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Materials Identification and Surveillance Project Item Evaluation

Item: Impure Plutonium Oxide (HRA905191)

Los Alamos
NATIONAL LABORATORY

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by

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1.0 INTRODUCTION

We report the demonstration of the canister puncturing system used to characterize gas evolution from impure plutonium oxide materials stored in accordance with the Department of Energy (DOE) standard DOE-STD-3013-96.[1] The gas sampling operation is applied to a can containing an impure mixture of plutonium oxide and thoria (27 g of plutonium and 78 g of thorium). This is the third impure plutonium oxide sample to be evaluated by this project.

Methods used to characterize the cans include photography and x-ray radiography. Methods used to characterize the oxide include x-ray powder diffraction and gas analysis mass-spectrometry.

2.0 EXPERIMENTAL METHODS

2.1 Materials

The history of this material is as follows: This actinide material (plutonium and thorium) was originated at Argonne National Laboratories, was repackaged at Hanford, and was sent to Los Alamos in 1980. A slightly bowed container lid suggested that the cans were possibly pressurized. The lid on the inner can was cocked as if internal pressure had opened the can.

2.2 Procedures

The canister was photographed at three different orientations, 0, 120 and 240 degrees. Also the top and bottom of the can were photographed. The photograph of the canister at 0 degrees can be seen in Figure 1. Next the canister was radiographed, also at three different orientations, 0, 120 and 240 degrees, see Figures 2-4.

The outer container, which was in good condition, was punctured, and the gas phase between the outer and inner container was sampled and analyzed with a mass spectrometer plumbed to the gas equipment. Next, the inner container was punctured, and the gas phase inside the inner container was sampled and analyzed. The outer container was opened with a can opener, see Figures 5 and 6. Inside the outer container, the inner container was enclosed in a plastic bag sealed shut with masking tape, see Fig. 7. The inside of the outer container was coated with a layer of brown-red powder, and copper shot was observed in the bottom of the can.

The inner container was sealed with black electrical tape, which was still elastic with adhesive in working condition, see Fig. 8. Its outside appeared corroded, particularly on the bottom seam. The inner container was opened, and it was found to be filled with a number of different things, a long plastic tube, a ribbed container with a metal lid which sat on its side, emery cloth, and paper, see Fig. 9. The inside of the container was covered with a brownish red powder or deposit. The plastic tube was sealed with black electrical tape and was brittle. It contained thin metal pieces, see Fig. 10. The ribbed container was closed with a screw type metal lid, and the metal lid was sealed with black electrical tape, see Fig. 11. The container was approximately three-quarters filled with a metallic gray powder, see Fig. 12. The powder had leaked out of the container and could be seen on the tape that held the lid on. The gas sampling fixture for the containers can be seen in Fig. 13.

The sample for x-ray powder diffraction consisted of three parts: a gray powder, a red powder, and various pieces of metal. Portions of the powders were taken for x-ray diffraction; only the largest piece of metal was analyzed. The powder samples were prepared by mixing with LaB-6, which was used as an internal standard, and then spreading the powder on double sided tape, which had been placed on a 1.25-in. metallographic mount. The largest piece of metal was mounted in an epoxy metallographic mount and polished; no internal standard was used. A Scintag XDS 2000 with a rotating sample holder was employed to collect the diffraction data. The tube was operated at 45 kV and 40 mA.

3.0 RESULTS AND DISCUSSION

After opening the inner container, it was realized that it was not pressurized. Instead, the inner can was cocked because it held a vial that was taller than the slip top can. The gas phase composition of the inner and outer can was found to be the same, Table I. This is in accord with the fact that the inner container was sealed only with a tape. Oxygen and nitrogen were depleted from the can atmosphere. Hydrogen and carbon dioxide were found in substantial amounts.

The x-ray powder diffraction results of the samples are summarized as follows. The gray powder was determined to be ThO_2, face-centered cubic, space group Fm3m, with a lattice constant of 5.593(3) Å. The red powder was found to be amorphous, and no further work was done on the red powder. The large piece of metal was found to be a two-phase mixture of Pu_7Th_3 and a solid solution of 80 % Th and 20 % Pu. The solid solution had a lattice constant of 5.0197(4) Å, which was used to deduce the composition from data supplied by D. M. Poole et al.[2]

Table I. Gas Phase Composition of the Inner and Outer Can

Species	Quantity (mole %)
CO_2	10.4
N_2	4.94
H_2	38.5
Ar	0.4
H_2O	0.07
O_2	0.006
He	0.02

4.0 CONCLUSIONS

The following conclusions were reached in the course of this study for impure oxide HRA905191:

- The cans were not pressurized.
- The oxygen gas in the cans was removed, apparently by reacting with the actinide materials, and hydrogen was produced.
- Nitrogen was depleted from the can atmosphere.
- The can puncturing system and gas analysis system works well.

