Nuclear Materials Measurement Impacts on Plutonium Storage and Disposition

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The Department of Energy requires accurate measurements of special nuclear material quantities and isotopics to support material control and accountability upon creation of an item. Usually the initial non-destructive assay measurements meet all requirements. However, at times the measurements must be repeated (e.g., verification measurements for intersite transfers) or more accurate measurements are required to meet a specific specification (e.g., a disposition program). Savannah River Site performs remeasurements to support feed characterization for the Mixed Oxide Fuel Fabrication Facility and to provide verification measurements for the plutonium and enriched uranium content in packages it has received for storage pending disposition. This paper describes key factors that impede accuracy in meeting the multiple program missions, including the presence of competing elements and isotopes; limited counting duration; and shielding in packages that were created decades earlier.

Surplus Plutonium Consolidation and Disposition

SRS has received more than 5000 containers stabilized to the Plutonium Long-Term Storage Standard, DOE-STD-3013,[1] for storage in the K-Area Materials Storage (KAMS) Facility pending disposition.[2] These containers include 1888 that resulted from the earliest stages of stabilization and packaging, performed at the Rocky Flats Environmental Technology Site (RFETS). Few of those containers had destructive analysis (DA) from samples of the contents; the nuclear materials contents were reported using calorimetry and gamma spectrometry (cal-gamma) on the 3013s after sealing. Cal-gamma is a non-destructive assay (NDA) technique. In early packaging, gamma measurements relied on counting times in the range of 15-30 minutes. Additional semi-quantitative data on chemical impurities from light elements are also available from Prompt Gamma Analysis that was performed on oxide-bearing cans.[3] The oxides range from highly impure (up to 70 wt.% non-actinide impurities) to higher plutonium contents that may be suitable for disposition as feed for the Mixed Oxide (MOX) Fuel Fabrication Facility (MFFF).[4]

Nuclear Material Verification

The material control and accountability (MC&A) program in KAMS attempts to verify the plutonium content and the plutonium-240-effective percentage for all offsite plutonium receipts inside their Type 9975 shipping container, using the shipper's isotopic data and a passive neutron measurement.[5] Some items fail the initial NDA confirmation. SRS identified 341 uranium measurements and 170 plutonium measurements that should be confirmed by cal-gamma.[6]

For enriched uranium, the accuracy of the initial measurements from Rocky Flats was hampered by the fact that uranium-235 has a very weak gamma signature when it decays, making it difficult to assign precision by calorimetry and gamma spectrometry unless the low-energy portion of the spectrum is specifically addressed. Previously many containers were assayed at another SRS facility in a californium-252 shuffler before that equipment was retired.

For plutonium and uranium, the accuracy of the initial measurements was known to be hampered by elevated levels of light elements, e.g., beryllium, fluorine, or magnesium. These elements produce a significant number of (α-n) reactions, making measurement of the plutonium-240-effective impractical. Beryllium is present in 60% of the items that were identified for analysis or verification measurements.

MOX Fuel Feed Validation

Separate from the MC&A questions, the National Nuclear Security Administration (NNSA) is evaluating 3013s with plutonium oxide as feed for the MFFF. The Interface Control Document (ICD) for the acceptance of these Alternate Feedstocks (AFS) sets limits on the isotopic composition of the plutonium, and also on the measured uncertainty in the measurements.[7] This limits would assist the MFFF in verifying that the composition of the fuel made from combining the plutonium from various feeds would meet reactor requirements without rework. Table 1 shows the current limits on plutonium isotopic ranges and absolute uncertainty (i.e., the uncertainty in each isotope as a fraction of the total plutonium).

