^{o}C in an oxidizing environment. A representative sample of the stabilized material is tested to ensure that the moisture content is less than 0.5% wt. In addition oxides must be considered to be represented in the Materials Identification and Surveillance (MIS) shelf life program at Los Alamos National Laboratory.

E. Documentation

A very formal methodology for maintaining documentation of stabilized and packaged plutonium materials has been developed in order to effectively manage long term storage and perform long term surveillance. The documentation is entirely electronic with a standardized database. Each site packaging materials per 3013 must populate the database for their sites materials. The database consists of “required” and “if available” fields. A single database of the entire DOE complex 3013’s is maintained at Los Alamos National Laboratory.

II. IMPLEMENTATION AT SRS

Each site within the DOE which is packaging per the 3013 Standard is required to submit implementation documentation. Specifics regarding preparation of metal materials, inner 3013 container and convenience can designs, furnace operation and control, moisture measurement and control for oxides, a weld quality plan for outer containers, representation of oxides in the MIS studies, and database implementation specifics for the stabilization practices of the site. The documentation may be revised during packaging and stabilization activities as technical issues arise and approaches change.

A. Packaging

As already mentioned, a single design of the outer 3013 container is used throughout the DOE complex. The inner 3013 container used at SRS is called a bagless can (historically, materials are bagged out of a glovebox in a convenience container in order to control contamination). The bagless system was developed at SRS in the early to mid 1990’s and became operational in FB-Line in 1997. The full 3013 program at SRS had not been developed at this time. No convenience can was used for packaging metals. Due to criticality limitations, the bagless containers were limited to 2150 grams (net wt.) of metal, however they were sized such that two could be stacked inside an outer 3013 container. These bagless cans are referred to as "short" cans. Contamination in the weld of the bagless can when packaging multiple metal pieces resulted in the design and implementation of an open top convenience bucket.

A 1.6 liter screw top 300 series stainless steel oxide convenience can was designed for use at SRS. The container is designed with several small slots at the can top in order to facilitate helium backfill. This convenience can is packaged within a single tall version of the bagless can (referred to as a "tall" can) for oxides.

Long metal pieces are packaged in the tall can configuration including the screw top convenience can. Specific FB-Line criticality limitations restrict the net wt. of metal to 2150 grams per bagless container.

B. General Requirements

The 19 watt heat generation limit is easily satisfied due to the relatively low specific heat generation of the SRS plutonium bearing materials. A bounding calculation was performed to ensure that each 3013 generated less then 19 watts (not only at the time of packaging, but also throughout its life). Each bagless container was decontaminated to less than 200 dpm per 100 sq. cm alpha prior to packaging into an outer 3013, the outer 3013 was weighed following successful completion of all screenings (verification of weld DAS parameters, visual exam of weld, leak check, and digital radiography check for porosity). The outer 3013 is welded in a clean area which necessitates it being less than 20 dpm per 100 sq. cm alpha.

For tall bagless cans, the mass of material packaged is verified to be compliant when the bagless can is welded. When two short bagless cans are packaged into an outer 3013, the net material mass of each bagless can is summed to verify compliance.

C. Stabilization of Metals

Most of the metals at SRS consists of “buttons” or pucks of plutonium. These buttons were produced in FB-Line for over 40 years. A final campaign, which ended in April 2002, completed stabilization of plutonium solutions in F-Canyon at SRS. SRS also has a number of ingots and scrap metal pieces (a number of which are mixed Pu/U metals and alloys from reactor research programs) from throughout the DOE complex. Pieces that are too long to fit into the “short” version of the bagless can are packaged into the tall one using the screw top convenience can.

All metal pieces are verified to be at least 50 g in mass and then brushed to remove non-adherent oxide. The brushed pieces are packaged within the same shift (otherwise the pieces are brushed again) in order to minimize oxide growth. In addition, all metal pieces are packaged in a dry air environment with dew points typically no greater than -10^{o}C. It should be noted that metal packaging began in 1997, well before the entire 3013 program was in place.

D. Stabilization of Oxides

Plutonium bearing oxides are batched together per material type (e.g. pure oxides from oxalate, oxides from burn metal, mixed oxides from the same source, fuels grade oxides, etc.), sieved, and weighed prior to stabilization.

Two Inconel retort type of furnaces with electric heaters are used for stabilization. A thermowell houses a Type S thermocouple which measures the oxide temperature (in the middle of the oxide, which is the coolest location). Additional thermocouples are used to measure the retort and heater temperatures. The sweep gas is air and its flow rate is set via a rotometer. Additionally, the pressure across the retort and in the off gas line is measured. Several interlocks and alarms are used to protect the furnace from over heating, over pressurization, under heating a stabilization run, and loss of sweep gas flow. Several heating curves were developed to facilitate complete stabilization for the different material types.

Stabilized oxide is dumped into the screw top convenience can and stored until the bagless can is welded (up to 6 convenience cans may be stored at a time in the glovebox line). Just prior to welding, a sample is taken and packaged into a screw top metal vial (with replaceable rubber o-ring gasket) for TGA moisture measurement.