ACKNOWLEDGMENTS

The experimental work was performed in part by John Telford and the photography work by Michael D. Greenbank and Leo J. Riedel. We wish to thank them for performing this work that aided our research efforts.

REFERENCES

1. Criteria for Preparing and Packaging Plutonium Metals and Oxides for Long-Term Storage. DOE-STD-3013-96.
2. D. M. Poole et al., J. Inst. Met., 86, 172, (1957).

Fig. 1. Photograph of the canister at 0 degrees.

Fig. 2. Radiograph of the canister at 0 degrees

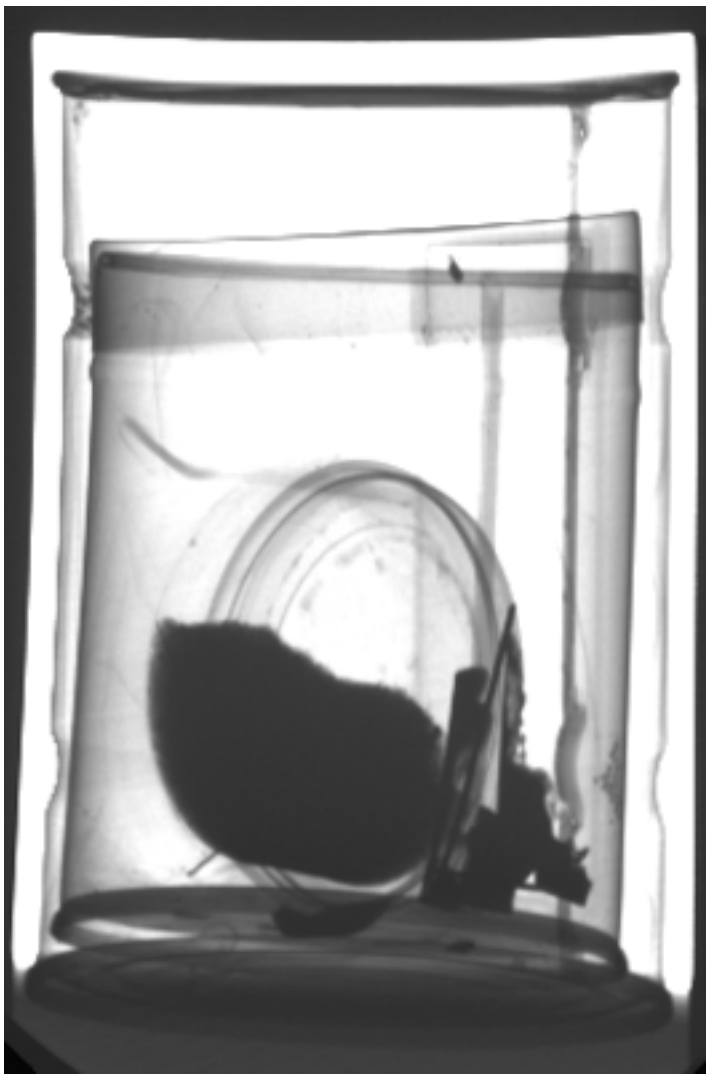

Fig. 3. Radiograph of the canister at 120 degrees.

Fig. 4. Radiograph of the canister at 240 degrees.

Fig. 5. Punctured top of the outer container.

