Materials Identification and

Surveillance Project Item Evaluation

Item: Impure Mixture of Plutonium Oxide and
Uranium Oxide (PUUOXBC05)

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by

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ABSTRACT

In this report Los Alamos researchers characterize properties relevant to storage of an impure mixture of plutonium oxide and uranium oxide (impure mixed oxide [MOX] that is composed of 43.8 mass % plutonium and 17.8 mass % uranium) in accordance with the Department of Energy (DOE) standard DOE-STD-3013-96. This is the first sample of an impure mixture of plutonium oxide and uranium oxide to be evaluated by the Materials Identification and Surveillance Project.

Methods used to characterize the mixture include mass loss-on-calcination measurements, mass loss-on-ignition (LOI) measurements, elemental analysis, plutonium and uranium isotopic analysis, particle analyses measurements, X-ray powder diffraction, thermal desorption mass spectrometry (TDMS), and surface-area analyses. LOI measurements show a steady decrease in magnitude as the calcining temperature is increased. In contrast, calcining at progressively increasing temperatures does not appear to significantly change the specific surface area of the impure MOX. The LOI value for the powder after final 950°C calcination is 0.4 mass %. Water and carbon dioxide are the major gaseous products formed at all temperatures.

1.0   INTRODUCTION

The criteria for preparing and packaging plutonium metals and oxides containing >50 mass % plutonium for long-term storage are described in the Department of Energy (DOE) standard DOE-STD-3013-96.[1] Although the standard is applicable to pure plutonium dioxide, PuO_2,[2] questions remain for applying these methods to impure materials. This particular impure mixed oxide (MOX), item PUUOXBC05, contains 43.8 mass % plutonium and 17.8 mass % uranium, and does not satisfy the DOE-STD-3013-96 criterion for plutonium content (>50 mass % plutonium);[1] however, after calcination the material contains 48.85 mass % plutonium. Within the DOE complex there is a large inventory of mixed plutonium-uranium oxides of varying ratios that need to be stabilized and stored.

The objective of this study is to continue our experimental effort to characterize impure plutonium oxides and to develop preparation methods that convert these materials into forms suitable for extended storage.

2.0   EXPERIMENTAL METHODS

2.1   Materials

This impure mixture of plutonium oxide and uranium oxide powder, item PUUOXBC05, was produced and then sealed in a food-pack container in November 1983. The can was opened and inspected in September 1996: thus, this material was stored in the Technical Area (TA) 55 vault for almost 13 years. The inner can was contained in a plastic bag. The plastic bag was thin and slightly torn on the bottom at the can seal lip. The inner can was slightly corroded on the bottom seal. The plastic bag and inner can were contained in a stainless steel outer dressing can that is in like-new condition. The oxide powder, a very dark brown color, filled approximately 3/4 of the inner can. Its chemical history is unknown.