E. Documentation

To support long term storage and surveillance of the containers, a standardized database was created. The database was to contain a record for each container that was packaged to the 3013 standard. This would allow surveillance to be performed on the entire population instead of each storage site to have their own program. An extensive program was set up at SRS to control data collection and the population of the electronic database.

As each container was generated, procedures, datasheets, and electronic files were collected and placed in a data package. A Use Every Time (UET) procedure is used during each stage of the stabilization and packaging process. The procedures include Appendices where relevant data are recorded. A QA review was performed on each procedure and data sheet before there was any input in the database. Any errors or omissions were corrected or when necessary, a Non-Conformance Report was issued.

When the data package was released by QA, all of the supporting procedures were scanned and data fields were populated. Engineering would perform a review of the data package to ensure that all of the required information had been collected and that would certify that the container met all of the 3013 requirements. When the item was packaged for shipment, additional information was generated that had to be collected and input into the database.

Once all of the information was entered, QA would perform an extensive review of each data field, electronic file, and scanned image. The review ensured that all of the required information had been inputted, that it matched the source documentation, and that the information was within required ranges. The data record was then copied to the complex-wide database.

III. OBSERVATIONS AND LESSONS LEARNED

A. Packaging

During packaging of multiple metal pieces, removable contamination (which occasionally exceeded 1,000 dpm/100 sq cm alpha) was detected on the bagless can. To facilitate contamination control for packaging multiple metal pieces, an open top convenience “bucket” was designed and used. Pieces were first placed into the bucket and then the bucket was lowered into the can. This single pass into the bagless can helped reduce the spread of contamination. Packaging of oxides resulted in similar contamination problems. Ultimately, an external slip lid can was used as a temporary contamination barrier (during storage and assay activities) until the bagless can was packaged into the outer 3013. The only other issue impacting packaging was discovered and resolved during startup testing [2].

B. General Requirements

These requirements were easily satisfied. Deliberate and consistent radiological conduct of operations ensured that minimal contamination levels were maintained.

C. Stabilization of Metals

Repackaging of metal buttons and ingots into bagless containers began several years before the 3013 program was formally established. Implementation requirements and “industry standard practices” with respect to 3013 Standard compliance had not been established. Nevertheless, basic requirements were implemented such as brushing to remove non-adherent oxide. In addition, dry air was required to be operational during metal packaging activities, and the time in which a metal item may be packaged after being brushed was limited to a single shift (if the item was not packaged the next shift would brush the item again to ensure non-adherent oxide did not grow onto the metal surface).

During the course of packaging activities (after start up of welding outer 3013 containers (April 2003) and after shipment of 3013 containers for long term storage at SRS, which began in October 2003), metal packaging procedures used prior to March 2002 were found to not implement the 1999 version of the Standard with respect to metal fragments. These procedures were not fully recovered until the bagless cans had been packaged into outer 3013's, and the procedures had not been reviewed prior to packaging and shipment for long term storage in a 9975 container.

The 1999 version required metal pieces to be at least 50 g, while the 1996 version (which was implemented) required metal pieces to be at least 1 mm thick and have a specific surface area less than 1.0 sq. cm/g. Per the completed packaging procedures, two bagless cans were found to contain less than 50 g metal fragments (three fragments in one can, packaged in June 2000; and one fragment in the other, packaged in September 1997) packaged with the larger metal button. This discovery resulted in the return of the 9975 containers from the long term storage facility to the packaging facility. The two items in question were repackaged and the fragments were removed. The primary lesson learned from this incident is that in order to ensure strict programmatic compliance, all historic records should be carefully reviewed. This is especially true when work has already been performed and credited prior to fully establishing the program.

The 50 g mass requirement for metal pieces proved to cause processing challenges. Much of the legacy metal scrap consisted of (primarily mixed Pu/U) materials from DOE nuclear reactor research programs. However, both metal and oxide materials were found within the original can. A significant portion of the metal pieces were less than 50 g. This resulted in development of alternative processing and disposition paths (metal oxidation and dissolution) depending on the composition of the less than 50 g metal material. The lesson learned is that the material form is uncertain when dealing legacy passivated metal material. Radiography was used to examine these materials in their original packagings. However, without comprehensive 3-D images (which were not available) it often provides inconclusive and even misleading indications of material size and mass. Along these lines, "legacy" storage containers pose hazards which require careful review and consideration.

As such, in accordance with Integrated Safety Management (ISM), an expert "legacy review" team was created to review records and radiographs to evaluate the "legacy" materials and their packagings. The team identified potential hazards and provided handling guidance. The relevant safety information and guidance was provided to the operators for review during pre-job briefs. This approach resulted in an appropriately graded handling of "legacy" materials in a very safe manner during the campaign.

