

*Materials Identification and Surveillance
Project Item Evaluation*

Item: Impure Plutonium Oxide (ARF-102-85-295)

*This work was supported by US Department of Energy,
Office of Environmental Management EM-66 Nuclear Materials
Stabilization Task Group.*

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MATERIALS IDENTIFICATION AND SURVEILLANCE PROJECT ITEM EVALUATION

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by

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ABSTRACT

In this report, we characterize properties relevant to storage of an impure plutonium oxide (26.59 mass % plutonium) in accordance with the Department of Energy (DOE) standard DOE-STD-3013-96.

Methods used to characterize the oxide include mass loss-on-calcination (LOC) measurements, mass loss-on-ignition (LOI) measurements, elemental analysis, plutonium isotopic analysis, particle analyses measurements, x-ray powder diffraction, thermal-desorption mass-spectrometry, and gas analyses. Methods used to characterize the container include x-ray radiography and photography.

Loss-on-ignition values are high due to the presence of high concentrations of chloride and stay relatively constant as the calcining temperature is increased. The material does not satisfy DOE-STD-3013-96 criteria for plutonium content and LOI.

1.0 INTRODUCTION

The requirements for extended storage of oxides containing >50 mass % plutonium are described in the Department of Energy (DOE) standard DOE-STD-3013-96.¹ Although procedures² are found to be valid for pure PuO₂ in accordance with this standard, questions remain for applying these methods to impure materials. This particular impure plutonium oxide item ARF-102-85-295 has 26.59 mass % plutonium and does not satisfy the criterion for plutonium content (>50 mass % plutonium). Within the DOE complex there is a large inventory of impure plutonium oxides of varying concentrations of different impurities awaiting stabilization and storage.

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 storage.

2.0 EXPERIMENTAL METHODS

2.1 Materials

The material ARF-102-85-295 was canned for shipment at Rocky Flats about 1985 and shipped shortly thereafter. Hanford processed much of this type oxide in the mid 1980s. To support dissolution processing, some items such as this one were sampled for analysis, then returned to the vault.

Table I shows the characterization data sent from Hanford site.

Table I. Characterization Data Sent from Hanford Site for the Impure Oxide ARF-102-85-295.

| | |
|-------------------------|--------------|
| Date Processed | 1985 |
| Gross weight (g) | 1035.5 |
| Net Weight (g) | 635 |
| LOI (wt.%) | <1.0 |
| Calorimetry Date | 25 June 1996 |
| Power (watts) | 0.4215 |

2.2 Procedures

Photography and Radiography

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 Fig. 1. Next the canister was radiographed, also at three different orientations, 0, 120 and 240 degrees. The radiograph of the canister at 0 degrees can be seen in Fig. 2. The radiographs suggested clumps or chunks of material but we were uncertain what form the material was really in. When the can was opened and the material was examined it became clear what we were seeing in the radiographs. The material was placed on the slip top can lid and photographed, Fig. 3. A 22-g piece of this material was calcined to determine its performance when heated in the calcining furnace. During the calcining run the furnace door was opened and the piece photographed. The temperatures in red indicate the temperature of the furnace when the photograph was taken. The last photograph is after the piece was heated at 950°C for two hours and then cooled to room temperature, see Fig. 4.

Gas sampling and analysis

For gas sampling the puncture device was epoxied to the top of the can, see Fig. 1 of the previous report.³ The sampling port was mated to the storage can using an O-ring and epoxy seal, which allows for a leak-tight connection to the punch assembly and volume-calibrated pressure-vacuum system (PVS). Two type-K thermocouples were also attached to the can. The can, sampling port assembly, and two gas sample vials are connected to the PVS, and the PVS is evacuated and flushed at least three times with high-purity helium gas. After the final flush, the PVS was evacuated to a pressure 5.0×10^{-6} torr and this evacuation was monitored on a residual gas analyzer (RGA). After the helium was pumped off, a rate-of-rise leak test is performed. The leak rate is acceptable if no argon gas is detected after 5 min under static vacuum and the pressure rise in the PVS system does not exceed 5.0 millitorr. All leaks were corrected before proceeding. The gas sample vials were valved out of the system and the PVS was isolated from the vacuum pump. The punch was unlocked and the can lid punctured. The punch was then retracted and locked in its starting (volume calibrated) position. The pressure was allowed to equilibrate between the can and the PVS. The equilibrium pressure and can temperature were recorded. The



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



Fig. 2. Radiograph of the canister at 0 degrees.



Fig. 3. ARF-actinide material on the slip top can lid.

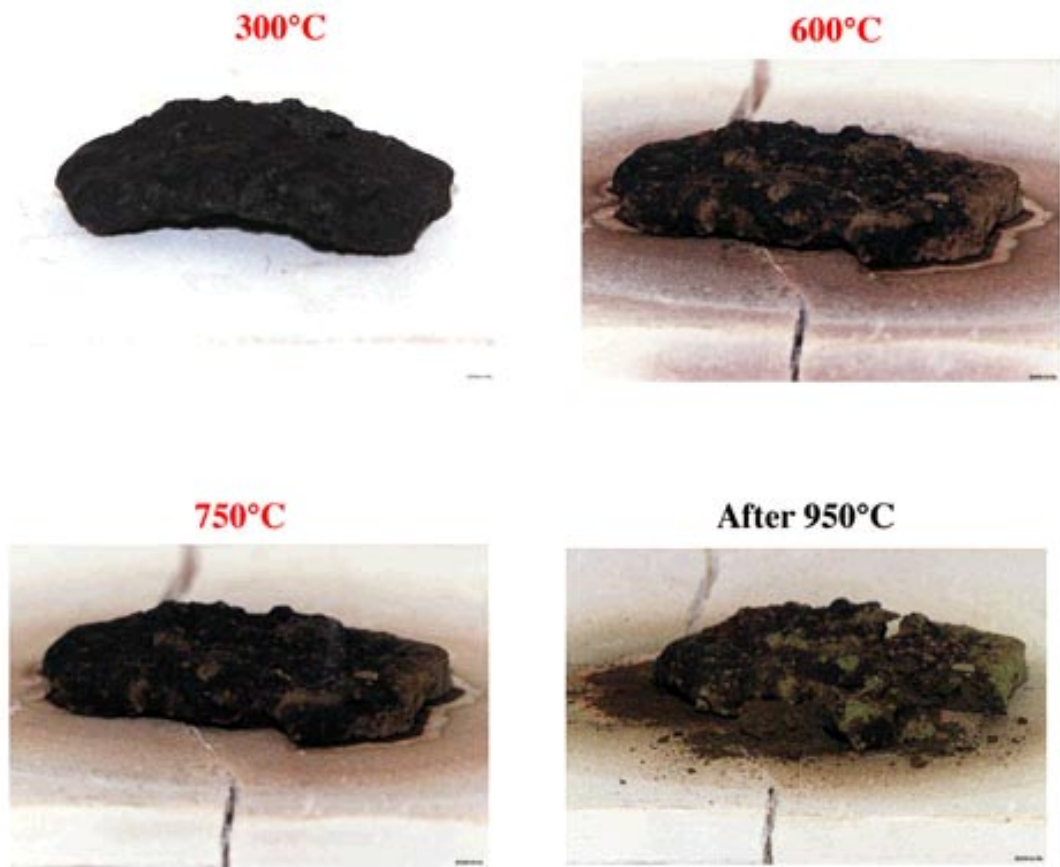


Fig. 4. Photos of the material while the furnace was at 300°C, 600°C, 775°C and after the piece was heated at 950°C for 2h and then cooled to room temperature.

initial can pressure was calculated based on the expanded equilibrium pressure, the can temperature, and the free volume of the food-pack can. The free volume of the food-pack can was calculated from measurements taken from radiographs of the can. A gas sample was captured in each of the two gas sample vials for future analysis, and the remaining gas in the PVS was analyzed using the RGA. The can puncturing, gas sampling and disassembly process has also been described in summary with photos in a previous report⁴. Next the can was removed from the PVS and opened, and the internal surfaces of the can were visually inspected and photographed. In nested configurations the outer surfaces of the nested can are also photographed. In cases where the nested can is another food-pack can, the puncturing process is repeated. The cycle is stopped when the inner can contains the oxide. Next the convenience container was opened and the temperature of the contents was measured in five places. The plutonium oxide was weighed on a material accountability and safeguards system (MASS) certified balance for accountability purposes. A 30-g sample of the material was then removed from the parent lot. Ten grams were used for thermal desorption mass spectrometry (TDMS), 10 g for long term storage experiments, and 10 grams held as an archive sample. The remainder of the item was sent for calcination and further tests. 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 reports.^{2,5,6,7}

The item was split into two portions, ARF102-295 remained as chunks, and ARF102-295A was rod-milled to powder. The sequence of sampling and testing is presented in Fig. 5. After samples were taken from the as-received material, (sample S), a portion of the 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''.

X-ray Diffraction Analysis

Samples were removed from ARF102-295 and ARF102-295A portions. From ARF102-295, sample ARF-3 was the as received material, ARF-5 was fired at 650°C, ARF-9 was fired at 900°C. From ARF102-295A, sample ARF-3a was the as received material, ARF-8 was fired at 650°C and ARF-13 was fired at 900°C. The samples were stored in screw-capped vials which were placed in an air-filled (approx. 40 % relative humidity) glovebox. The ARF samples were examined by x-ray powder diffraction using a SCINTAG XDS-2000 diffractometer. The samples were prepared for x-ray diffraction by

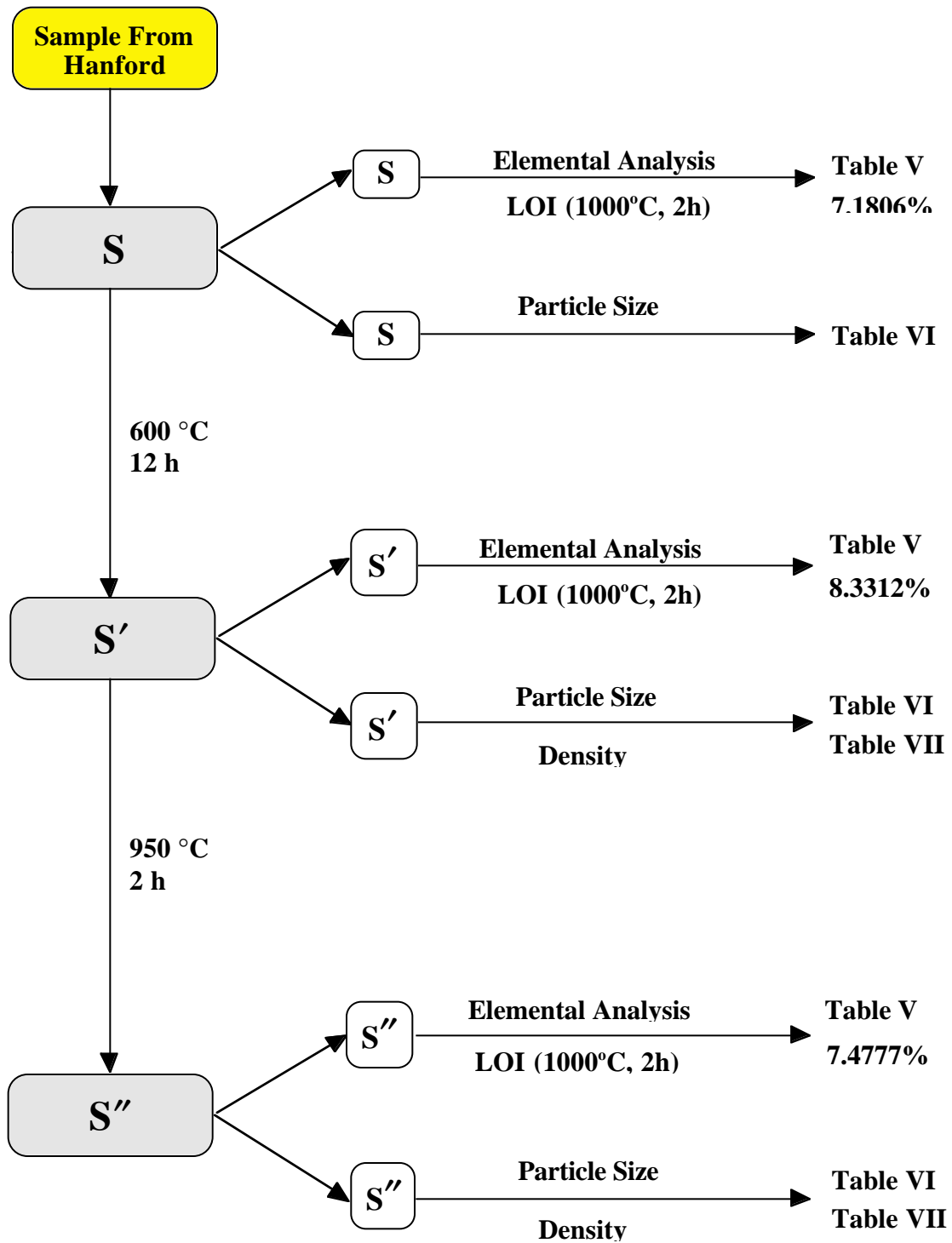


Fig. 5. Characterization of the impure plutonium oxide ARF-102-85-295.

light grinding with an Al_2O_3 mortar and pestle. A portion of the sample, approximately 50 milligrams, was then loaded on to strip of double-sided tape which was mounted on an epoxy cylinder 1.25 in diameter and 1.25 in tall. The sample and cylinder were then wrapped with plastic film, checked for contamination and then placed into a specially designed sample holder, which is mounted on to the goniometer. The correct sample height with respect to the detector-tube zero point plane was set with an external standard. Each sample was exposed to nickel filtered copper radiation; the x-ray tube settings were 45 kilovolts and 40 milliamps. A 0.05 degree beam slit, medium Soller slits, and a 0.02 degree detector slit was used. Diffraction data were collected from 25 to 135 degrees in two theta; the scan time was 6 h.

Thermal Gravimetric/Differential Thermal Analysis

The same samples were analyzed with a Rheometrics model PL-STA 2000 TGA/DSC with provided thermal gravimetric and differential thermal analysis (TGA/DTA) over a given temperature regime. The samples were consistently handled in the following way. Each sample was exposed to room air for a maximum of 15 min while that sample was weighed and then loaded into the apparatus. The samples were held in an alumina crucible. Once the sample was in the apparatus, the sample chamber, whose volume is approximately 15 cm^2 , was flushed with oil-free bottled air. The flow rate was 15 ml/ min; the sample chamber was purged for 3 min before the experiment was started.

Once the sample chamber was purged, a temperature program was initiated. This program sets the temperature to 25°C for a three minute isotherm and then ramp the temperature to 1000°C at a rate of $10^\circ\text{C}/\text{min}$. During this temperature ramp, the mass of the sample was monitored at a rate of one datum point per second. The experiment concluded when the temperature reached 1000°C . These conditions were selected to closely model the LOI experiments.