Table 1. Isotopic Ranges for Alternate Feedstock Feeds for MFFF

Isotope	Range (wt.%)	Absolute Uncertainty at 2 standard deviations (wt.%)
Plutonium-236	≤ 1 ppb	N/A
Plutonium-238	≤ 0.05%	± 0.002
Plutonium-239	90% ≤ X ≤ 95%	± 0.44
Plutonium-240	5% ≤ X ≤ 9%	± 0.44
Plutonium-241	≤ 1%	± 0.02
Plutonium-242	≤ 0.1%	N/A

Factors That Affect Accuracy of Nuclear Materials Measurements

In the context of these materials, key features of the material and site operations that affect the ability to meet measurement requirements include:

- Counting duration
- Equipment arrangement and facility background radiation
- Specific impurities (e.g., Be, B, F, Mg)
- Inhomogeneity of the matrix

Specifically for enriched uranium, additional factors include:

- Low-energy nature of gamma peak for uranium-235
- "Insufficient" plutonium (i.e., a matrix with at least tens of percent plutonium generates sufficient prompt gamma radiation to enhance uranium peaks)

Other factors that affect plutonium include:

- High americium-241 content if used with calorimetry to correlate gamma spectrometry

A special case for the MFFF specifications is the plutonium-242 target value. This isotope cannot be measured with cal-gamma; in usual NDA, a value can be calculated by correlation with the distribution of other isotopes. However, the plutonium from process operations does not follow the standard correlations because it has components that have passed through previous operations since the time it was produced in a reactor. In particular, material from RFETS may have passed through several phases of americium removal during its lifetime. Material from other sites may have a component that resulted from items that were enhanced in plutonium-242. Therefore, NNSA concluded that reported contents of this isotope from the 3013 packaging programs were not reliable compared to knowledge of process history and the origins of the plutonium.

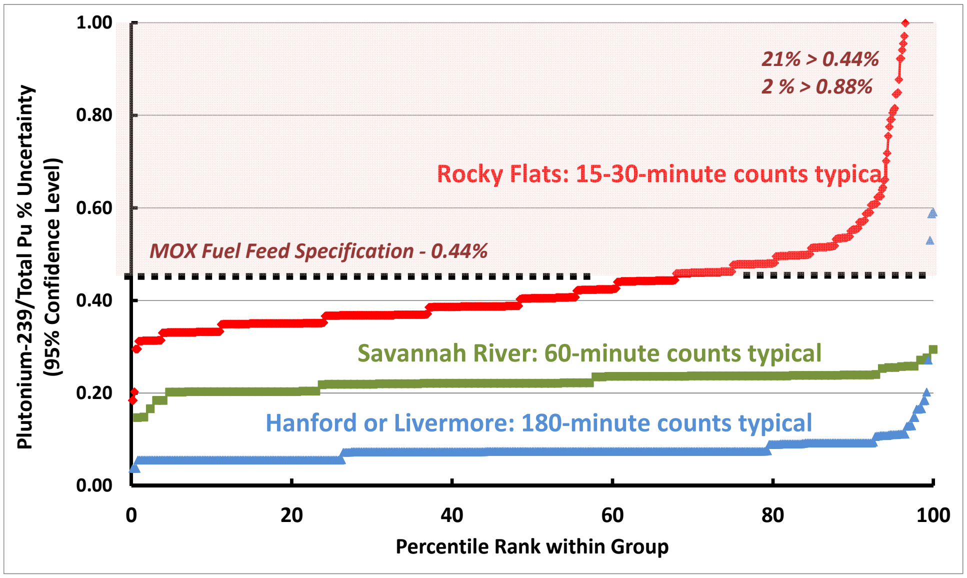
Scope of MFFF Feed Isotopic Uncertainty

Approximately 170 items were found to meet all other requirements for MFFF Alternate Feedstocks, but exceeded one or more of the isotopic uncertainty limits. Most commonly the limits were reached first with plutonium-239 uncertainty, closely followed by the plutonium-240 uncertainty.

Initially, counting time was judged to be the primary factor in the uncertainty measurements. Other sites performed gamma isotopic measurements for longer time periods: Savannah River, typically for 60 minutes; and Hanford or Livermore, typically for 180 minutes. Figure 1 shows a distributions of the site reports for plutonium-239 uncertainty, at 2 standard deviations. Approximately 21% of the RFETS data exceeded the ICD goal of 0.44%, but only 2% exceeded twice this amount. A few of the highest reported uncertainties were identified as temporary equipment upsets that invalidated the individual measurements, and thus a conservative value was assigned by RFETS.