2.2   Procedures

The experimental procedures used to measure loss on calcination, loss on ignition (LOI), specific surface area, and particle parameters are described in previous Los Alamos memoranda and reports.[2,3,4] The sequence of sampling and testing is presented in Fig. 1.
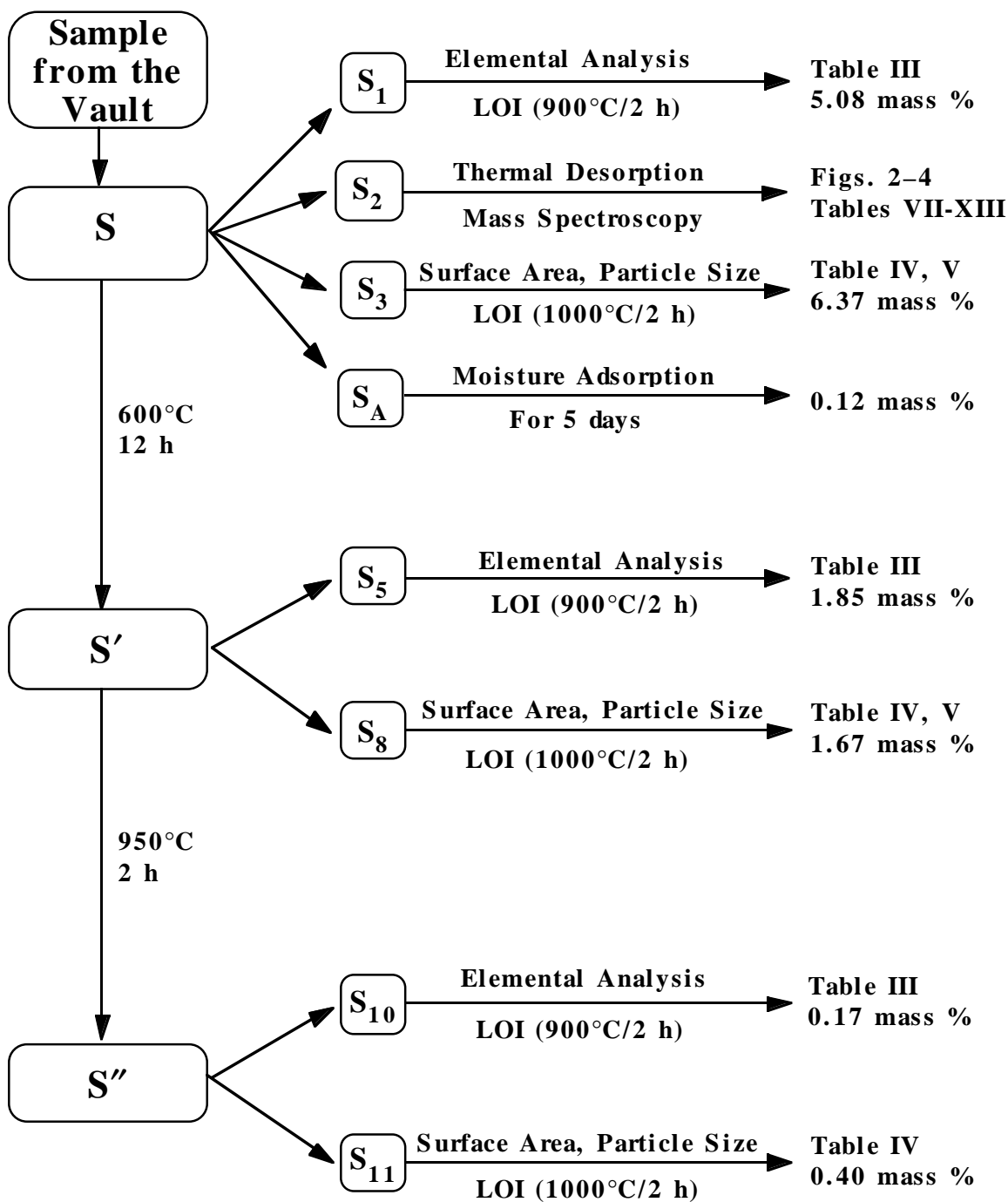
Fig. 1. **Characterization of the impure mixture of plutonium oxide and uranium oxide.**

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After samples were taken from the as-received material, sample S (the remaining powder) was calcined at 600°C to produce sample S'. After samples were taken from the 600°C calcined powder, all of the remaining powder was calcined at 950°C to produce sample S".

In brief, LOI data were obtained as follows. Each of three sample sets was halved and placed in a platinum calcining boat. All six boats were heated in the LOI furnace during the same furnace run. The furnace was ramped to 1000°C in approximately 4 h, held at 1000°C for 2 h, and cooled to 200°C in approximately 12 h. The samples were held at 200°C until they were removed from the furnace. After removing the samples from the furnace, they were immediately placed in a desiccator until they were cooled to room temperature and weighed. Materials S and S′ were placed in ceramic calcining boats and then calcined in a calibrated muffle furnace.

Elemental analyses were performed on the as-received precalcined powder (sample S), 600°C calcined powder (sample S′), and 950°C calcined powder (sample S″). Uranium content was measured and plutonium and uranium isotopic analyses were performed only for the as-received material.

Before particle size analysis was performed, each sample was heated at 350°C for 16 h in a vacuum. Particle size distribution was analyzed on a NOVA-1000 Brunauer-Emmett-Teller (BET) Surface Area Analyzer™. Five-point BET analysis was used to determine the specific surface area and a single-point specific surface area analysis was also used. Each powder set (precalcined, 600°C calcined, and 950°C calcined) was split and measured using both the five-point BET analysis and the single-point specific surface area analysis.

Samples S_1, S_5, and S_{10} were examined by X-ray powder diffraction (see Fig. 1). The samples were prepared for X-ray diffraction by light grinding with an Al_2O_3 mortar and pestle. A 1 mg portion of the sample was then loaded into a 0.2 mm quartz capillary. The capillary was cut to the appropriate length, sealed with epoxy, and centered in a Debye-Scherrer™ camera containing Kodak DEF-392™ film. The sample was exposed to nickel-filtered copper radiation for 6.5 h; the X-ray tube settings were 35 kV and 25 mA.