3.0 RESULTS AND DISCUSSION

Before the first calcination, 48.2266 g of S in chunks form was set in a glovebox. After 2 days of exposure to the glovebox atmosphere, the weight of S increased by 0.003 mass %. For the as-received milled powder weight gain on water uptake was 108.3984 final, less 108.3908 initial or 0.0076 g or 0.007% over seven days. These results are shown in Table II. It is interesting to notice that all different powders S, S', and S'' show very similar affinity for water. The values in Table II are most likely values for equilibrium

loading of water. As we have seen before⁵, for the impure plutonium dioxide ATL27960 calcined at 950°C for 2 h, one day was enough for equilibrium to be established.

Table II. Water Uptake Adsorption Measurements for the Impure Oxide ARF-102-85-295

| Time & Amount of Water | Powder S ^a | | Powder S ^b | | Powder S ^c | |
|---------------------------|-----------------------|--------|-----------------------|--------|-----------------------|--------|
| | Powder | Chunks | Powder | Chunks | Powder | Chunks |
| Time (days) | 7 | 2 | - | 5 | 2 | 2 |
| Mass (%) | 0.007 | 0.003 | - | 0.002 | 0.000 | 0.001 |

^aPowder S is the as-received material from Hanford.

^bPowder 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.

Each time the powder was calcined, the percent mass loss was measured. During the second calcination step the percent mass loss was much more than the percent mass loss during the first calcination step. The results are shown below in Table III.

Table III. Percent Mass Loss During Calcination of Sample ARF-102-85-295

| Calcination Temperature (°C) | Heating Time (h) | Mass Loss | Mass Loss |
|------------------------------------|------------------------|---------------|---------------|
| | | (%) Powder | (%) Chunks |
| 600 | 12 | 0.41 | 0.21 |
| 950 | 2 | 28.82 | 31.14 |

Table IV shows that LOI values are very high and they stay about the same regardless of thermal treatment. This can be explained on the fact that this material is high in chloride. The reported Hanford-LOI value is <1.0 mass %. This very low LOI value is understood because the LOI test was done only at 600°C instead at 1000°C. This impure oxide does not conform to the DOE-STD-3013-96¹ LOI requirement of <0.5 mass % loss tested at 1000°C for 2 h, even after treatment at 950°C.

Table IV. LOI Analyses Results of the Impure Oxide ARF-102-85-295

| LOI Conditions | LOI for S ^a (mass %) | | LOI for S' ^b (mass %) | | LOI for S'' ^c (mass %) | |
|-------------------|------------------------------------|---------|-------------------------------------|--------|--------------------------------------|--------|
| | Powder | Chunks | Powder | Chunks | Powder | Chunks |
| 1000°C for 2 h | 7.1806 | 11.8452 | 8.3312 | 9.6495 | 7.4777 | 9.9484 |

^aImpure oxide as-received.

^bImpure oxide, S, was calcined at 600°C for 12 h to yield S'.

^cImpure oxide, S', was calcined at 950°C for 2 h to yield S''.

Table V shows the results for the elements analyzed. Carbon concentration decreased after calcination. Chloride concentration decreased slightly after the 600°C calcining step and substantially after the 950°C calcining step. The low chloride concentration for the sample calcined at 950°C is consistent with the LOI values as a function of LOI temperature conditions. Plutonium concentration slightly decreased after the 600°C calcining step and substantially increased after the 950°C calcination. Obviously, this large increase in plutonium concentration is due to the loss of chloride salts. The net amount of plutonium is understood to remain constant.

Table V. Elemental Analysis of As-Received Powder S^a and Calcined Powders S'^b and S''^c

| Element | Powder S (µg/g) | Powder S' (µg/g) | Powder S'' (µg/g) |
|------------------|--------------------|---------------------|----------------------|
| Plutonium | 265900 | 263500 | 394800 |
| Chloride | <204000 | 181000 | 77000 |
| Carbon | 3600 | 70 | 200 |

^aImpure oxide as-received.

^bImpure oxide, S, was calcined at 600°C for 12 h to yield S'.

^cImpure oxide, S', was calcined at 950°C for 2 h to yield S''.

The specific surface area of ARF-102-85-295 material was not measured.

Particle analysis results for the material in powder form are shown in Table VI.

Table VI. Particle Analysis Results of Precalcined Powder S^a and Calcined Powders S^b and S^c

| Property | Powder S | Powder S' | Powder S'' |
|---------------------------------------|----------|-----------|------------|
| Spherical Equivalent Mean (μm) | 17.05 | 18.65 | 18.5 |
| Diameter-by-Volume Mean (μm) | 54.75 | 66.3 | 67.65 |

^aImpure oxide as-received.

^bImpure oxide, S, was calcined at 600°C for 12 h to yield S'.

^cImpure oxide, S', was calcined at 950°C for 2 h to yield S''.

The tap and bulk density of the as-received oxide was not measured; however they were measured for the materials in powder form calcined at 600°C and 950°C. The values are shown in Table VII.

Table VII. Tap Density and Bulk Density of Calcined Powders S^a and S^b

| Property | Powder S' | Powder S'' |
|----------------------------|-----------|------------|
| Tap Density (g/cc) | 4.65 | 4.16 |
| Bulk Density (g/cc) | 5.26 | 4.76 |

^aImpure oxide, S, was calcined at 600°C for 12 h to yield S'.

^bImpure oxide, S', was calcined at 950°C for 2 h to yield S''.

The results for the ARF samples of the XRD runs are summarized in Tables 1 through 6 in Appendix 1; in addition the powder patterns for each sample and the phase identification are shown in Fig. 1 through 6, Appendix 1. These results indicate that the samples are composed of PuO_{2-x}, NaCl, KCl, and chromium oxide whose stoichiometry is not known. The exact composition of each sample could not be calculated because the composition and density of the chromium oxide could not be determined. Tables 1 through 6 contain a listing for each sample of the values for two theta, d, Miller indices, background, peak areas which reflect the intensities of the peaks, and the peak widths. At the end of each listing for a given sample, the table reports the calculated lattice constant for that sample. The lattice constant was calculated by a least squares technique in which the points were weighted by the square root of sine theta. The oxygen to plutonium ratio for

each sample was calculated from the lattice constant and the data from Gardner et al.⁸ The results of the TGA runs are summarized in Table 7, Appendix 1.

Interpretation of the x-ray diffraction results is complicated by the chemistry of the KCl-NaCl system. KCl and NaCl form a series of solid solutions with a miscibility gap below 500°C; upon melting KCl and NaCl are completely miscible⁹. Therefore the intensities of the KCl and NaCl lines (their amounts) in the heat treated samples depends on the temperature and the cooling rate. This statement is also true of the as received material if that material had been heat treated sometime in its history. Changes in the relative peak intensities of the phases in the ARF samples indicate that the thermal processing removes KCl and NaCl while the concentration of the phase identified as chromium oxide remains essentially unchanged. At 900°C more KCl than NaCl is removed because the vapor pressure of KCl is greater than that of NaCl¹⁰. The calculated lattice constants of the plutonium oxide also show that thermal processing increases the O/Pu ratio for ARF-3, 5, and 9. Sample ARF-3A, the milled powder, contained “stoichiometric” PuO₂.

The identification of chromium oxide seems questionable. Of all the chromium oxides and other known in the international center for diffraction data (ICDD), the chromium oxide selected fits the best. This oxide has Cr primarily in the +2 oxidation state although some +3 may exist, this is the reason that the composition is unknown. It seems that if CrO is the phase, the heat treatment in air would oxidize the CrO to some extent. The results show that the CrO does not change with heat treatment. This result should be checked against the elemental analysis for the starting materials. Taken together, the TGA/DTA results show that the ARF samples undergo five thermal events; although not all samples undergo all five as summarized in Table TGA/DTA-1. The thermal events between 25 and 200°C could be interpreted as the removal of water in a three stage process. The percent mass loss in the range of 25 to 500°C strongly suggests that samples ARF-9 and 13 and possibly ARF-3 and 3a picked up water during shipment for TGA analysis. The cause of the thermal event at roughly 450°C in ARF-9 and 13 is unknown. The thermal event at approximately 650°C, common to all samples, is interpreted as the melting of the KCl-NaCl. The large mass losses subsequently recorded are due to the vaporization of the KCl and NaCl. The residues that remained after thermal treatment at 900°C could only be removed from the crucible by scrapping. The melting of the salt at 650°C will impact the processing of this material in two ways, (1) the as processed material will not be a free flowing powder, and (2) large quantities of salt will be deposited in the processing equipment.

The gas analysis results for the outer and inner can are shown in Table VIII. Since the outer can was open to the atmosphere it is understandable that the gas analysis is very close to that of the air. The differences are attributed to error in the RGA calibration.

Table VIII. Can Puncturing Results and the Gas Composition for the Outer and Inner Can Gas Phase.

| Property | Outer Can | Inner Can | Air |
|-----------------------------|-------------|--------------|-----------------------|
| Temperature (°C) | 19.9 | 21.5 | |
| Equilibrium Pressure (Torr) | 578.9 | 475 (584.24) | |
| Calculated Pressure (Torr) | NA | 456.2 | |
| Can Seal | leaking | leaking | |
| Gas Species (mole %) | | | |
| N ₂ | 80.5 | 97.1 | 78.08 |
| O ₂ | 18.6 | 0.0042 | 20.94 |
| Ar | 0.75 | 0.91 | 0.934 |
| CO ₂ | 0.043 | 1.27 | 0.0314 |
| He | 0.0022 | 0.44 | 5.24x10 ⁻⁴ |
| H ₂ | 0.0058 | 0.0054 | 5.0x10 ⁻⁵ |
| H ₂ O | 0.05 | 0.02 | not listed |
| CH ₄ | not present | 0.052 | not listed |
| 45 | not present | 0.14 | not listed |

The 1996 isotopic distribution of plutonium is shown in Table IX.

Table IX. Isotopic Distribution of Plutonium for the “As-received” Powder S.

| Isotope | Hanford Analysis | Los Alamos Analysis |
|---------|------------------|---------------------|
| Pu-238 | 0.0077 | 0.0076 |
| Pu-239 | 94.1925 | 94.1808 |
| Pu-240 | 5.673 | 5.6858 |
| Pu-241 | 0.107 | 0.1069 |
| Pu-242 | 0.0199 | 0.0189 |
| Total | 100 | 100 |

4.0 CONCLUSION

This particular impure plutonium oxide item ARF-102-85-295 has 26.59 mass % plutonium. The very high LOI value is a result of the very high concentration of chloride. The material does not satisfy DOE-STD-3013-96 criteria for plutonium content and LOI.

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Appendix 1. XRD and TGA results.

Table 1. X-ray Diffraction Results for Sample ARF-3

| 2-Theta | d(A) | Int | I% | FWHM | Phase-ID | h | k | l |
|---------|--------|-----|-------|-------|-----------------|---|---|---|
| 27.322 | 3.2614 | 5 | 4.9 | 0.214 | Halite, syn | 1 | 1 | 1 |
| 28.353 | 3.1451 | 102 | 100.0 | 0.315 | Sylvite, syn | 2 | 0 | 0 |
| 30.087 | 2.9677 | 6 | 5.9 | 0.360 | Cr-O | 2 | 2 | 0 |
| 31.611 | 2.8280 | 37 | 36.3 | 0.206 | Halite, syn | 2 | 0 | 0 |
| 33.139 | 2.7010 | 25 | 24.5 | 0.417 | PuO2 | 0 | 0 | 2 |
| 35.507 | 2.5262 | 13 | 12.7 | 0.308 | Cr-O | 3 | 1 | 1 |
| 40.498 | 2.2256 | 30 | 29.4 | 0.287 | Sylvite, syn | 2 | 2 | 0 |
| 43.134 | 2.0955 | 5 | 4.9 | 0.150 | Cr-O | 4 | 0 | 0 |
| 45.400 | 1.9960 | 15 | 14.7 | 0.241 | Halite, syn | 2 | 2 | 0 |
| 47.468 | 1.9138 | 20 | 19.6 | 0.327 | PuO2 | 0 | 2 | 2 |
| 50.127 | 1.8183 | 5 | 4.9 | 0.100 | Sylvite, syn | 2 | 2 | 2 |
| 52.704 | 1.7353 | 2 | 2.0 | 0.067 | | | | |
| 56.331 | 1.6319 | 28 | 27.5 | 0.406 | Halite, syn | 2 | 2 | 2 |
| 57.040 | 1.6133 | 1 | 1.0 | 0.100 | Cr-O | 5 | 1 | 1 |
| 58.341 | 1.5804 | 5 | 4.9 | 0.110 | | | | |
| 58.679 | 1.5721 | 10 | 9.8 | 0.818 | Sylvite, syn | 4 | 0 | 0 |
| 59.041 | 1.5633 | 8 | 7.8 | 0.514 | PuO2 | 2 | 2 | 2 |
| 62.681 | 1.4810 | 7 | 6.9 | 0.262 | Cr-O | 4 | 4 | 0 |
| 66.320 | 1.4083 | 9 | 8.8 | 0.338 | Sylvite, syn | 4 | 2 | 0 |
| 73.783 | 1.2832 | 2 | 2.0 | 0.164 | Sylvite, syn | 4 | 2 | 2 |
| 75.263 | 1.2616 | 6 | 5.9 | 0.300 | Halite, syn | 4 | 2 | 0 |
| 76.833 | 1.2396 | 10 | 9.8 | 0.818 | PuO2 | 1 | 3 | 3 |
| 78.217 | 1.2211 | 3 | 2.9 | 0.084 | | | | |
| 79.133 | 1.2093 | 5 | 4.9 | 0.409 | PuO2 | 0 | 2 | 4 |
| 83.584 | 1.1558 | 2 | 2.0 | 0.078 | Halite, syn | 4 | 2 | 2 |
| 88.693 | 1.1020 | 5 | 4.9 | 0.090 | PuO2 | 2 | 2 | 4 |
| 89.210 | 1.0969 | 5 | 4.9 | 0.250 | | | | |
| 89.604 | 1.0931 | 1 | 1.0 | 0.045 | | | | |
| 95.314 | 1.0422 | 9 | 8.8 | 0.506 | PuO2 | 1 | 1 | 5 |
| 102.891 | 0.9850 | 2 | 2.0 | 0.069 | | | | |
| 108.604 | 0.9485 | 2 | 2.0 | 0.067 | Sylvite, syn | 6 | 2 | 2 |
| 110.358 | 0.9383 | 6 | 5.9 | 0.089 | Halite, syn | 6 | 0 | 0 |
| 115.328 | 0.9117 | 3 | 2.9 | 0.104 | PuO2 | 1 | 3 | 5 |
| 118.774 | 0.8950 | 1 | 1.0 | 0.082 | | | | |

| | | | | | | | | |
|---------|--------|---|-----|-------|------|---|---|---|
| 122.789 | 0.8774 | 3 | 2.9 | 0.077 | | | | |
| 129.102 | 0.8531 | 3 | 2.9 | 0.245 | PuO2 | 0 | 2 | 6 |