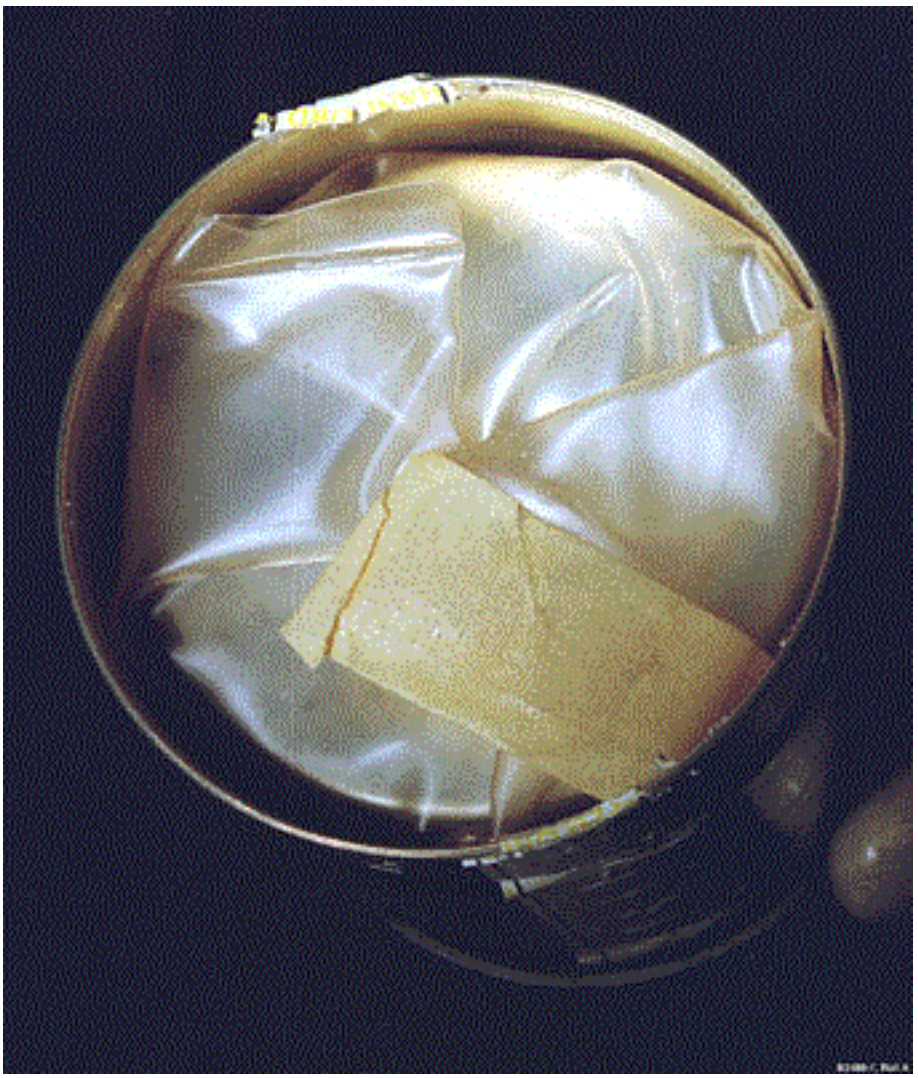

Fig. 6. Outer container opened.

Fig. 7. Inner container enclosed in plastic bag sealed with masking tape.

Fig. 8. Inner container sealed with black electrical tape.

Fig. 9. Inner container opened.

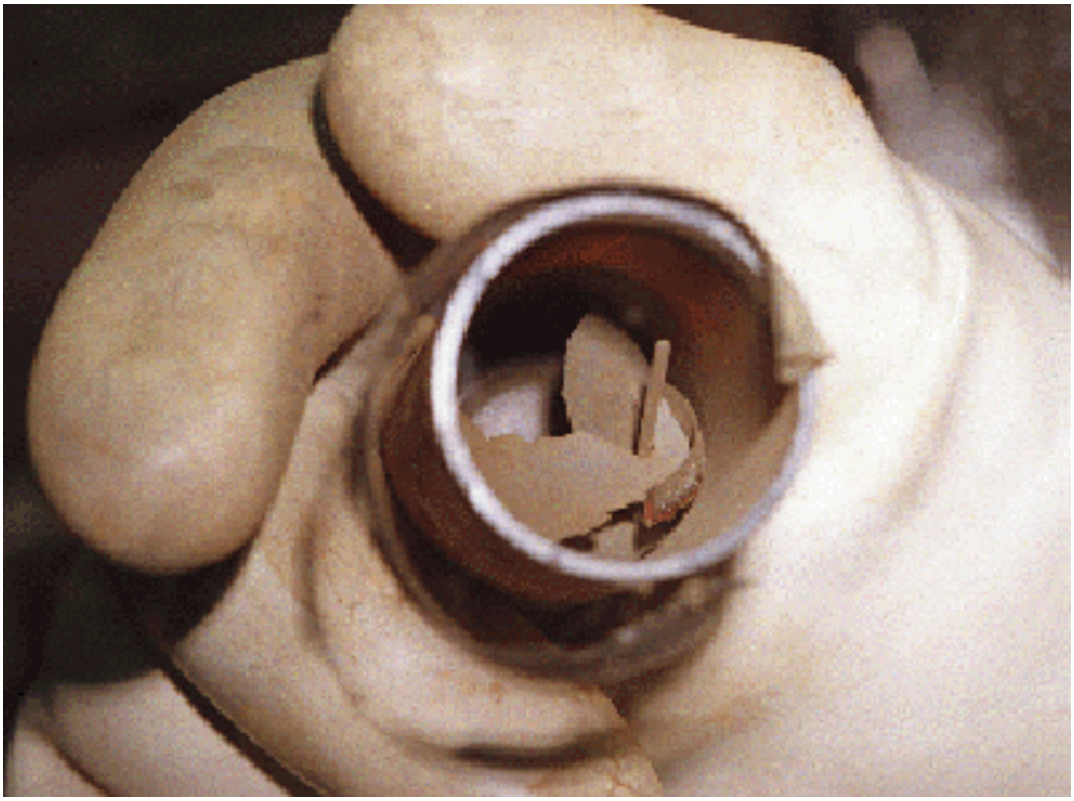

Fig. 10. Plastic tube contained thin metal pieces.

Fig. 11. Ribbed container with a screw type metal lid.

Fig. 12. Metallic gray powder.

Fig. 13. Gas sampling fixture.