Thermal desorption mass spectrometry (TDMS) was performed on the as-received sample S_2 (see Fig. 1). An amount of 9.959 g of the material was placed in a 10 cm^3 tantalum cup. The weighed cup and oxide were placed inside a magnesium oxide, MgO, crucible and this assembly placed in the reactor constructed of a 1.5-in. schedule 40 stainless steel tube approximately 16-in. long with a welded plug on the bottom and a

Conflat™ flange on the top. A thermocouple pass through and high-pressure nipple were welded to a mating Conflat™ flange. When the reactor was assembled, the thermocouple was located between 0.25 in. and 0.5 in. above the oxide surface. The reactor was connected to the pressure vacuum system, evacuated, and leak checked. A 1200°C clamshell tube furnace is placed around the reactor that was maintained under dynamic vacuum conditions during heating. The furnace was programmed for an 8 h rise from room temperature to 950°C. The residual gas analyzer (RGA) is calibrated using standard procedures. Mass spectra were taken approximately every 15 min, and the reactor temperature and pressure were recorded throughout the run.

3.0   RESULTS AND DISCUSSION

Before the first calcination, 112.6 g of powder S was set in a glovebox. After 5 days of exposure to the glovebox atmosphere, the weight of powder S increased by 0.12 mass %.

Each time the powder was calcined the percent mass loss was measured. During the second calcination step the percent mass loss was half of the percent mass loss during the first calcination step. The results are shown below in Table I.

Table I.   Percent Mass Loss During Calcination of Sample PUUOXBC05[a]

Calcination Temperature (°C)	Heating Time (h)	Mass Before Calcination (g)	Mass After Calcination (g)	Mass Loss (%)
600	12	1 160.2	1 108.4	4.4
950	2	1 058.7	1 036.3	2.1

[a]The whole as-received batch was first calcined at 600°C. After sampling the 600°C calcined powder, this entire batch was calcined at 950°C.

Table II shows that LOI values steadily decrease in magnitude as the calcining temperature is increased. The LOI value (at 1000°C for 2 h) for the 950°C calcined powder is 0.4 mass %. The effect of the temperature at which the LOI measurement is performed is noticeable.

Table II. LOI Analyses Results of the Impure Mixture of Plutonium Oxide and Uranium Oxide

LOI Conditions	Powder S[a] LOI (mass %)	Powder S′[b] LOI (mass %)	Powder S″[c] LOI (mass %)
900°C for 2 h	5.08	1.85	0.17
1000°C for 2 h	6.37	1.67	0.40

[a]Powder S is the impure oxide from the vault before any calcination.
[b]Powder S′ is obtained after impure oxide S is calcined at 600°C for 12 h.
[c] Powder S″ is obtained after impure oxide S' is calcined at 950°C for 2 h.

Plutonium, nickel, calcium, copper, magnesium, and zinc concentrations increased after the 600°C calcining step and subsequently decreased after the 950°C calcining step. Lithium and carbon were the only elements for which concentration decreased after both the 600°C and the 950°C calcining. Table III shows these results and the elements found.

The as-received powder S has a specific surface area of 1.05 m^2/g; and the 950°C calcined powder S" has a specific surface area of 0.78 m^2/g. The specific surface area of 600°C calcined material (sample S′) is larger than the specific surface area of both the as-received and the 950°C (sample S″) calcined material. One of the two measurements at 600°C was over 70% larger than the other; so, one of the two measurements may be in error. Nevertheless, for this impure mixed oxide (PUUOXBCO5) heating the powder to 600°C and 950°C does not appear to significantly change its specific surface area. In contrast, the previous oxides studied, ATL27960 and PEOR3258,[5] show that calcination significantly changed their specific surface areas (see Table IV). The two impure oxides calcined at 950°C show almost identical specific surface area values of about 0.8 m^2/g. This value of specific surface area for the impure oxides is six times smaller than that for the pure plutonium oxide. This smaller value can be attributed to sintering during the heating process facilitated by the presence of inorganic salts as impurities.

Table III.   Elemental Analysis of Precalcined Powder S and Calcined Powders S′ and S″

Element	Powder S[a] (μg/g)	Powder S′[b] (μg/g)	Powder S″[c] (μg/g)
Pu	438 100	496 500	488 500
U	177 700	not done	not done
C	230	120	90
Ca	16 000	19 000	16 000
Cu	60 000	90 000	19 000
Fe	1 600	1 500	1 700
Ga	870	1 130	1 160
K	2 500	2 600	1 000
Li	330	200	175
Mg	52 000	61 000	47 000
Na	1 500	1 400	1 400
Ni	365	495	450
Zn	57 000	61 000	54 000

[a]Powder S is the impure oxide from the vault before any calcination.
[b]Powder S′ is obtained after impure oxide S is calcined at 600°C for 12 h.
[c]Powder S″ is obtained after impure oxide S′ is calcined at 950°C for 2 h.