Calculated lattice constant for PuO2: 5.4081(3)
 Calculated O/Pu: 1.967

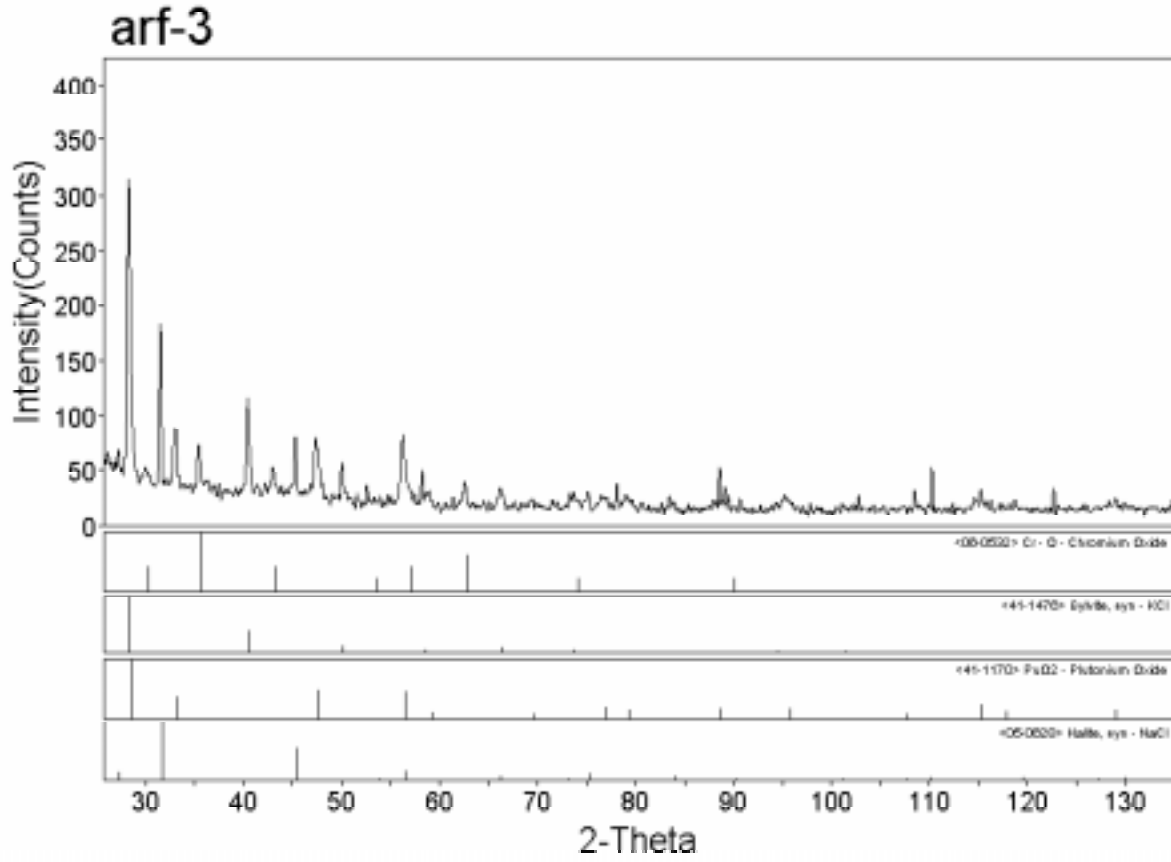


Table 2. X-ray Diffraction Results for Sample ARF-3A

| 2-Theta | d(A) | Int | I% | FWHM | Phase-ID | h | k | l |
|---------|--------|-----|-------|-------|-----------------|---|---|---|
| 28.580 | 3.1207 | 107 | 100.0 | 0.267 | PuO2 | 1 | 1 | 1 |
| 30.154 | 2.9613 | 5 | 4.7 | 0.250 | Cr-O | 2 | 2 | 0 |
| 31.600 | 2.8290 | 6 | 5.6 | 0.235 | Halite, syn | 2 | 0 | 0 |
| 33.143 | 2.7007 | 46 | 43.0 | 0.238 | PuO2 | 0 | 0 | 2 |
| 35.644 | 2.5168 | 17 | 15.9 | 0.243 | Cr-O | 3 | 1 | 1 |
| 40.512 | 2.2249 | 7 | 6.5 | 0.332 | Sylvite, syn | 2 | 2 | 0 |
| 43.241 | 2.0906 | 15 | 14.0 | 0.587 | Cr-O | 4 | 0 | 0 |
| 45.341 | 1.9985 | 5 | 4.7 | 0.265 | Halite, syn | 2 | 2 | 0 |
| 47.602 | 1.9087 | 71 | 66.4 | 0.349 | PuO2 | 0 | 2 | 2 |
| 50.097 | 1.8193 | 3 | 2.8 | 0.208 | Sylvite, syn | 2 | 2 | 2 |
| 53.741 | 1.7042 | 7 | 6.5 | 0.420 | Cr-O | 4 | 2 | 2 |
| 56.483 | 1.6278 | 64 | 59.8 | 0.360 | Halite, syn | 2 | 2 | 2 |
| 57.359 | 1.6051 | 12 | 11.2 | 0.491 | Cr-O | 5 | 1 | 1 |
| 59.190 | 1.5597 | 11 | 10.3 | 0.291 | PuO2 | 2 | 2 | 2 |
| 62.885 | 1.4766 | 16 | 15.0 | 0.514 | Cr-O | 4 | 4 | 0 |
| 63.198 | 1.4701 | 9 | 8.4 | 0.368 | | | | |
| 65.071 | 1.4322 | 5 | 4.7 | 0.196 | | | | |
| 66.412 | 1.4065 | 3 | 2.8 | 0.169 | Sylvite, syn | 4 | 2 | 0 |
| 69.590 | 1.3499 | 10 | 9.3 | 0.333 | PuO2 | 0 | 0 | 4 |
| 69.816 | 1.3460 | 5 | 4.7 | 0.225 | | | | |
| 76.962 | 1.2379 | 33 | 30.8 | 0.594 | PuO2 | 1 | 3 | 3 |
| 79.325 | 1.2068 | 22 | 20.6 | 0.495 | PuO2 | 0 | 2 | 4 |
| 88.707 | 1.1018 | 21 | 19.6 | 0.497 | PuO2 | 2 | 2 | 4 |
| 95.746 | 1.0386 | 14 | 13.1 | 0.371 | PuO2 | 1 | 1 | 5 |
| 107.530 | 0.9550 | 5 | 4.7 | 0.173 | PuO2 | 0 | 4 | 4 |
| 108.370 | 0.9499 | 5 | 4.7 | 0.145 | Sylvite, syn | 6 | 2 | 2 |
| 115.358 | 0.9115 | 25 | 23.4 | 0.776 | PuO2 | 1 | 3 | 5 |
| 117.185 | 0.9025 | 4 | 3.7 | 0.171 | | | | |
| 119.169 | 0.8932 | 2 | 1.9 | 0.100 | | | | |
| 125.030 | 0.8683 | 4 | 3.7 | 0.157 | | | | |
| 129.000 | 0.8534 | 19 | 17.8 | 0.658 | PuO2 | 0 | 2 | 6 |

Calculated lattice constant for PuO2: 5.3974(4)

Calculated O/Pu: 1.996

arf-3a

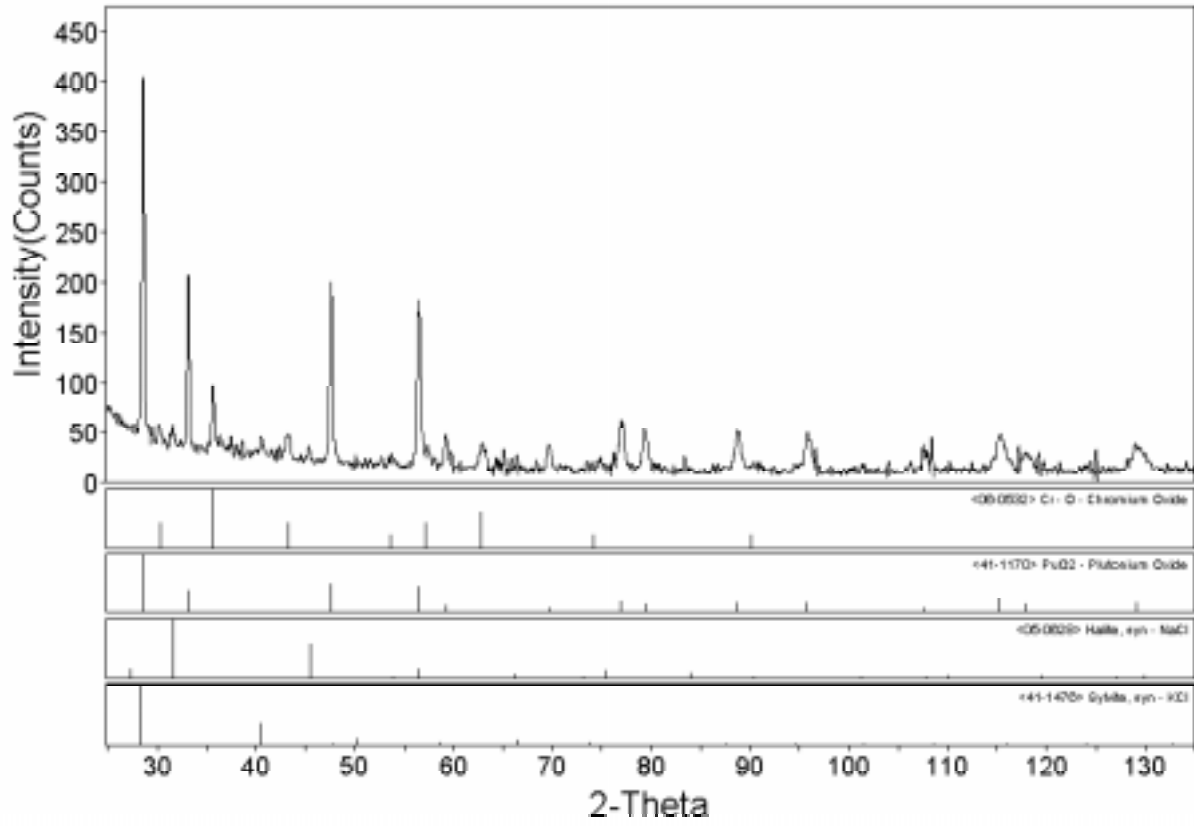


Table 3. X-ray Diffraction Results for Sample ARF5

| 2-Theta | d(A) | Int | I% | FWHM | Phase-ID | h | k | l |
|---------|--------|-----|-------|-------|-----------------|---|---|---|
| 27.279 | 3.2665 | 3 | 2.8 | 0.208 | Halite, syn | 1 | 1 | 1 |
| 28.289 | 3.1522 | 92 | 86.0 | 0.427 | Sylvite, syn | 2 | 0 | 0 |
| 28.452 | 3.1345 | 107 | 100.0 | 0.544 | Halite, syn | 2 | 0 | 0 |
| 31.525 | 2.8355 | 25 | 23.4 | 0.179 | Halite, syn | 2 | 0 | 0 |
| 33.067 | 2.7068 | 23 | 21.5 | 0.287 | PuO2 | 0 | 0 | 2 |
| 35.638 | 2.5172 | 4 | 3.7 | 0.327 | Cr-O | 3 | 1 | 1 |
| 40.441 | 2.2286 | 22 | 20.6 | 0.257 | Sylvite, syn | 2 | 2 | 0 |
| 45.249 | 2.0023 | 10 | 9.3 | 0.180 | Halite, syn | 2 | 2 | 0 |
| 47.541 | 1.9110 | 27 | 25.2 | 0.426 | PuO2 | 0 | 2 | 2 |
| 50.044 | 1.8211 | 6 | 5.6 | 0.164 | | | | |
| 50.280 | 1.8131 | 6 | 5.6 | 0.169 | Sylvite, syn | 2 | 2 | 2 |
| 56.363 | 1.6310 | 33 | 30.8 | 0.530 | Halite, syn | 2 | 2 | 2 |
| 56.797 | 1.6196 | 11 | 10.3 | 0.330 | PuO2 | 1 | 1 | 3 |
| 57.180 | 1.6097 | 1 | 0.9 | 0.100 | Cr-O | 5 | 1 | 1 |
| 58.558 | 1.5750 | 2 | 1.9 | 0.164 | Sylvite, syn | 4 | 0 | 0 |
| 59.151 | 1.5606 | 2 | 1.9 | 0.129 | PuO2 | 2 | 2 | 2 |
| 66.330 | 1.4081 | 10 | 9.3 | 0.500 | Sylvite, syn | 4 | 2 | 0 |
| 73.807 | 1.2828 | 3 | 2.8 | 0.193 | Sylvite, syn | 4 | 2 | 2 |
| 74.270 | 1.2759 | 1 | 0.9 | 0.090 | Cr-O | 5 | 3 | 3 |
| 74.967 | 1.2658 | 3 | 2.8 | 0.129 | | | | |
| 76.683 | 1.2417 | 3 | 2.8 | 0.129 | | | | |
| 76.984 | 1.2376 | 4 | 3.7 | 0.144 | PuO2 | 1 | 3 | 3 |
| 77.957 | 1.2246 | 4 | 3.7 | 0.129 | | | | |
| 80.938 | 1.1868 | 3 | 2.8 | 0.123 | | | | |
| 88.705 | 1.1019 | 3 | 2.8 | 0.135 | PuO2 | 2 | 2 | 4 |
| 101.764 | 0.9928 | 3 | 2.8 | 0.123 | Sylvite, syn | 6 | 2 | 0 |
| 104.904 | 0.9715 | 4 | 3.7 | 0.133 | | | | |
| 111.646 | 0.9311 | 4 | 3.7 | 0.120 | | | | |
| 115.076 | 0.9129 | 3 | 2.8 | 0.123 | PuO2 | 1 | 3 | 5 |
| 119.189 | 0.8931 | 6 | 5.6 | 0.164 | | | | |
| 133.401 | 0.8387 | 5 | 4.7 | 0.132 | | | | |

Calculated lattice constant for PuO2: 5.4016(3)