SRS proposed a plan to qualify the items that exceeded the 0.44% threshold, by analysis or, if necessary, by remeasurement, with highest priority given to the highest reported uncertainties.

Figure 1. Distribution of Isotopic Uncertainty for Alternate Feedstocks for MFFF

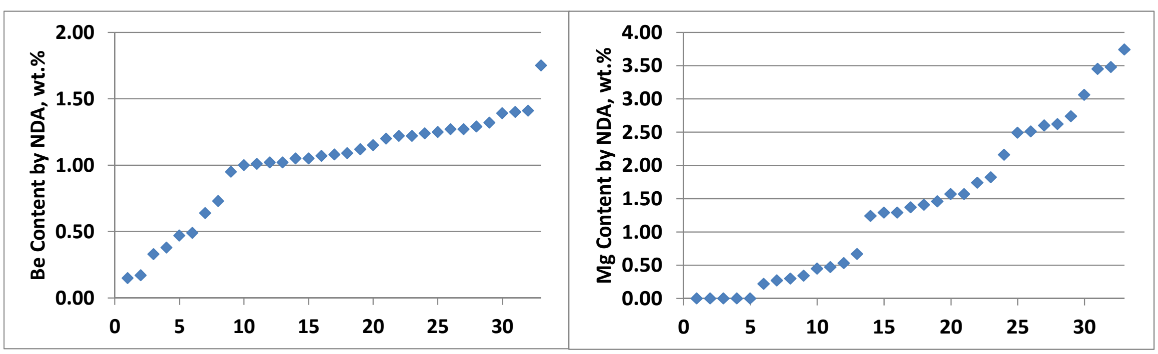


Initial Results from Verification Program

There is an overlap between the items requiring MC&A verification and those that do not meet the MFFF uncertainty goals, but the MC&A items are generally higher in impurity content, and those from RFETS with uranium anomalies generally exceed the MFFF limits of 30 wt.% enriched uranium. However, the requested uncertainties for MFFF plutonium feed are tighter than the Limits of Error targets for MC&A verification.

Figure 2 shows the distribution of beryllium and magnesium in the first 33 items that were fully characterized for plutonium in the verification program. Data points represent PGA elemental data.

Figure 2. Beryllium and Magnesium in Items Requiring MC&A Verification



The charts confirm that the items selected for MC&A verification are dominated by items with beryllium, and secondarily for other light elements, as expected from process history.

When the MFFF remeasurement program was proposed, a significant fraction of the highest-uncertainty items were found to be in the Verification program, but also to have high beryllium. Figure 3 shows the PGA beryllium levels for three priority groupings. Clearly beryllium, and not just counting time, was a major contributor. The small tail at the lower right shows reported beryllium content for more than 1200 other Alternate Feedstock oxide items. It should also be noted that the sites other than RFETS were unlikely to have items with significant Be content.