Table IV.   Comparison of Specific Surface Area Results for Pure Plutonium Oxide, Impure Plutonium Oxide, and Impure MOX

Oxide	As-received (m²/g)	Calcined at 600°C (m²/g)	Calcined at 950°C (m²/g)
Pure plutonium oxide[a]	11.3	10.2	4.8
Impure plutonium oxide[b]	15.6	4.3	0.8
Impure MOX[c]	1.05	1.75	0.78

[a]Data for the pure plutonium oxide (PEOR3258) are from Ref. 2.
[b]Data for the impure plutonium oxide (ATL27960) are from Ref. 5.
[c]Data for the impure mixed oxide (PUUOXBC05) are in this report.

As expected, particle analysis showed an increase in the values of spherical equiva-lent mean and diameter-by-volume mean after the first calcination step, but particle analysis unexpectedly showed a decrease in these values after the second calcination step (see Table V).

Table V.   Particle Analysis Results of Precalcined Powder S and Calcined Powders S′ and S″

Property	Powder S[a]	Powder S′[b]	Powder S″[c]
Spherical equivalent mean (μm)	14.2	19.1	16.65
Diameter-by-volume mean (μm)	50.35	75.05	60.9

[a]Powder S is the impure oxide from the vault before any calcination.
[b]Powder S′ is obtained after impure oxide S is calcined at 600°C for 12 h.
[c]Powder S″ is obtained after impure oxide S′ is calcined at 950°C for 2 h.

The isotopic distribution of plutonium and uranium was also measured for the as-received powder S and the results are shown in Table VI.

Table VI.   Isotopic Distribution of Plutonium and Uranium for the "As-received" Powder S

Isotope	Atomic %	Mass %
Pu-238	0.0142	0.0141
Pu-239	94.8383	94.8171
Pu-240	4.9999	5.0197
Pu-241	0.1156	0.1165
Pu-242	0.0322	0.0325
Pu-244	0.0000	0.0000
Total	100	100
U-233	0.0000	0.0000
U-234	0.0498	0.0490
U-235	4.3734	4.3206
U-236	0.0474	0.0470
U-238	95.5295	95.5835
Total	100	100

The X-ray powder diffraction results for samples S_1, S_5, and S_{10} are summarized as follows. The as-received material, S_1, was determined to be a face-centered cubic, space group Fm3m, solid solution of PuO_2 and UO_2 with a lattice constant of 5.4067(5) Å. The composition of this solid solution, based on the lattice constant, was $Pu_{0.82}U_{0.18}O_{2+x}$.[6,7] However, this formula does not agree very well with Pu:U ratio based on chemical analyses (see Table III). In addition, CuO and MgO were found as minor phases. Sample S_5 was found to be $Pu_{0.82}U_{0.18}O_{2+x}$, with a lattice constant of 5.3973(5) Å, and with CuO and MgO as minor phases. Sample S_5 contained less CuO than sample S_1. Sample S_{10} was a mixture of $Pu_{0.82}U_{0.18}O_{2+x}$, with a lattice constant 5.3832(8) Å, and MgO. Copper oxide was not found. The lattice contraction upon firing indicated that a substantial amount of uranium was oxidized to +5. The oxygen stoichiometry could not be determined quantitatively, however $0 < x < 0.12$. Based on this range of oxygen composition, the maximum mass increase due to the oxidation of UO_2 in the sample is 0.72 g per 100 g of sample.

The TDMS results for sample S_2 are summarized as follows. A plot of reactor temperature and pressure as a function of time is shown in Fig. 2. A plot of reactor pressure as a function of temperature is shown in Fig. 3. Figure 4 shows the major gas constituents as a function of temperature. A large pressure spike is observed in Fig. 2 at approximately 2 h, and the temperature of the reactor is approximately 150°C. The temperature plot in Fig. 2 shows an endothermic drop in temperature at 125°C that directly corresponds to the pressure rise. This endothermic process appears between 125°C and 225°C while water is vaporized. An endotherm appears when the observed temperature is lower than the programmed temperature. The lower temperature remains as long as the endothermic process continues to cool the sample. A false exotherm is observed after the endothermic process is completed and the temperature recovers.