Calculated O/Pu: 1.985

arf-5

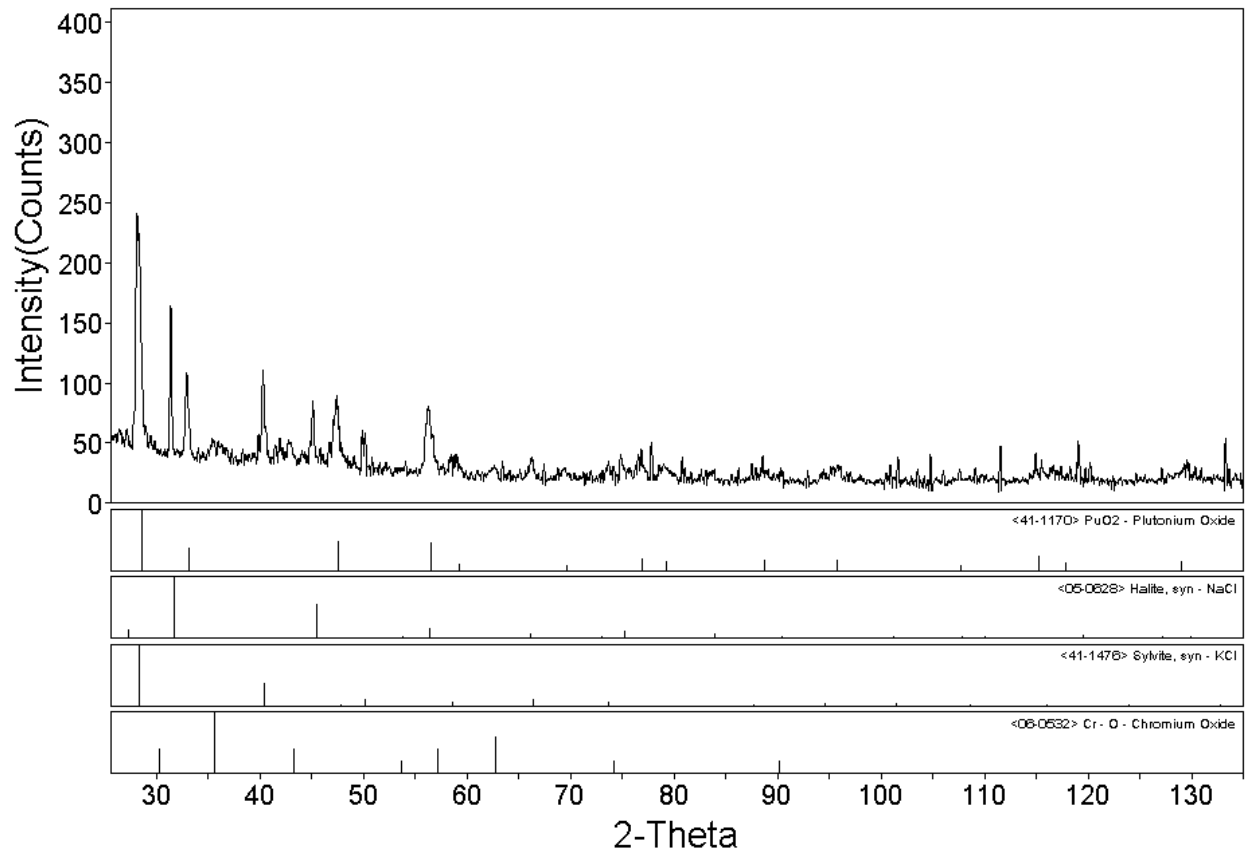


Table 4. X-ray Diffraction Results for Sample ARF8

| 2-Theta | d(A) | Int | I% | FWHM | Phase-ID | h | k | l |
|---------|--------|-----|-------|-------|-----------------|---|---|---|
| 27.322 | 3.2614 | 3 | 2.5 | 0.093 | Halite, syn | 1 | 1 | 1 |
| 28.300 | 3.1509 | 95 | 78.5 | 0.441 | Sylvite, syn | 2 | 0 | 0 |
| 28.495 | 3.1298 | 121 | 100.0 | 0.500 | PuO2 | 1 | 1 | 1 |
| 31.560 | 2.8325 | 28 | 23.1 | 0.176 | Halite, syn | 2 | 0 | 0 |
| 33.079 | 2.7058 | 29 | 24.0 | 0.284 | PuO2 | 0 | 0 | 2 |
| 35.433 | 2.5313 | 6 | 5.0 | 0.180 | Cr-O | 3 | 1 | 1 |
| 40.467 | 2.2272 | 20 | 16.5 | 0.222 | Sylvite, syn | 2 | 2 | 0 |
| 40.617 | 2.2194 | 16 | 13.2 | 0.257 | | | | |
| 42.852 | 2.1086 | 4 | 3.3 | 0.150 | | | | |
| 43.081 | 2.0980 | 11 | 9.1 | 0.396 | Cr-O | 4 | 0 | 0 |
| 45.270 | 2.0014 | 18 | 14.9 | 0.198 | Halite, syn | 2 | 2 | 0 |
| 47.578 | 1.9096 | 52 | 43.0 | 0.532 | PuO2 | 0 | 2 | 2 |
| 50.128 | 1.8183 | 5 | 4.1 | 0.145 | Sylvite, syn | 2 | 2 | 2 |
| 56.399 | 1.6301 | 39 | 32.2 | 0.444 | Halite, syn | 2 | 2 | 2 |
| 56.687 | 1.6225 | 23 | 19.0 | 0.493 | PuO2 | 1 | 1 | 3 |
| 59.064 | 1.5627 | 5 | 4.1 | 0.225 | PuO2 | 2 | 2 | 2 |
| 62.719 | 1.4802 | 9 | 7.4 | 0.405 | Cr-O | 4 | 4 | 0 |
| 66.290 | 1.4088 | 4 | 3.3 | 0.133 | Halite, syn | 4 | 0 | 0 |
| 74.905 | 1.2667 | 6 | 5.0 | 0.216 | | | | |
| 76.885 | 1.2389 | 12 | 9.9 | 0.450 | PuO2 | 1 | 3 | 3 |
| 78.739 | 1.2143 | 3 | 2.5 | 0.108 | | | | |
| 81.030 | 1.1857 | 3 | 2.5 | 0.100 | | | | |
| 86.401 | 1.1252 | 2 | 1.7 | 0.100 | | | | |
| 88.663 | 1.1023 | 4 | 3.3 | 0.300 | PuO2 | 2 | 2 | 4 |
| 90.414 | 1.0854 | 2 | 1.7 | 0.095 | Halite, syn | 5 | 1 | 1 |
| 95.845 | 1.0378 | 11 | 9.1 | 0.900 | PuO2 | 1 | 1 | 5 |
| 108.919 | 0.9466 | 3 | 2.5 | 0.123 | | | | |
| 112.239 | 0.9278 | 2 | 1.7 | 0.086 | | | | |
| 115.064 | 0.9130 | 4 | 3.3 | 0.150 | PuO2 | 1 | 3 | 5 |
| 121.453 | 0.8830 | 5 | 4.1 | 0.132 | | | | |
| 125.409 | 0.8668 | 2 | 1.7 | 0.082 | | | | |
| 129.147 | 0.8529 | 3 | 2.5 | 0.123 | PuO2 | 0 | 2 | 6 |

Calculated lattice constant for PuO2: 5.3985(4)

Calculated O/Pu: 1.993

arf-8

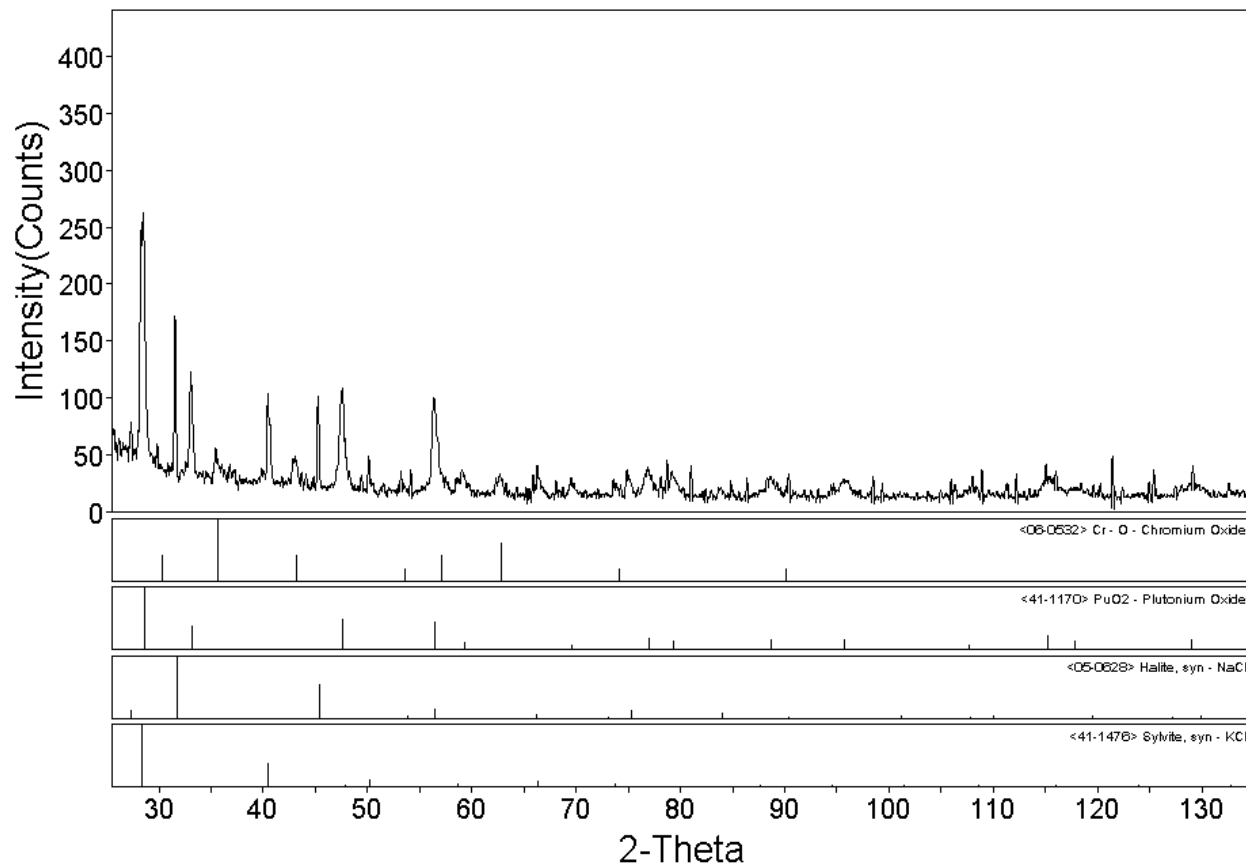


Table 5. X-ray Diffraction Results for Sample ARF9

| 2-Theta | d(A) | Int | I% | FWHM | Phase-ID | h | k | l |
|---------|--------|-----|-------|-------|-----------------|---|---|---|
| 28.541 | 3.1249 | 103 | 100.0 | 0.255 | PuO2 | 1 | 1 | 1 |
| 29.888 | 2.9870 | 4 | 3.9 | 0.129 | | | | |
| 30.163 | 2.9604 | 5 | 4.9 | 0.150 | Cr-O | 2 | 2 | 0 |
| 31.524 | 2.8357 | 8 | 7.8 | 0.157 | Halite, syn | 2 | 0 | 0 |
| 33.116 | 2.7028 | 41 | 39.8 | 0.225 | PuO2 | 0 | 0 | 2 |
| 35.545 | 2.5236 | 23 | 22.3 | 0.252 | Cr-O | 3 | 1 | 1 |
| 40.534 | 2.2237 | 7 | 6.8 | 0.286 | Sylvite, syn | 2 | 2 | 0 |
| 43.205 | 2.0922 | 13 | 12.6 | 0.292 | Cr-O | 4 | 0 | 0 |
| 47.561 | 1.9103 | 51 | 49.5 | 0.319 | PuO2 | 0 | 2 | 2 |
| 56.478 | 1.6280 | 51 | 49.5 | 0.335 | Halite, syn | 2 | 2 | 2 |
| 57.228 | 1.6084 | 22 | 21.4 | 0.600 | Cr-O | 5 | 1 | 1 |
| 58.687 | 1.5718 | 3 | 2.9 | 0.129 | Sylvite, syn | 4 | 0 | 0 |
| 59.188 | 1.5597 | 12 | 11.7 | 0.318 | PuO2 | 2 | 2 | 2 |
| 62.441 | 1.4861 | 3 | 2.9 | 0.135 | | | | |
| 62.937 | 1.4755 | 13 | 12.6 | 0.355 | Cr-O | 4 | 4 | 0 |
| 69.556 | 1.3504 | 9 | 8.7 | 0.261 | PuO2 | 0 | 0 | 4 |
| 76.921 | 1.2384 | 24 | 23.3 | 0.502 | PuO2 | 1 | 3 | 3 |
| 77.131 | 1.2356 | 11 | 10.7 | 0.291 | | | | |
| 78.118 | 1.2224 | 2 | 1.9 | 0.095 | | | | |
| 79.376 | 1.2062 | 16 | 15.5 | 0.424 | PuO2 | 0 | 2 | 4 |
| 85.156 | 1.1385 | 6 | 5.8 | 0.132 | | | | |
| 88.650 | 1.1024 | 14 | 13.6 | 0.371 | PuO2 | 2 | 2 | 4 |
| 95.873 | 1.0375 | 17 | 16.5 | 0.546 | PuO2 | 1 | 1 | 5 |
| 96.147 | 1.0353 | 6 | 5.8 | 0.257 | | | | |
| 99.784 | 1.0071 | 2 | 1.9 | 0.090 | | | | |
| 103.603 | 0.9802 | 2 | 1.9 | 0.095 | | | | |
| 105.016 | 0.9708 | 4 | 3.9 | 0.129 | | | | |
| 107.186 | 0.9571 | 2 | 1.9 | 0.090 | | | | |
| 115.151 | 0.9125 | 10 | 9.7 | 0.429 | PuO2 | 1 | 3 | 5 |
| 115.475 | 0.9109 | 13 | 12.6 | 0.390 | | | | |
| 118.121 | 0.8981 | 4 | 3.9 | 0.157 | PuO2 | 2 | 4 | 4 |
| 121.106 | 0.8846 | 3 | 2.9 | 0.129 | | | | |
| 126.485 | 0.8627 | 3 | 2.9 | 0.104 | | | | |
| 129.746 | 0.8508 | 3 | 2.9 | 0.108 | Halite, syn | 6 | 2 | 2 |

Calculated lattice constant for PuO2: 5.3969(4)