Figure 3. Beryllium Impact on Uncertainty for RFETS-Origin Alternate Feedstocks

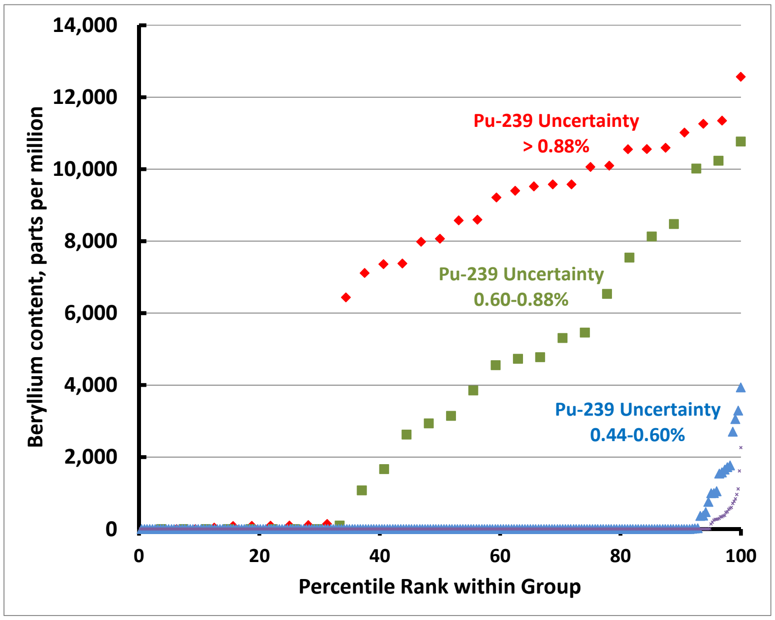


Figure 4 shows the distribution of Be and Mg measurements across the entire population of RFETS oxide candidates for AFS. Trend lines can be misleading because there are other factors that affect the uncertainty measurements, and most items will not contain detectable quantities of the analyzed element. However, a strong trend can be seen for increased isotopic uncertainty with increased Be content, with only a small (if any) correlation seen for Mg.

Figure 4. Isotopic Uncertainty Trends for RFETS Beryllium and Magnesium

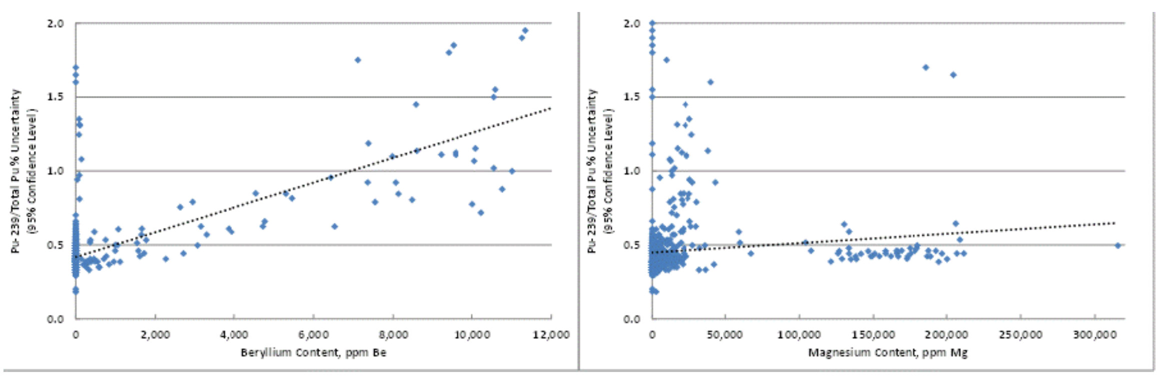
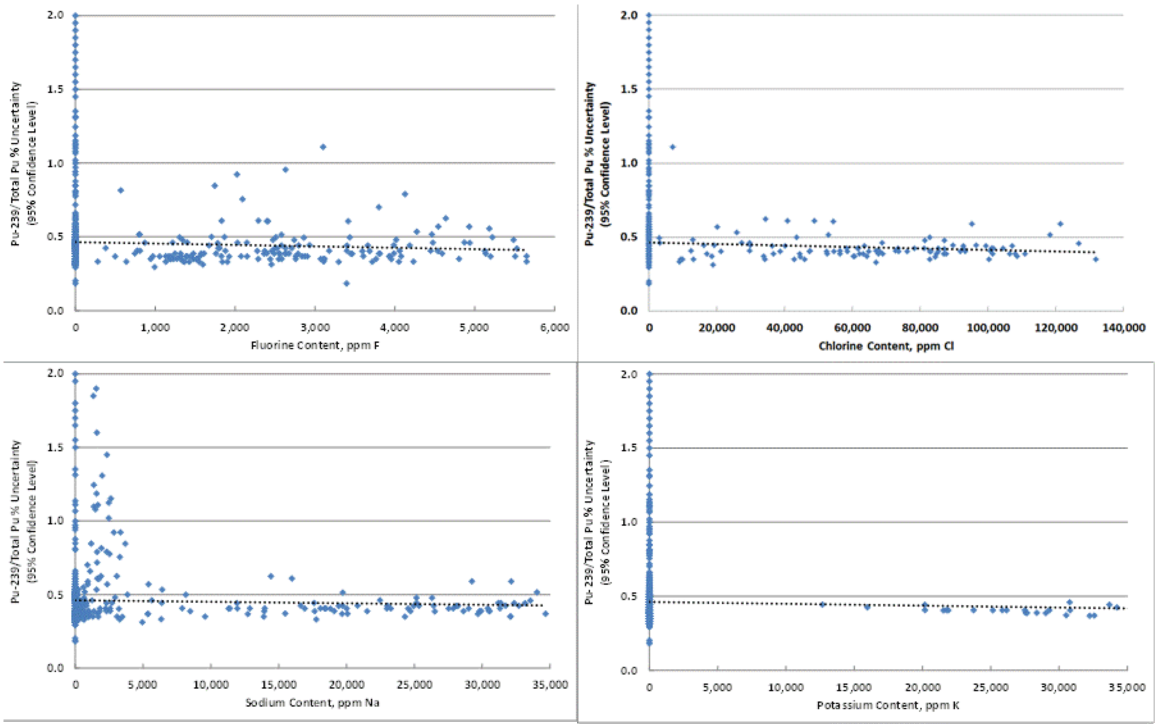