The gas composition is shown in Table VII. At this point, the major constituent of the gas mixture is water. The pressure then drops slightly (Fig. 3) followed by a slight pressure increase between 2.4 h and 2.75 h. The gas analysis shows a loss of O_2 and CO_2 and a small increase in the water concentration (Table VIII). Some minor unidentified peaks are also present. The water pressure now drops off radically with a small shoulder observed at about 275°C (Table IX). Figure 4 shows C_2H_6, CO_2, and CO are essentially constant between 150°C and 350°C. Their percent amounts increase because water decreases somewhat. All gases continue to fall off until a pressure minimum is reached at about 380°C. The pressure then starts to rise to a pressure plateau at about 510°C (Table X). This is followed by a sharp pressure rise at 570°C (Table XI). An exotherm appears between 525°C and 725°C where CO_2 is the major gaseous product. The gas composition

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Fig. 2. Reactor temperature and pressure as a function of time during thermal desorption of the impure mixture of plutonium oxide and uranium oxide.
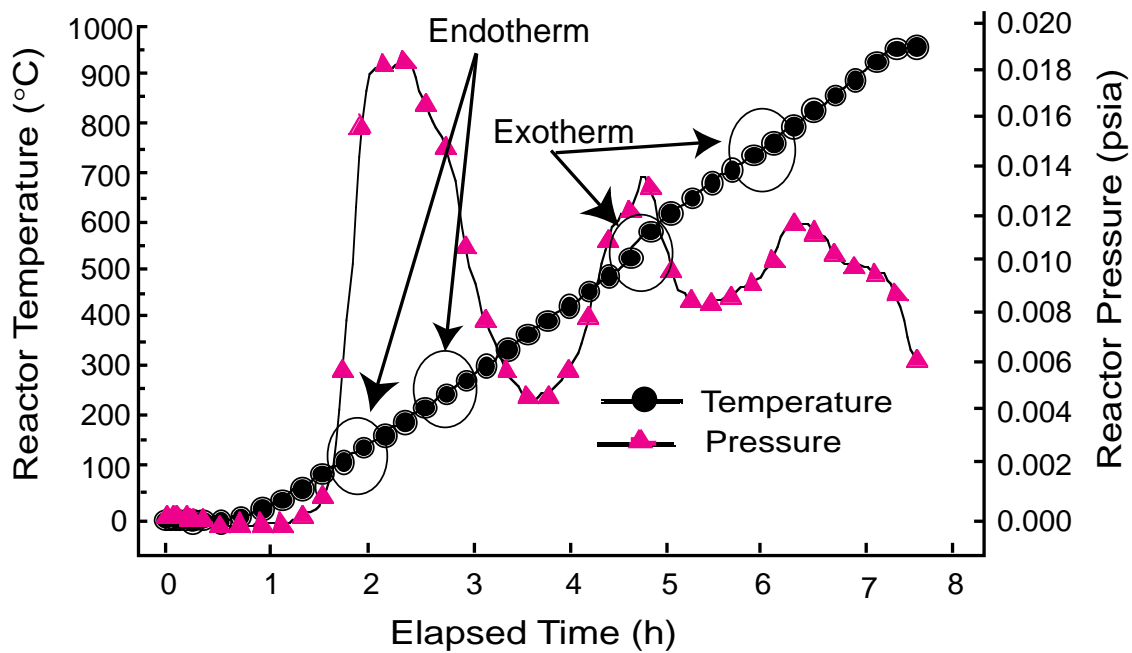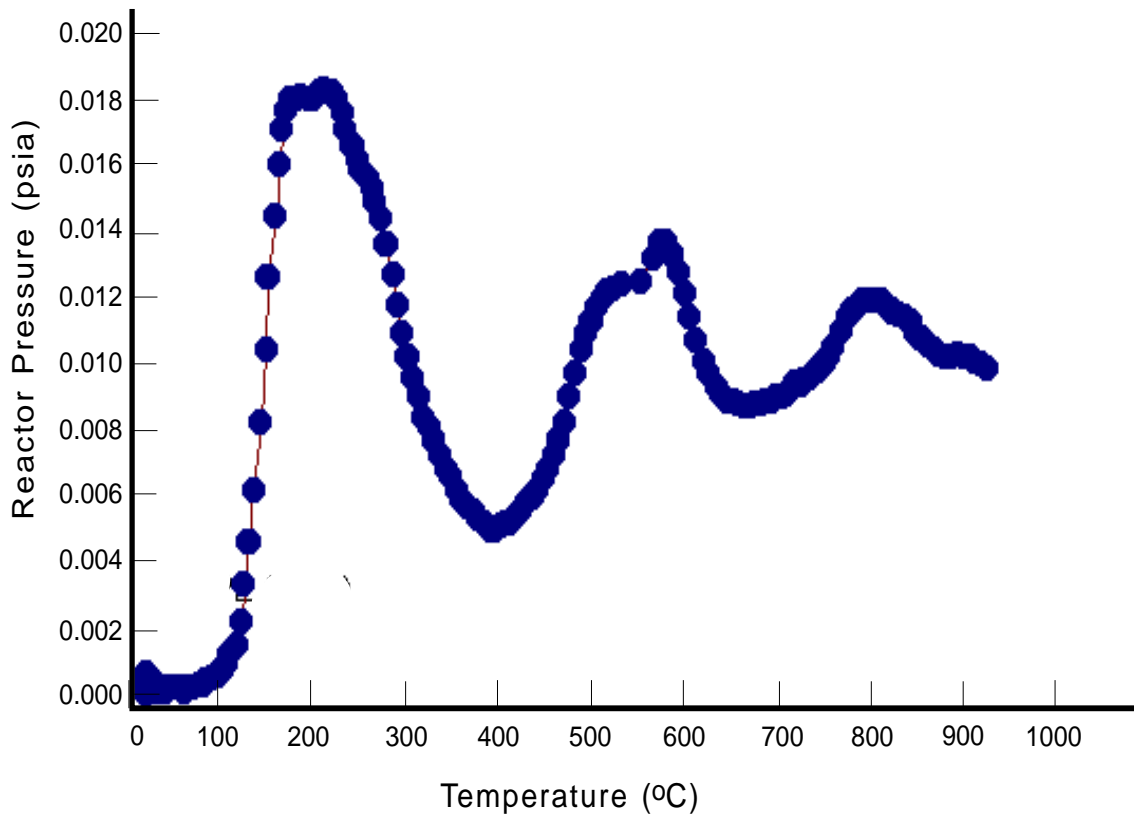
Fig. 3. **Reactor pressure as a function of temperature during thermal desorption of the impure mixture of plutonium oxide and uranium oxide.**
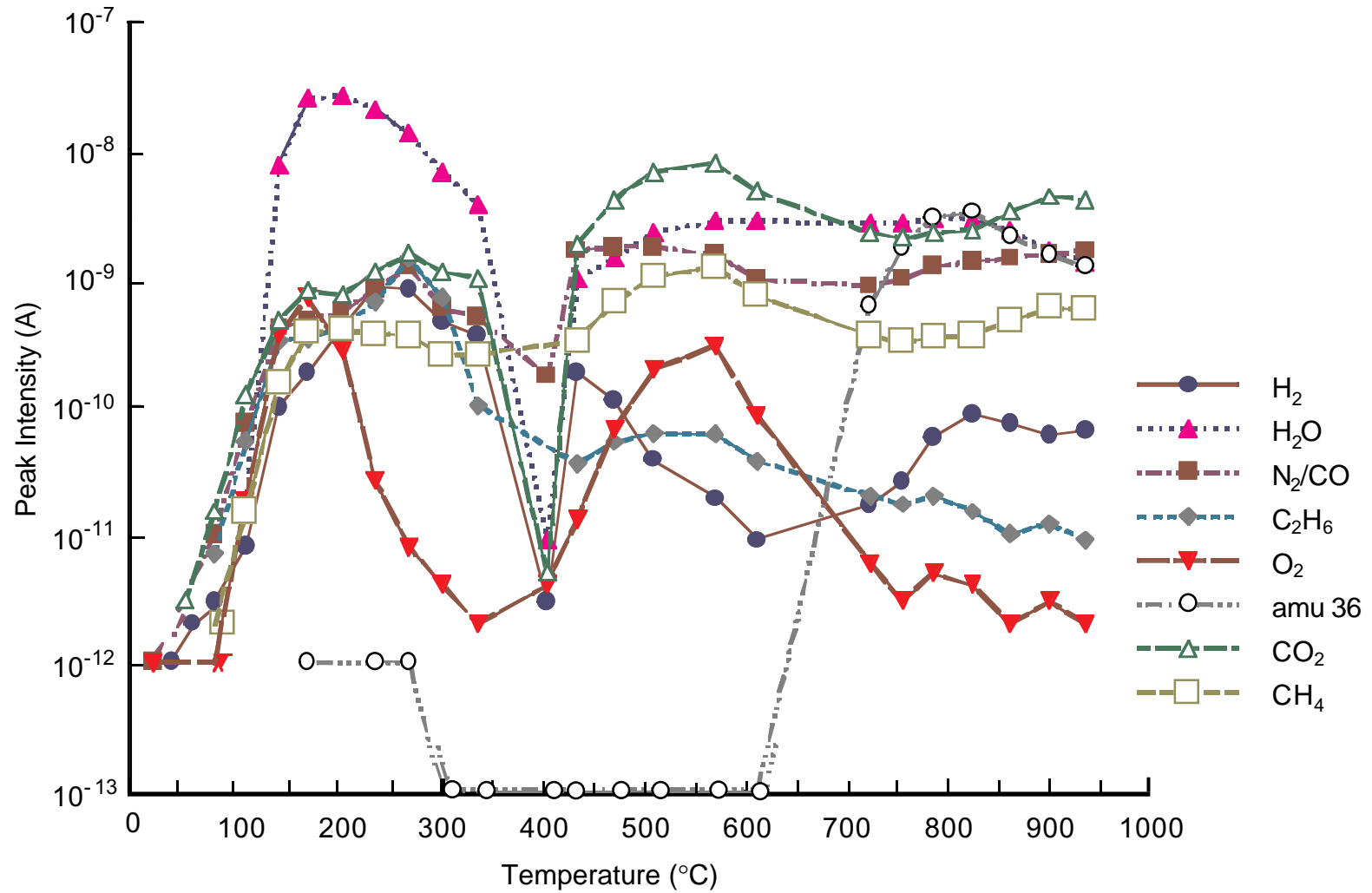
Fig. 4. Major gas constituents as a function of temperature during thermal desorption of the impure mixture of plutonium oxide and uranium oxide.