Calculated O/Pu: 1.997

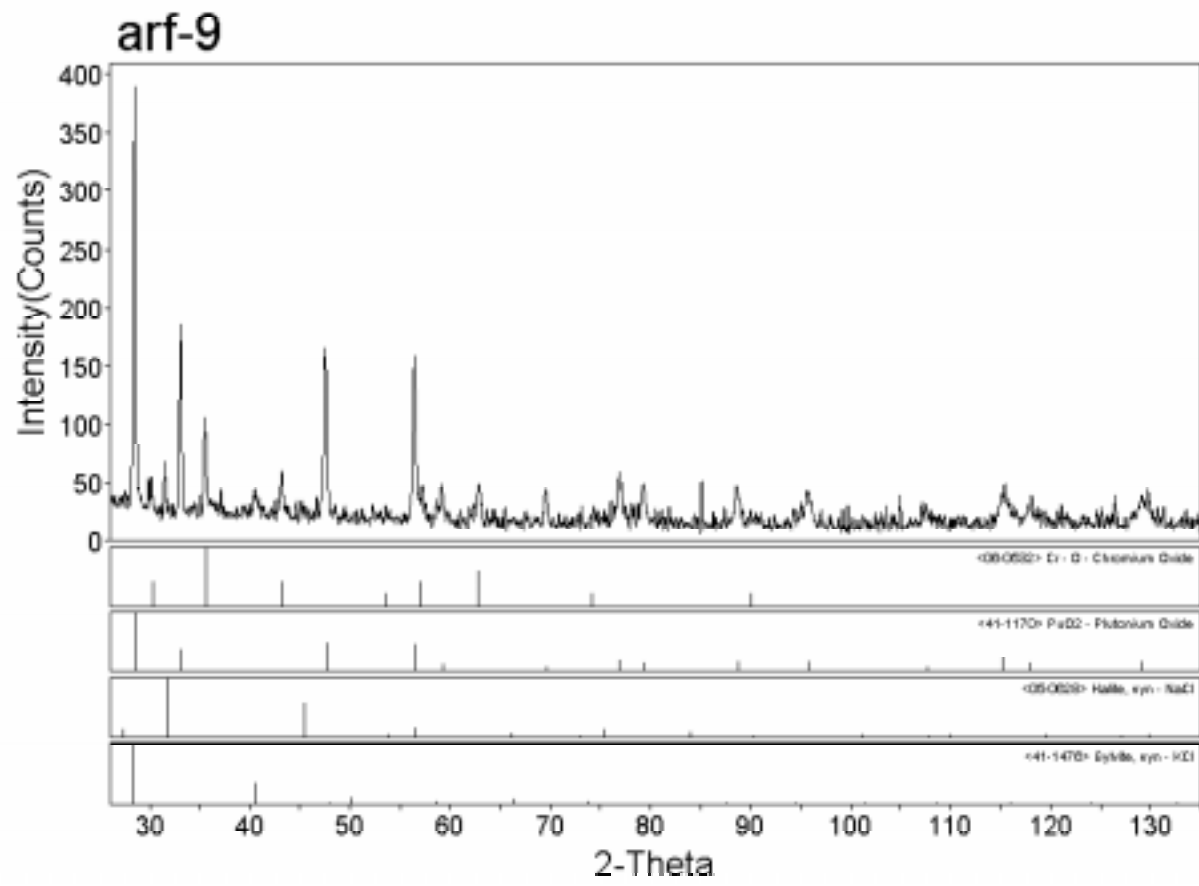


Table 6. X-ray Diffraction Results for Sample ARF13

| 2-Theta | d(A) | Int | I% | FWHM | Phase-ID | h | k | l |
|---------|--------|-----|-------|-------|-----------------|---|---|---|
| 28.577 | 3.1210 | 106 | 100.0 | 0.266 | PuO2 | 1 | 1 | 1 |
| 30.197 | 2.9571 | 7 | 6.6 | 0.315 | Cr-O | 2 | 2 | 0 |
| 31.579 | 2.8309 | 7 | 6.6 | 0.286 | Halite, syn | 2 | 0 | 0 |
| 33.145 | 2.7006 | 46 | 43.4 | 0.239 | PuO2 | 0 | 0 | 2 |
| 35.646 | 2.5166 | 17 | 16.0 | 0.247 | Cr-O | 3 | 1 | 1 |
| 40.528 | 2.2240 | 6 | 5.7 | 0.284 | Sylvite, syn | 2 | 2 | 0 |
| 43.228 | 2.0912 | 15 | 14.2 | 0.614 | Cr-O | 4 | 0 | 0 |
| 45.336 | 1.9987 | 7 | 6.6 | 0.371 | Halite, syn | 2 | 2 | 0 |
| 47.604 | 1.9086 | 70 | 66.0 | 0.346 | PuO2 | 0 | 2 | 2 |
| 50.087 | 1.8197 | 2 | 1.9 | 0.164 | Sylvite, syn | 2 | 2 | 2 |
| 53.722 | 1.7048 | 8 | 7.5 | 0.514 | Cr-O | 4 | 2 | 2 |
| 56.498 | 1.6274 | 64 | 60.4 | 0.362 | PuO2 | 1 | 1 | 3 |
| 57.357 | 1.6051 | 10 | 9.4 | 0.409 | Cr-O | 5 | 1 | 1 |
| 59.212 | 1.5592 | 12 | 11.3 | 0.318 | PuO2 | 2 | 2 | 2 |
| 62.889 | 1.4766 | 17 | 16.0 | 0.567 | Cr-O | 4 | 4 | 0 |
| 63.188 | 1.4703 | 8 | 7.5 | 0.343 | | | | |
| 65.074 | 1.4322 | 5 | 4.7 | 0.196 | | | | |
| 66.419 | 1.4064 | 3 | 2.8 | 0.142 | Sylvite, syn | 4 | 2 | 0 |
| 69.590 | 1.3499 | 10 | 9.4 | 0.333 | PuO2 | 0 | 0 | 4 |
| 76.971 | 1.2378 | 34 | 32.1 | 0.612 | PuO2 | 1 | 3 | 3 |
| 79.379 | 1.2062 | 23 | 21.7 | 0.517 | PuO2 | 0 | 2 | 4 |
| 83.380 | 1.1581 | 3 | 2.8 | 0.180 | PuO2 | 2 | 2 | 4 |
| 88.733 | 1.1016 | 24 | 22.6 | 0.554 | PuO2 | 2 | 2 | 4 |
| 95.754 | 1.0385 | 14 | 13.2 | 0.382 | PuO2 | 1 | 1 | 5 |
| 107.533 | 0.9550 | 6 | 5.7 | 0.216 | PuO2 | 0 | 4 | 4 |
| 108.378 | 0.9499 | 5 | 4.7 | 0.150 | Sylvite, syn | 6 | 2 | 2 |
| 115.374 | 0.9114 | 22 | 20.8 | 0.683 | PuO2 | 1 | 3 | 5 |
| 117.182 | 0.9025 | 3 | 2.8 | 0.129 | | | | |
| 119.187 | 0.8931 | 3 | 2.8 | 0.142 | | | | |
| 125.026 | 0.8683 | 4 | 3.8 | 0.157 | | | | |
| 128.973 | 0.8535 | 19 | 17.9 | 0.684 | PuO2 | 0 | 2 | 6 |

Calculated lattice constant for PuO2: 5.3966(3)

Calculated O/Pu: 1.998

arf-13

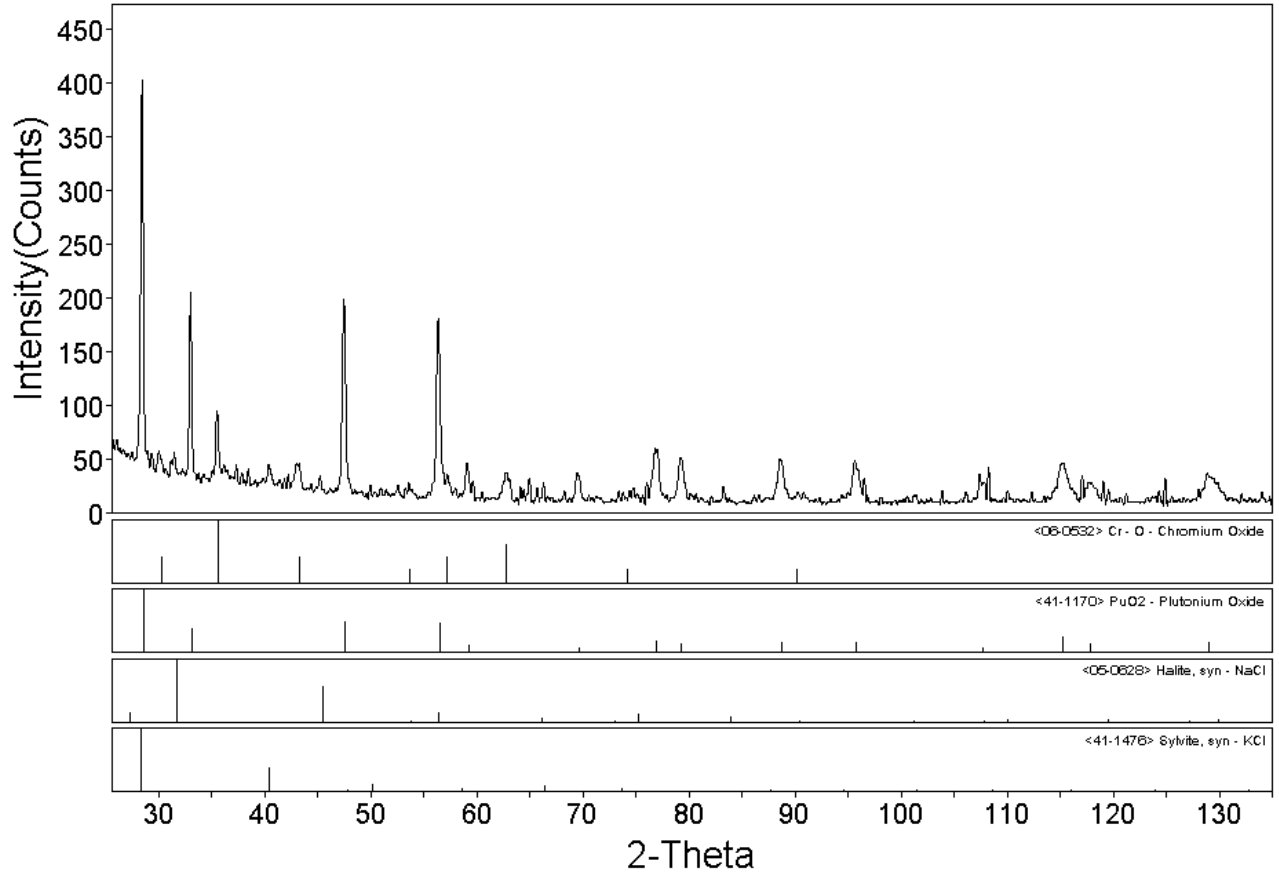
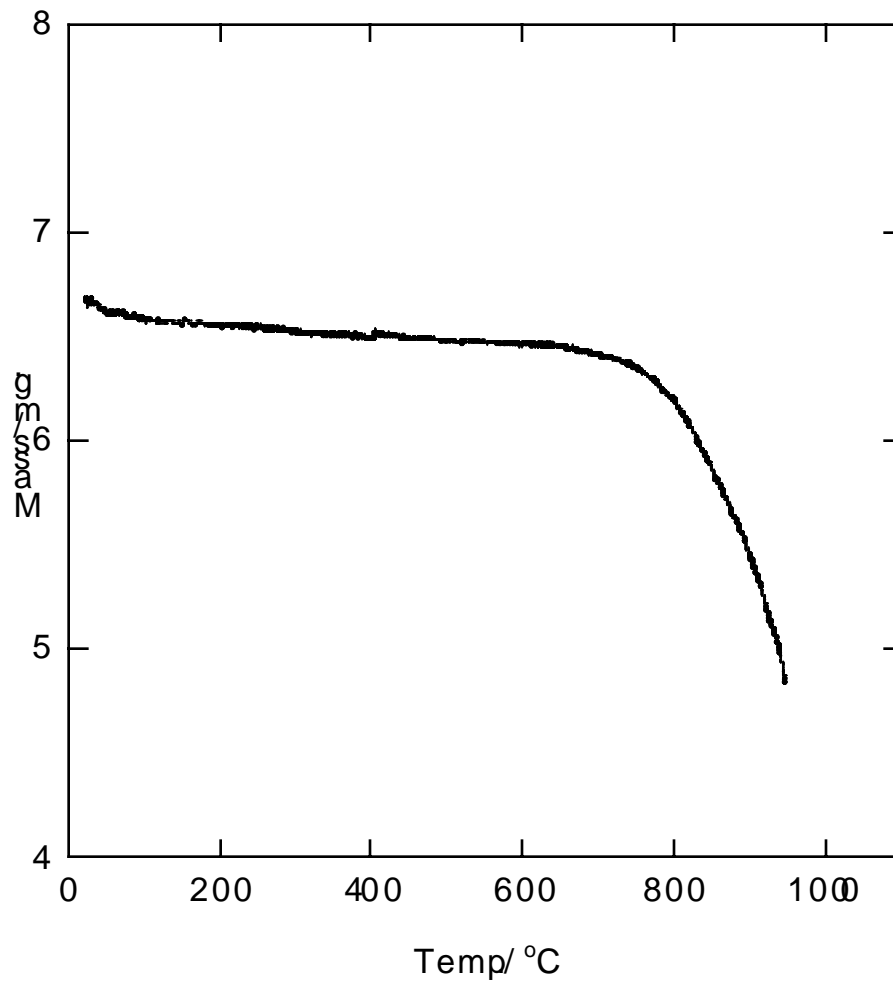


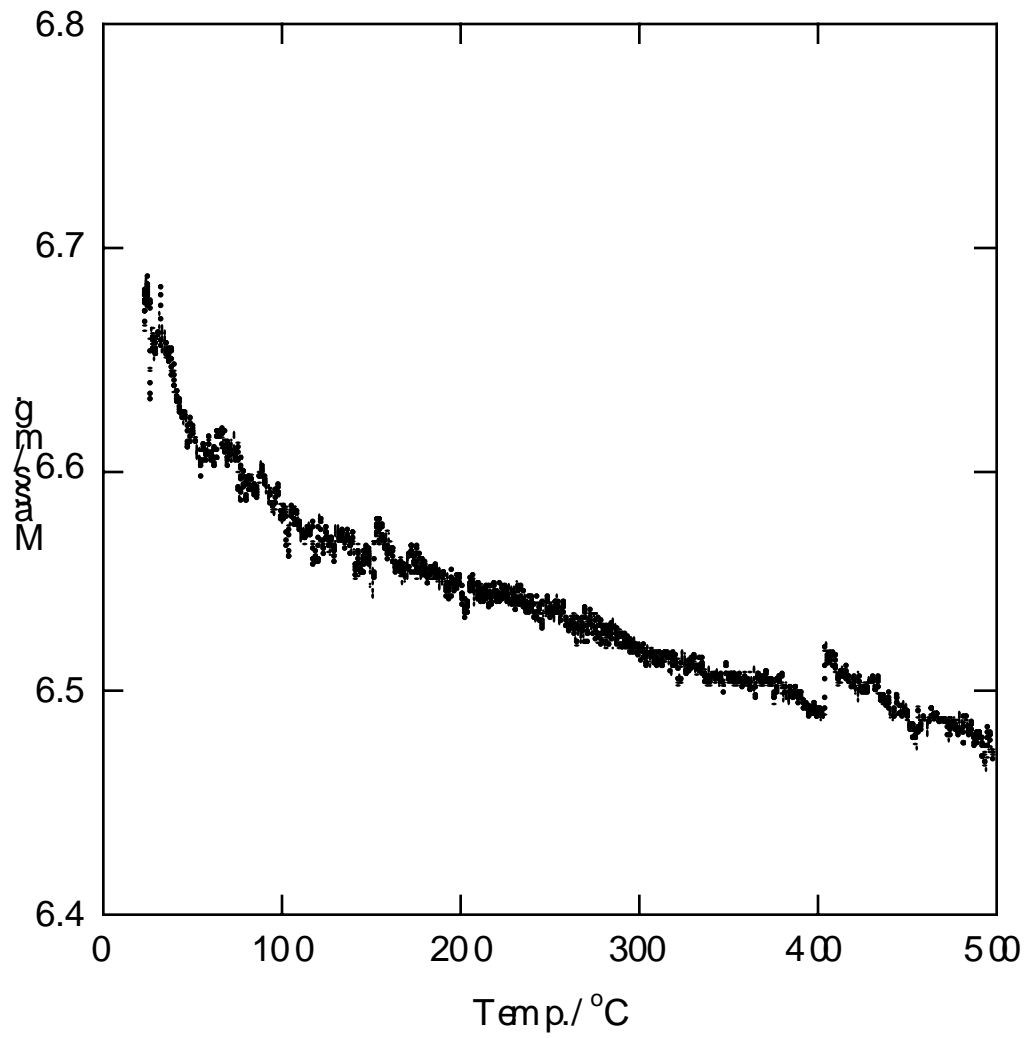
Table 7. Summary of TGA/DTA Results for the ARF Samples.