D. Stabilization of Oxides

The plutonium bearing oxides were organized into material families and sub groups within the families prior to processing. Large groups of relatively pure materials were processed first. Stabilization of pure materials was highly successful, other than occasional equipment problems (e.g. furnace thermocouple failure). Furnace inspections indicated minimal "wear" within the furnaces. These materials easily passed the TGA moisture measurement.

Occasional discrepancies with records regarding the net weight of the source material were discovered - however these were not completely unexpected, and in general did not cause processing delays. However, in one case a significant mass discrepancy was discovered upon opening and weighing the material (prior to processing). An extensive records search found that the item had actually gone to the dissolver cabinet. A likely scenario is the can was opened and the dissolving activities were suspended. The material was not dissolved and additional left over material (from same material family) was added to it. The repackaged container was never assayed nor records updated to reflect the change.

Special contingencies were made for processing of a large group of material which were known to be chloride/fluoride bearing. These included ensuring a replacement thermowell was available (corrosion of the existing one was anticipated), increased furnace inspections, and a redesign of the furnace off gas system to facilitate cleaning out blockages. The off gas system did plug twice over the 60 furnace runs associated with this group of material. The furnace, thermowell and even the stabilization pans did not excessively corrode during this campaign. Salt waste was generated and had to be removed from the furnace periodically (brushed and collected). This was assayed to confirm minimal plutonium accumulation and discarded as TRU waste.

Processing of less pure material proved to be more problematic. In some cases the material showed significant non-hydrogenous weight loss (as high as 9% during stabilization and 1.7% during TGA measurement). Even though the 3013 Standard has no quantified limit for non-hydrogenous weight loss (moisture limit is 0.5% wt.), such a result was considered unacceptable. Six such items were restabilized in the furnace and subsequent TGA measurements indicated a much lower total weight loss (less than 0.6%).

Temperature excursions occurred during some of the runs.

An increase in 400-500^{o}C was detected by the cake thermocouple for a very short duration. In a couple of cases the presence of beryllium in the material resulted in an increase in neutron dose rates (the bagless can measured up to 200 mrem/hr at 30 cm). Existing facility practices and procedures proved to be adequate to handle these few cases.

Stabilization of the last small groups of materials proved to be most problematic. These final 13 furnace runs consisted of an assortment of oxides (some of which were mixed Pu and U). These "cats and dogs" were items that had been stored for years and never dissolved. Some of the material came from off specification liquid processing activities. Others were one or two of a kinds sent from other DOE facilities. The contingencies developed for processing the chloride bearing materials proved to be invaluable during these final stabilization runs. The off gas line plugged numerous times. This resulted in the furnace shutting down and restabilization after the off gas line had been cleaned (pluggage in the 1/8 in. line occurred very near the furnace). Some of these runs resulted in a hard crust forming over the oxide in the stabilization pan. The crust had to be broken up prior to packaging. These runs resulted in moderate to significant furnace corrosion. In one such run the material hardened during stabilization to the point that several hours were required to break up the material. The end of the thermowell which is submerged into the oxide being stabilized ruptured when the stabilization pan was lowered. This run also resulted in a significant quantity of fluffy salt waste being generated (after several days it became more powder and crystalline).

E. <u>Documentation</u>

Each site used a slightly different process for stabilizing and packaging material. The original database format and requirements document was based on the process that was set up at Rocky Flats. The format was set up to match their process and record information based on how they generated cans. As additional sites began packaging per the 3013 standard, the database had to be modified or backfit to support the variations in each sites process. Due to these differences, some of the information important to surveillance may be in a scanned procedure instead of input as a sortable field in the database. Also, many of the electronic files are in different formats. These differences could end up burdening the surveillance program.

IV. CONCLUSIONS

A total of 919 outer 3013 containers (617 metal and 302 oxide) of stabilized plutonium bearing materials were produced from April 2003 through January 2005. Implementation was in two phases with metals beginning in April, 2003 and oxides beginning in October, 2003. This very aggressive campaign averaged nearly 19 stabilized oxide containers per month (from October, 2003 to January, 2005), and 28 metal containers per month (from April, 2003 to January, 2005). The overall average was nearly 42 containers per month for 22 months.

A summary of lessons learned include: First, a careful and thorough review of work previously completed should be performed prior to crediting the work as being programmatically compliant (e.g. previously packaged metal items). Second, paperwork and radiographs of legacy materials are often inconclusive and even misleading. A "legacy" review team was used to carefully evaluate "legacy" materials and containers. Using a graded approach, they provided guidance for operators to ensure safe handling and processing of these "legacy" materials. Third, as expected, stabilization of impure oxides results in significant furnace degradation. Fourth, as expected, a few materials required longer furnace runs to ensure complete stabilization. Fifth, the current 3013 database contains a vast quantity of useful information which may become cumbersome to efficiently use during surveillance.

V. ACKNOWLEDGMENTS

The information contained in this article was developed during the course of work under Contract No. DE-AC09-96SR18500 with the U.S. Department of Energy.

VI. REFERENCES

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