and distribution in this region does not change substantially. The pressure now drops to a second minimum followed by a rise to a pressure plateau at about 780°C (Table XII). The major constituent, atomic mass unit (amu) 36, has not been identified. The pressure drops off to a plateau at about 910°C (Table XIII), but the gas composition does not change radically.

The reactor was cooled and disassembled. A large amount of unidentified material was plated out on the cooler reactor surfaces. The tantalum cup and oxide were re-weighed and the mass loss, 11.6% on thermal treatment during TDMS (up to 1000°C within 8 h), was calculated based on the weight difference. This is about a factor of 2 greater than that measured by conventional LOI analyses (Table II). A possible explanation for the difference in these values may be that the TDMS was run under dynamic vacuum conditions, resulting in vaporization of some volatile residues. Mass loss may also be due to the tantalum cup used. It is known that Ta_2O_5 forms under oxidizing conditions and is highly volatile.

Water is seen throughout the process; however, the largest quantity is observed between 175°C and 340°C. The CO and CO_2 that evolved and the pressure stages suggest the decomposition of carbonates that formed during storage, as well as the desorption of adsorbed CO_2. The methane and ethane that evolve during thermal desorption may result from the decomposition of small amounts of hydrocarbon contamination in the sample, or they may result from catalytic reaction of hydrogen and carbon on platinum-rich oxide surface. Chemical analyses of the sample show, in addition to plutonium and uranium, high quantities of sodium, zinc, potassium, lithium, calcium, and magnesium, but the specific chemical forms of these six elements are unknown. The residues plated out on the reactor surface probably contain these materials. TDMS measures only permanent gases. Volatile inorganics are plated out on the cooler surfaces of the reactor and do not reach the analyzer. There were no peaks observed past 90 amu. Many unidentified peaks were observed at the higher temperatures (Tables XII and XIII), but make up only about 0.5% of the total gas mixture. Attempts to match these peaks with volatile inorganic species failed. At this temperature it is unlikely that complex organic molecules would survive. Work will continue to identify these peaks.

In mass spectroscopy, the gas molecules that comprise the analyte are ionized. The ions are then accelerated across a potential difference, through a mass filter, to a detector. The mass filter separates the ions so that only a specific ion reaches the detector. The results are generally displayed as a table or graph showing the ratio of ion mass to charge

in contrast to the ion current (in amperes). The ion current is directly proportional to the amount of that ion present in the analyte. The quantitative analysis of mass spectra requires that each mass peak be identified as a parent or daughter peak of the compounds in the analyte. After each peak is identified, it is normalized to nitrogen, based on the ionization cross section for that compound. The quantity of that molecule is directly related to the total normalized ion current for that molecule. The quantity of a specific molecule in a given analyte is calculated using the sample pressure, molecule ion current, and the total ion current for the analyte. In the data from this test it was not possible to identify all of the peaks and, therefore, the quantitative results described above are not presented. The ratio of the ion current of a specific peak over the total ion current is presented in the tables below and represents approximately a ±10% error in the amount of a given molecule when compared with real quantitative results.