| Sample ID | Mass Loss / % Range 25 to 1000°C | Mass Loss / % Range 25 to 500°C | Thermal Events/ C |
|-----------|-------------------------------------|------------------------------------|--|
| ARF-3 | 27.62 | 2.9 | 41.8 96.9 651.4 |
| ARF-3A | 29.61 | 1.9 | 37.4 99.8 149.3 649.1 |
| ARF-5 | 28.46 | 0.32 | 88.3 649.4 |
| ARF-8 | 26.30 | 0.35 | 92.8 648.9 |
| ARF-9 | 11.74 | 0.99 | 99.6 146.8 447.8 646.3 |
| ARF-13 | 13.63 | 2.0 | 38.5 104.3 152.3 458.9 648.4 |

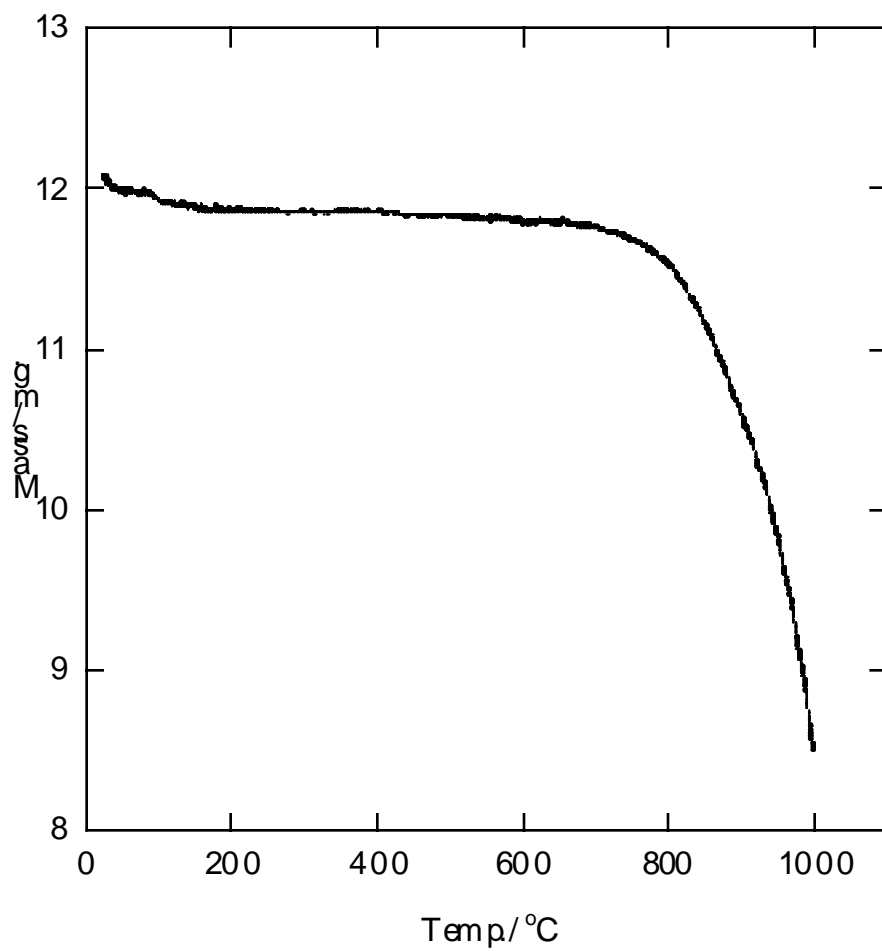