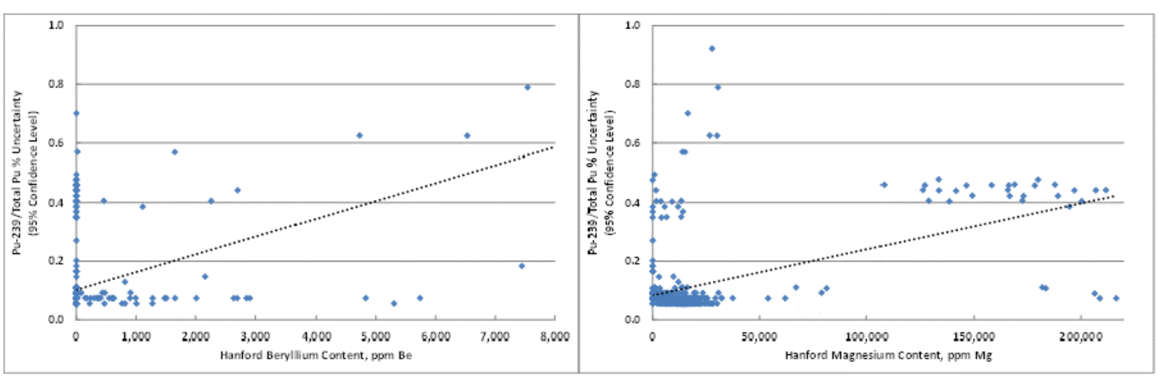
Figure 5 shows similar correlations for fluorine, chlorine, sodium, and potassium (other light elements detectable by PGA). No significant correlation with uncertainty is observed. A shallow trend line is observed for a plot of total impurities versus isotopic uncertainty, suggesting that up to approximately 27 wt.% non-actinide impurities (the limit for AFS), the data that exceed the impurity target are most influenced by counting time and beryllium.

Figure 5. Isotopic Uncertainty Trends for Other RFETS Light Elements



Although only a few items packaged at Hanford exceeded the isotopic uncertainty specification, oxides were also measured by cal-gamma. A portion of the Hanford inventory was stabilized by magnesium hydroxide precipitation and the magnesium may impede high measurement precision. Figure 6 shows charts for Hanford isotopic uncertainty for plutonium-239 versus beryllium and magnesium. Although few items contained Be, a correlation is possibly visible with isotopic uncertainty. The distribution of Mg is bimodal because of the differing stabilization processes, but a mild (but not universal) correlation is also possible.

Figure 6. Isotopic Uncertainty Trends for Hanford Beryllium and Magnesium



Path Forward

SRS has completed verification measurements on the highest priority items and continues to perform analyses on the lower priority items. Remeasurement of potential Alternate Feedstocks for the MFFF is suspended awaiting further definition of the project schedule. To a great degree, the factors influencing NDA special nuclear material measurements for the plutonium materials in DOE-STD-3013 containers are well understood.

References

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