Table VII.   Thermal Desorption-Gas Composition of Sample S$_2$ at 175°C

Species or amu	Quantity (Ratios Expressed as %)
H_2O	90.77
CO_2	2.99
O_2	2.57
CO	1.74
C_2H_2	1.25
H_2	0.68

Table VIII.   Thermal Desorption-Gas Composition of Sample S$_2$ at 210°C

Species or amu	Quantity (Ratios Expressed as %)
H_2O	91.6
CO_2	2.5
CO	1.9
H_2	1.4
C_2H_2	1.4
O_2	0.95
20	0.18
40	0.021
61	0.010

Table IX.   **Thermal Desorption-Gas Composition of Sample S_2 at 275°C**

Species or amu	Quantity (Ratios Expressed as %)
H_2O	72.4
CO_2	8.4
C_2H_2	7.8
CO	6.7
H_2	4.5
20	0.13
O_2	0.045
50	0.033
40	0.017

Table X.   **Thermal Desorption-Gas Composition of Sample S_2 at 520°C**

Species or amu	Quantity (Ratios Espressed as %)
CO_2	60.8
H_2O	20.6
CO	15.9
O_2	1.78
C_2H_2	0.57
H_2	0.36
20	0.037
64	0.009

Table XI.   **Thermal Desorption-Gas Composition of Sample S_2 at 577°C**

Species or amu	Quantity (Ratios Expressed as %)
CO_2	62.7
H_2O	22.0
CO	12.2
O_2	2.4
C_2H_2	0.49
H_2	0.15
20	0.049

Table XII.   Thermal Desorption-Gas Composition of Sample S_2 at 780°C

Species or amu	Quantity (Ratios Expressed as %)
36	31.4
H_2O	30.3
CO_2	23.8
CO	13.0
H_2	0.62
C_2H_2	0.23
20	0.15
40	0.12
60	0.076
73	0.071
O_2	0.054
55	0.054
57	0.043
56	0.032
61	0.032
74	0.032
77	0.021
64	0.011
69	0.011
70	0.011
85	0.011
87	0.011

Table XIII.   Thermal Desorption-Gas Composition of Sample S_2 at 980°C

Species or amu	Quantity (Ratios Expressed as %)
CO_2	47.13
H_2O	17.95
CO	16.83
36	16.83
H_2	0.66
C_2H_2	0.13
20	0.079
60	0.067
56	0.045
57	0.045
73	0.045
O_2	0.036
55	0.034
61	0.034
40	0.022
85	0.022
69	0.011
74	0.011
87	0.011

4.0   CONCLUSIONS

We continue studies to define the conditions for preparing and handling impure plutonium oxide for long-term storage. It becomes progressively apparent, as it was expected, that the identity and concentration of impurities determine the physical and chemical behavior of the impure oxide.

We obtained the following results in the course of this study for the impure mixed oxide PUUOXBC05:

- Thermal treatment makes the impure MOX conform to the DOE-STD-3013-96[1] LOI requirement of <0.5 mass % loss tested at 1000°C for 2 h.
- Heating the powder to 600°C and 950°C does not appear to significantly change the specific surface area.
- The specific surface area of the powder calcined at 950°C is 6 times smaller than that of the pure plutonium oxide calcined at 950°C and is comparable to the specific surface area of the previously studied impure oxide (ATL27960), approximately 0.8 m^2/g. Impurities appear to facilitate sintering.
- For a 100°C increase (i.e., 1000°C for 2 h instead of 900°C for 2 h) an LOI of 0.40 mass % was found instead 0.17 mass % for the calcined sample S''. LOI conditions appear to be important for impure oxides.
- The calculated mass loss of 11.6% on thermal treatment during TDMS (up to 1000°C within 8 h) for the as-received sample S, is about a factor of 2 greater than that measured by conventional LOI analyses, 6.37% (at 1000°C for 2 h), possibly because the TDMS was run under dynamic vacuum conditions that resulted in vaporization of volatile residues. Another possible reason for the observed large value of 11.6% mass loss on thermal treatment during TDMS is the longer time used, 8 h compared with 2 h by conventional LOI analyses. The vaporization of tantalum (tantalum cup) as T_2O_5 on thermal treatment of the MOX sample during TDMS cannot be excluded.
- Chemisorbed water is the primary gaseous product observed during calcination of the oxide and appears throughout the TDMS process. The largest quantity of water is seen between 175°C and 340°C.
- The CO_2 that evolves during thermal desorption at low temperatures should be due to the desorption of absorbed CO_2. The CO_2 that evolves during thermal desorption at high temperatures suggests the decomposition of carbonates that formed during storage. Water and carbon dioxide are the major gases formed between 450°C and 950°C.
- The methane and ethane that evolve during thermal desorption may result from the decomposition of small amounts of hydrocarbon contamination in the sample, or they may result from catalytic reaction of hydrogen and carbon on a plutonium-rich oxide surface.

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