TGA of ARF-3



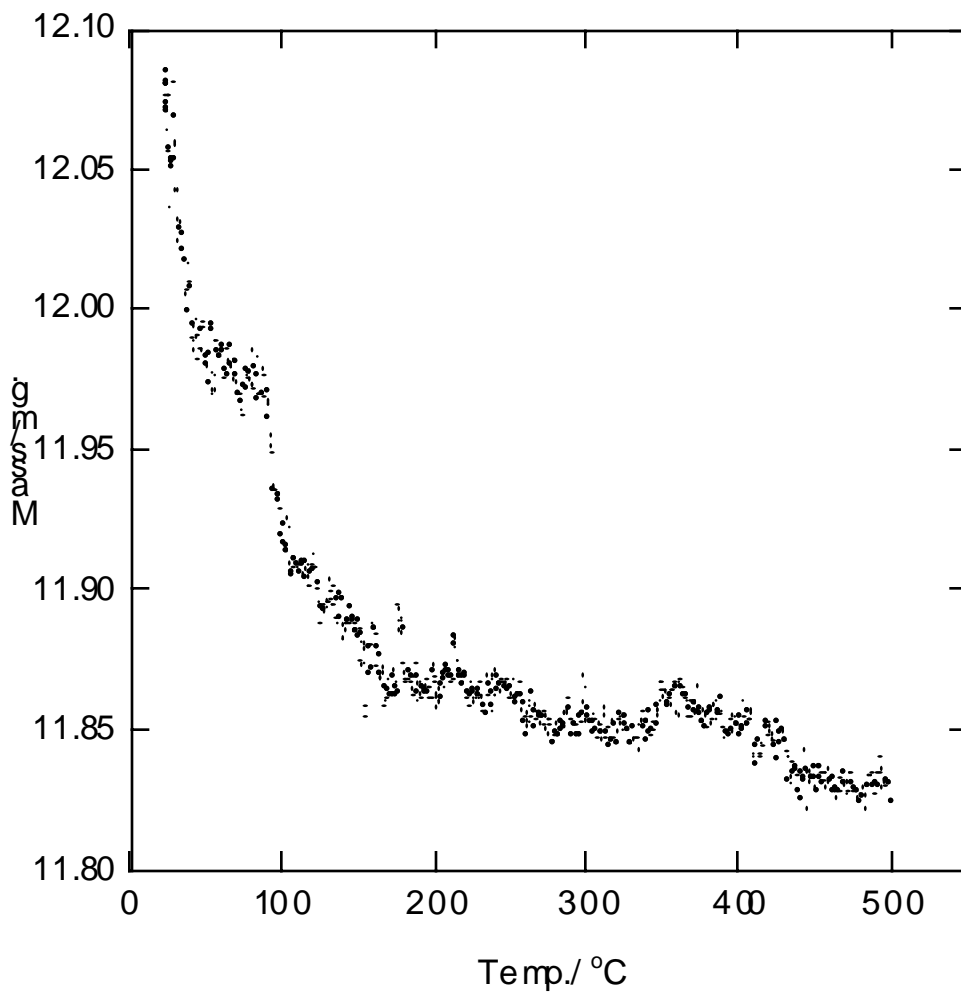
TGA of ARF 3 25 to 500 °C



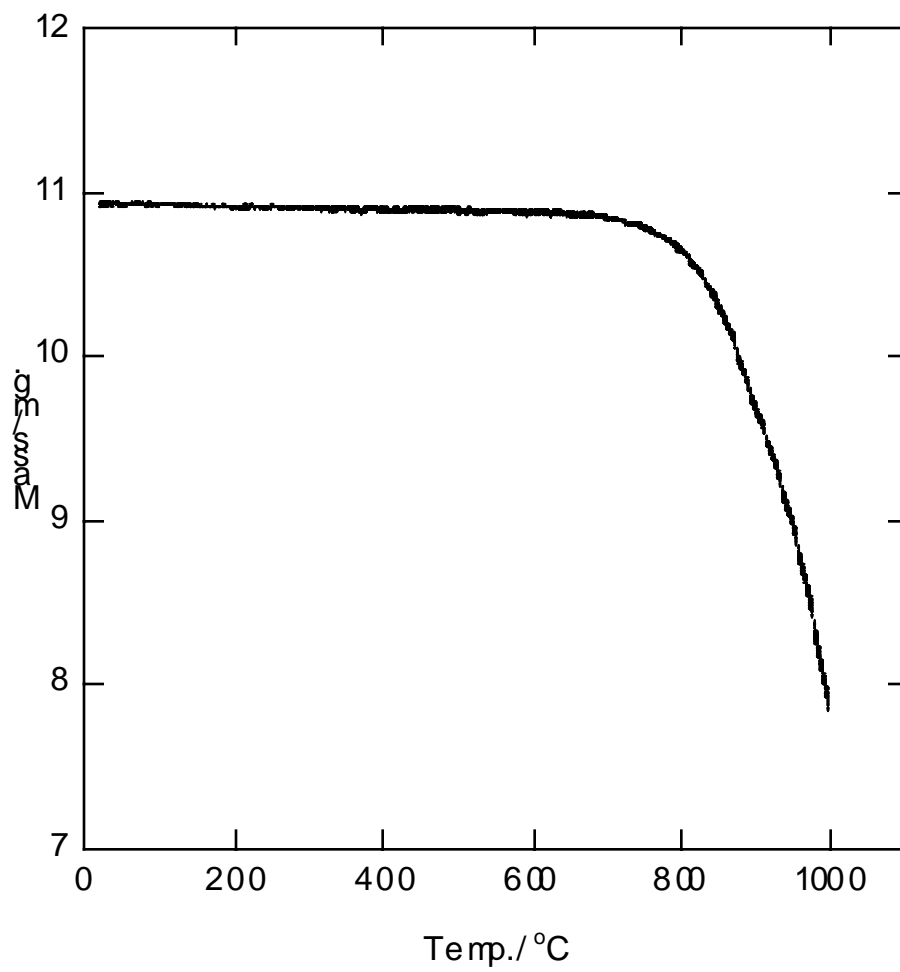
TGA of ARF 3a



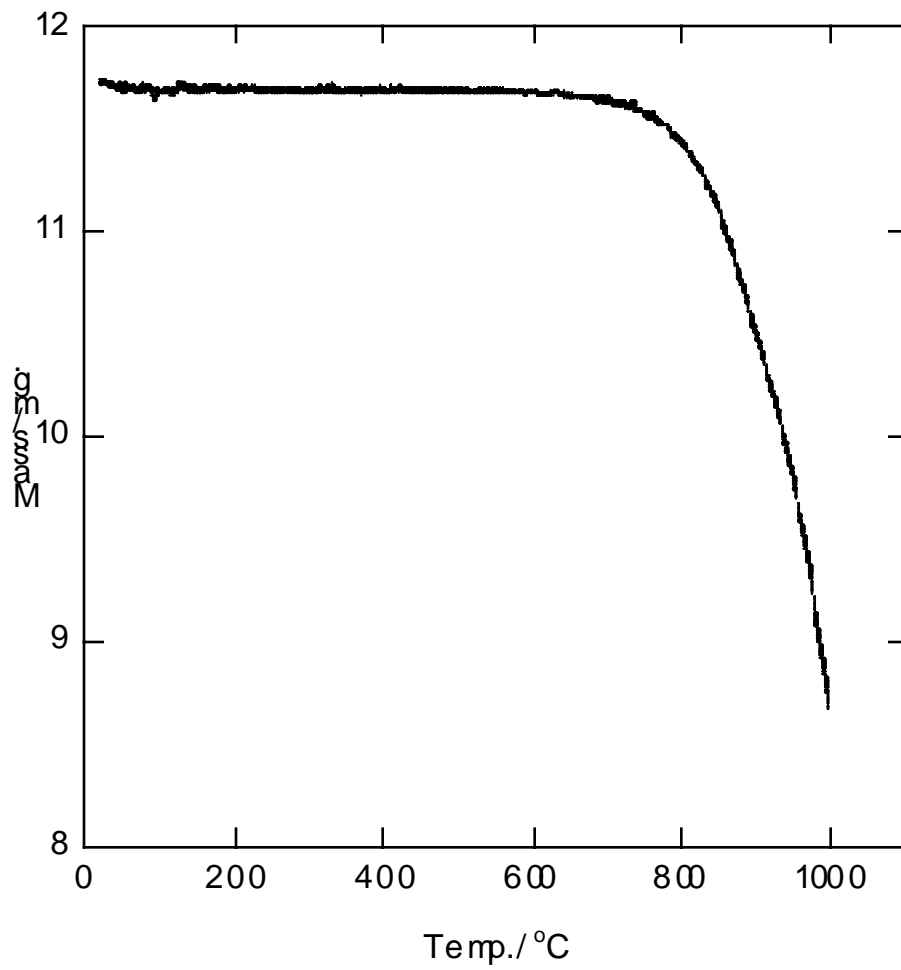
TGA of ARF 3a 25 to 500 °C



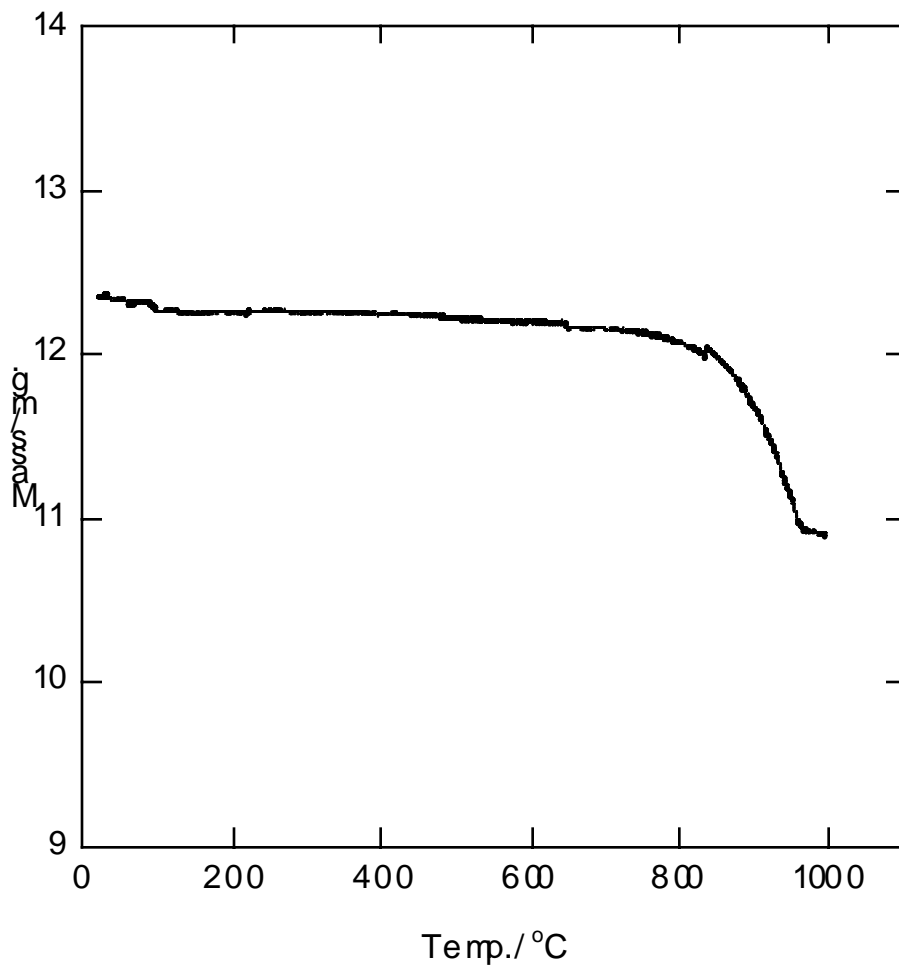
TGA of ARF-5



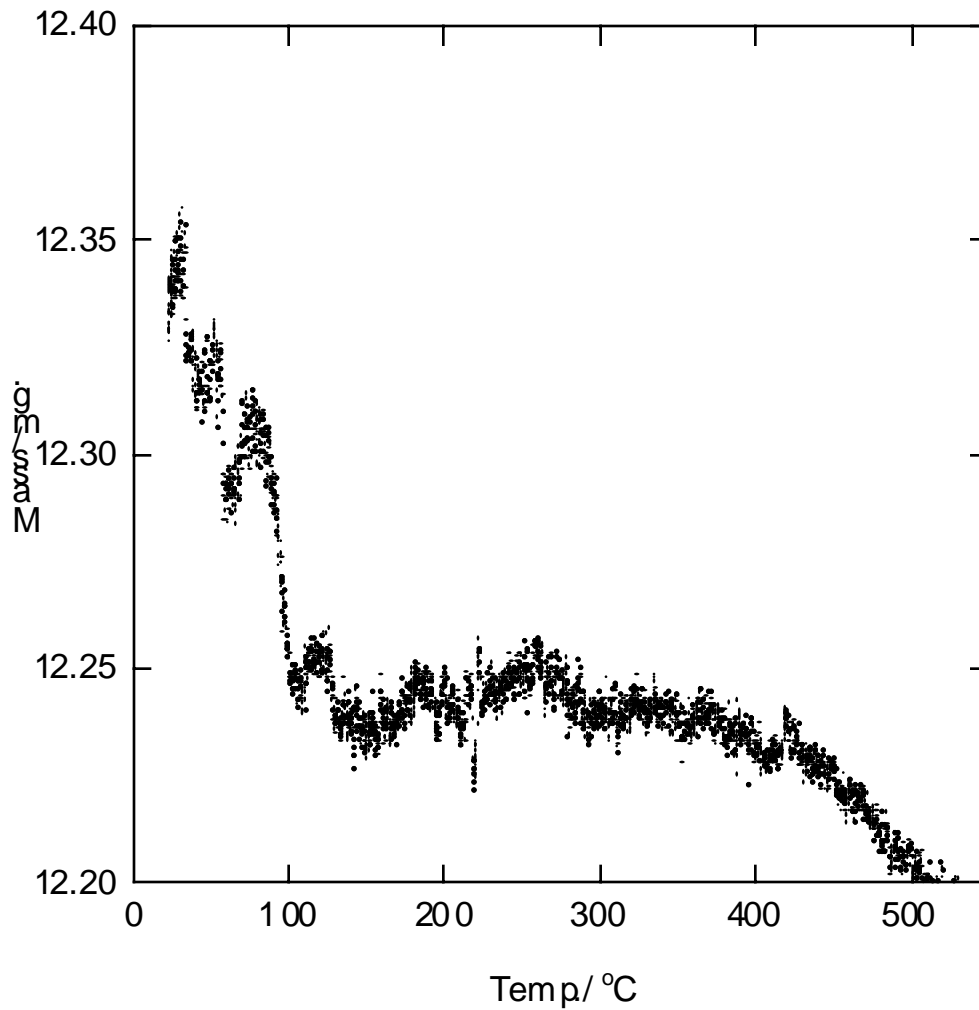
TGA of ARF-8



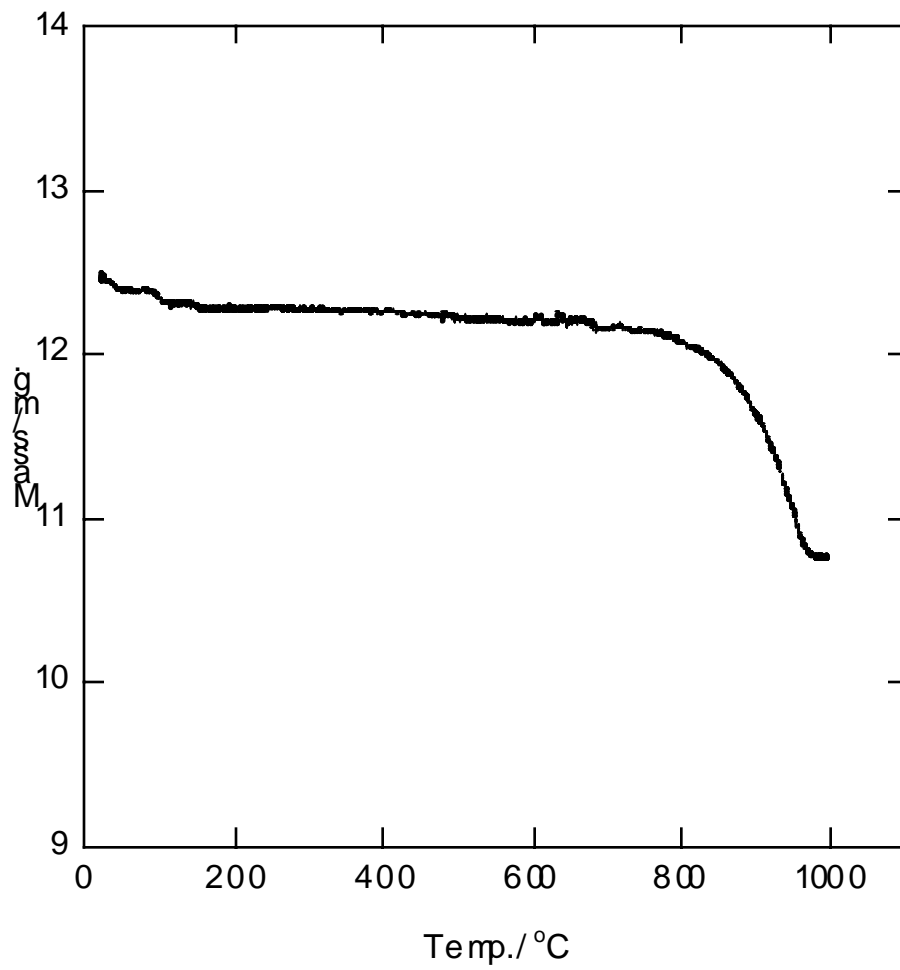
TGA of ARF-9



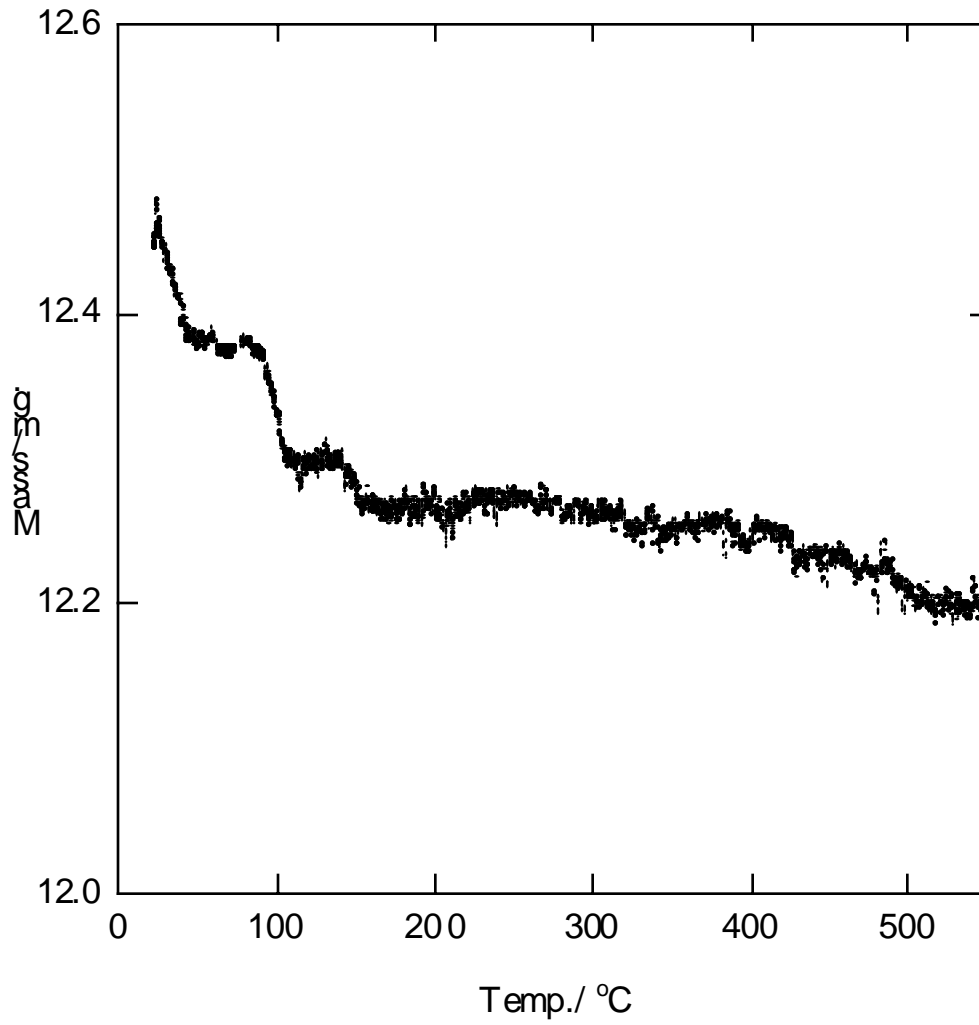
TGA of ARF 9 25 to 500 °C



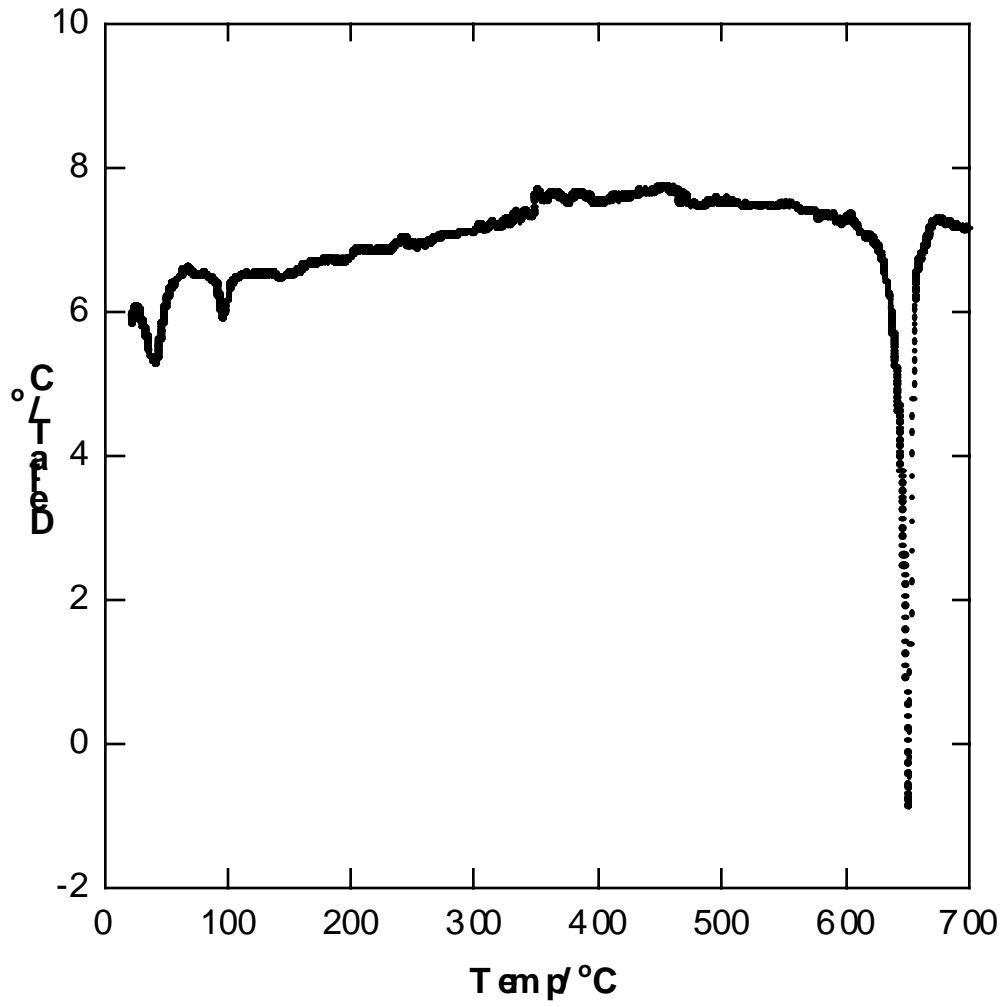
TGA of ARF-13



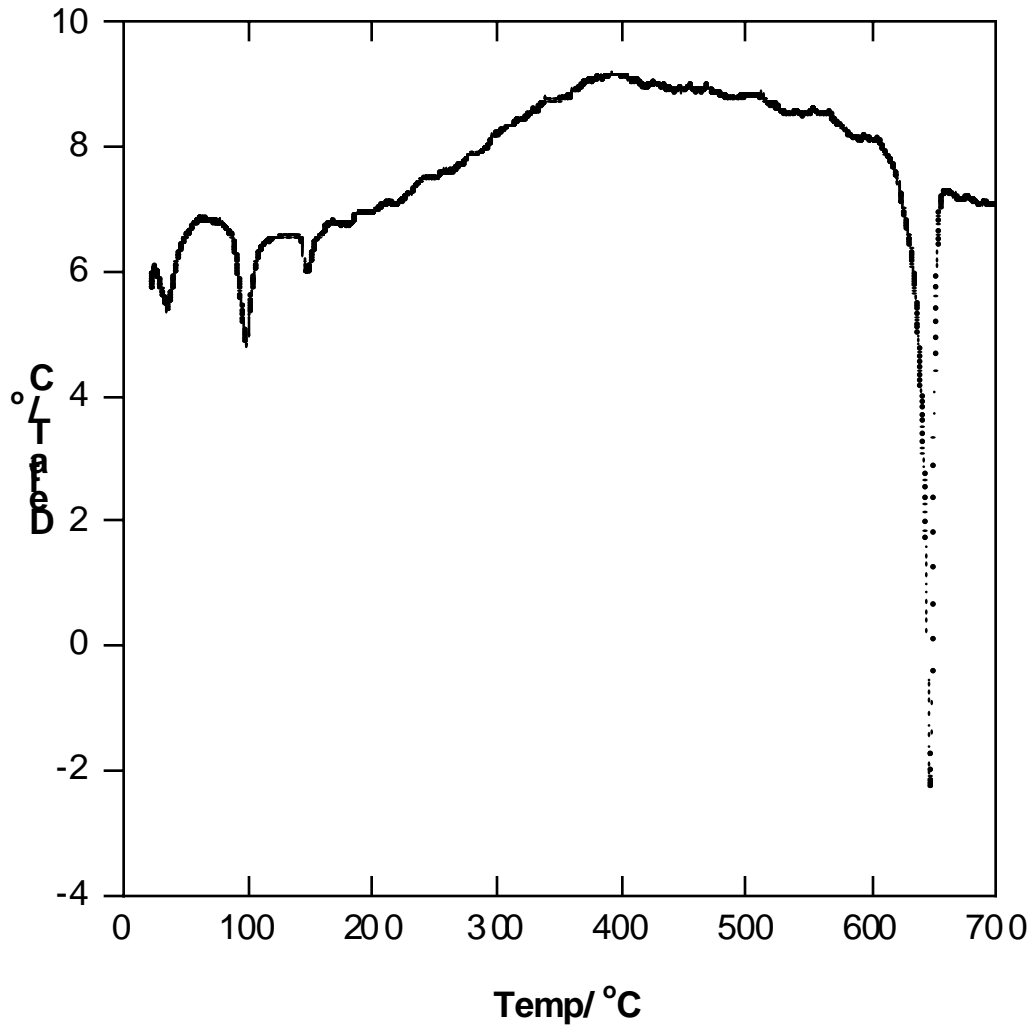
TGA of ARF-13 25 to 500 °C



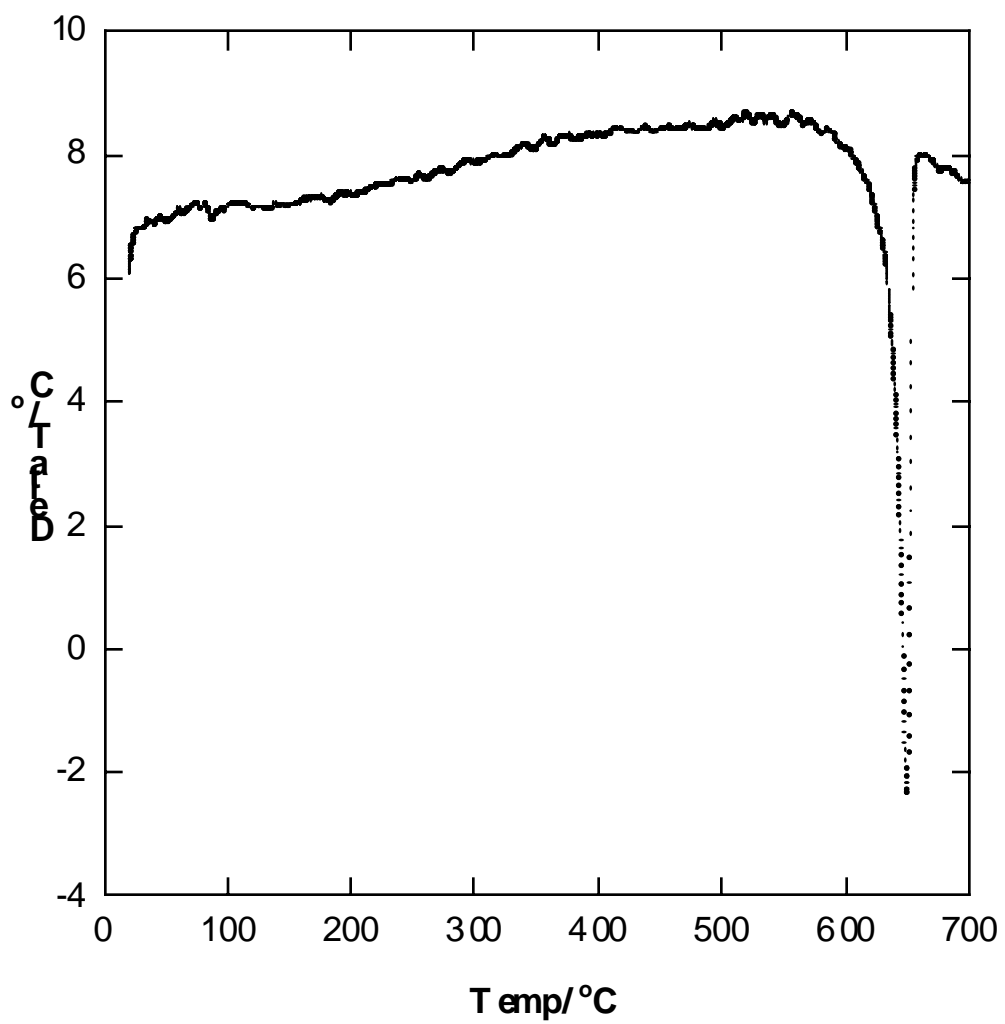
DTA of ARF-3



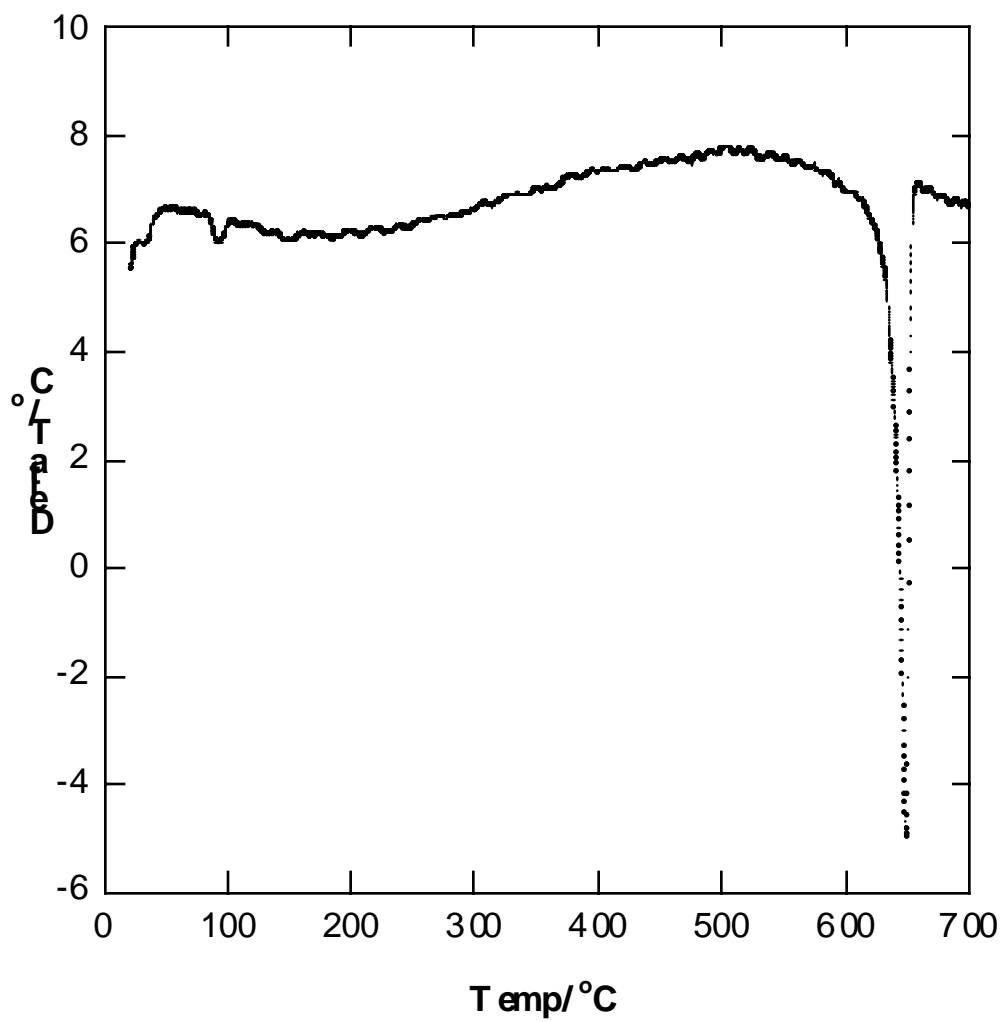
DTA of ARF 3a



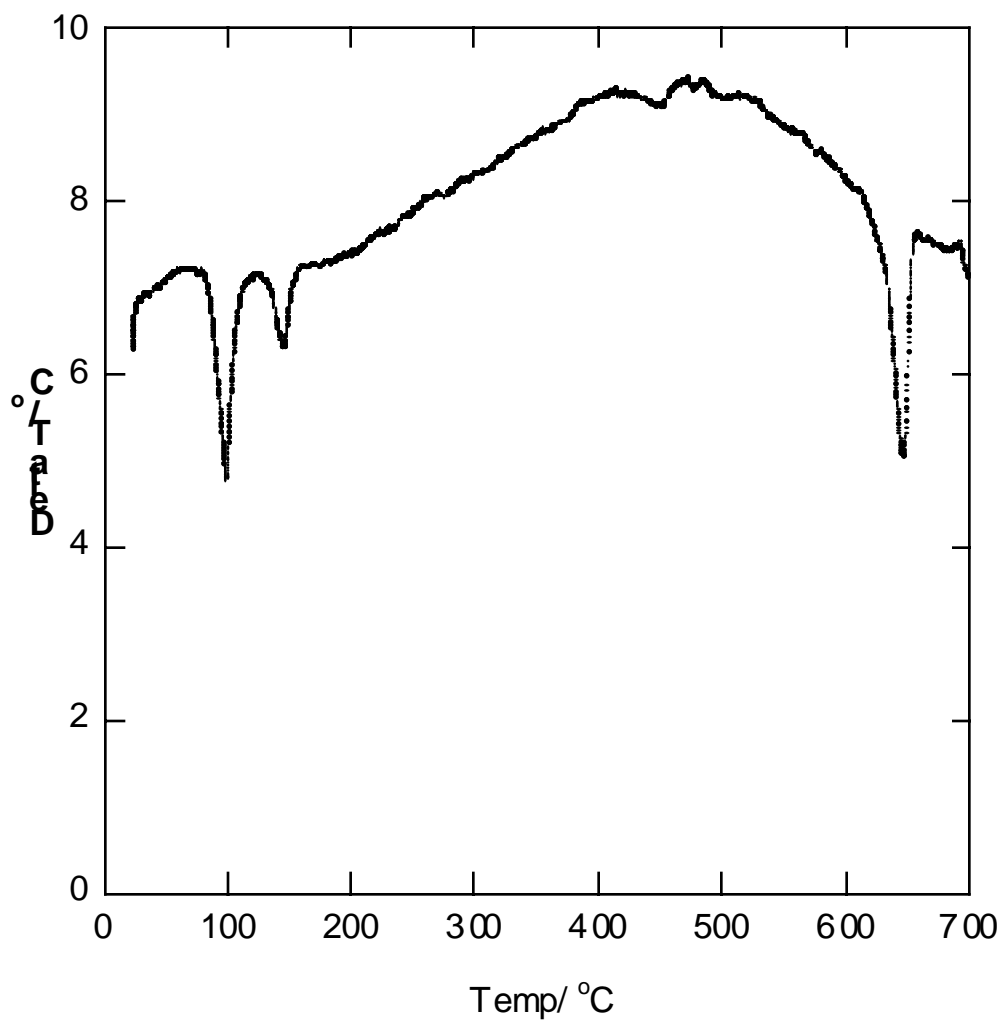
DTA of ARF-5



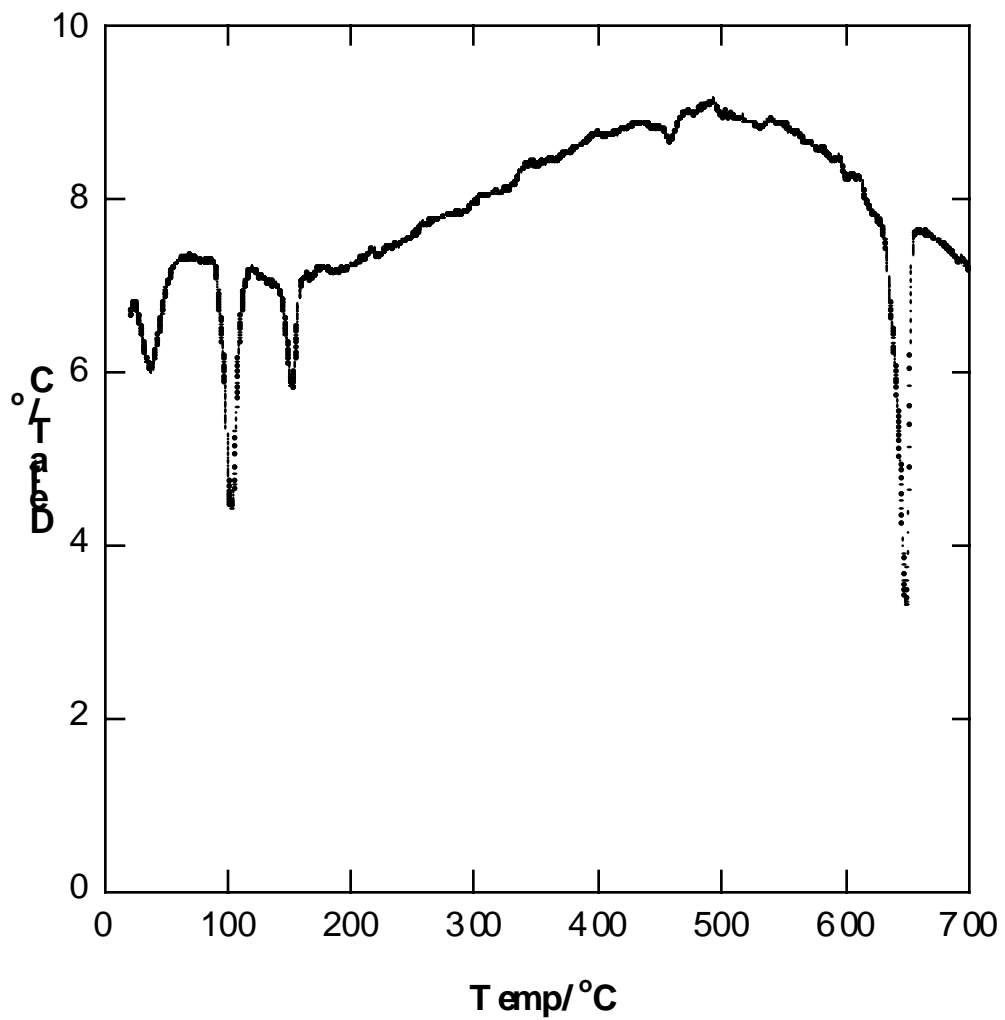
DTA of ARF-8



DTA of ARF-9



DTA of ARF-13



APPENDIX 2.

TD-MS results.

These results will be added later.

Los Alamos

NATIONAL LABORATORY

Los Alamos, New Mexico 87545