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Improving Neutron Assays of Dilute Surplus Plutonium Using Gamma-Ray Measurements

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Abstract

Neutron coincidence counting has been chosen as the method for assay of diluted, surplus plutonium in Criticality Control Overpacks (CCOs) produced at the Savannah River Site. However, some portion of the material consists of scrap oxide containing low-Z elemental impurities. These impurities interact with Pu and Am alpha particles to create neutrons. In large quantities, these (α,n) neutrons can cause erroneous values with standard neutron analysis techniques and, in fact, can even prevent the analysis from succeeding at all. These large uncertainties and biases can result in measurements falling outside the required bounds for both Nuclear Material Control & Accountability and Waste Acceptance Criteria.

Some of the relevant (α,n) reactions provide an associated gamma ray and, if not, sometimes there is a similar reaction (for example, (α,p)) that produces gamma rays that can be detected. A methodology is under development to allow for the estimation of the quantity and impact of (α,n) neutrons on a Pu neutron assay. This makes it possible to calculate alpha (the ratio of (α,n) to Pu fission neutrons) and utilizing a known-alpha analysis, greatly reduces the associated uncertainty and bias of the measurement. Preliminary implementation has shown positive results and elucidated avenues for further development.

Introduction

The Savannah River Site (SRS) is currently in the beginning stages of a lengthy Dilute and Dispose process intended to eventually discard excess plutonium materials to the Waste Isolation Pilot Plant (WIPP). The plutonium materials are adulterated and eventually packaged into criticality control overpack (CCO) containers. These CCOs are currently required to be assayed to assure the materials meet the WIPP Waste Acceptance Criteria (WIPP-WAC) and to meet Nuclear Material Control and Accountability (NMC&A) requirements. The chosen assay method was neutron coincidence counting using Mirion high efficiency neutron counters (HENCs) [1-3].

Neutron coincidence counting, in general, is fairly sensitive to high levels of background neutrons. While the bulk of the material to be processed is relatively pure PuO_2, some of material was generated by various processes across the DOE complex and contains some quantity of low-Z contaminants. Some of these contaminants, particularly those with $Z \leq 20$, can produce high levels of background neutrons (through (α,n) reactions) that lead to bias in the HENC measurements as well as much larger uncertainties in the final results [4-6].

To begin to address this problem, the materials have been separated into measurement categories or "families" based on the levels of contaminants present. Those materials within a "family" are expected to perform similarly (potentially over a large range) with regard to the bias and uncertainty of the results. Ultimately, the families are defined by the quantity of (α,n) neutrons relative to spontaneous fission neutrons from Plutonium. This ratio is referred to as the "alpha" value for a particular neutron coincidence measurement. Families are further divided into sub-families defined by the predominant impurity (Beryllium, Fluorine, Uranium, etc.).

In most cases (with carbon being the notable exception), neutrons from (α,n) reactions are accompanied by gamma rays. Often these gamma rays are from the (α,n) reaction itself, but sometimes they are from a reaction that happens concurrently, like (α,p). A method was developed to use these gamma rays to estimate the number of neutrons from (α,n) reactions and to predict their impact on the HENC results. The ultimate goal is to reduce or, potentially, eliminate the bias and uncertainty from the contaminating neutrons. The method utilizes a known-alpha analysis method that is sometimes used with neutron coincidence measurements.

Method Description

In the known-α approach [7-10], both the singles and the doubles are used in order to improve the accuracy of the HENC results. This assumes that the alpha value α is well-behaved, but that the $^{240}Pu_{eff}$ mass m and the multiplication M are unknown. In the most common application of this method at SRS, the α value is calculated based on a calorimeter measurement of $^{240}Pu_{eff}$ and the measured singles rate from the HENC. In this work, α is calculated based on the measurement of gamma rays from (α,x) reactions as well as some basic assumptions about the α value of pure oxide material.

The expected singles S and doubles D for an item assayed on a given neutron counter is as follows:

$$S = m\, F_0\, \varepsilon\, \nu_{s1}\, M\, (1+\alpha) \quad (1)$$

$$D = \tfrac{1}{2} m\, F_0\, \varepsilon^2\, f_D\, \nu_{s2}\, M^2\, [1 + k_\alpha (M-1)(1+\alpha)] \quad (2)$$

The efficiency ε and doubles gate fraction f_D are calibration constants specific to the instrument, and F_0, ν_{s1}, ν_{s2}, and k_α are physical constants [10].

If α is known, eliminating m from these equations produces an equation for M:

$$k_\alpha(1+\alpha)M^2 + [1-k_\alpha(1+\alpha)]M - r = 0 \quad (3)$$

where

$$k_\alpha = (\nu_{s1}\nu_{i2})/(\nu_{s2}(\nu_{i1}-1)) \text{ which in this case equals } 2.166 \quad (4)$$

$$r = ((D/S)(1+\alpha))/\rho_0 \quad (5)$$

where ρ_0 is the doubles/singles ratio for a nonmultiplying sample containing only ^{240}Pu.

Solving the quadratic equation for M gives

$$M = (-b_\alpha + (b_\alpha^2 + 4a_\alpha r))/2a_\alpha \quad (6)$$

where

$$a_\alpha = k_\alpha(1+\alpha) \quad (7)$$

$$b_\alpha = 1 - a_\alpha \quad (8)$$

Once the multiplication M is calculated in this manner, the improved $^{240}Pu_{eff}$ is calculated by rearranging equation (1) from above:

$$^{240}Pu_{eff} = S/(F_0\ \varepsilon\ \nu_{s1}\ M\ (1+\alpha)) \quad (9)$$

Specific Algorithmic Approach

Efficiency Calculation

In order to calculate the total number of (α,n) reactions occurring based on a measurement of the gamma rays emitted, it is necessary to know the absolute efficiency for detection at the relevant gamma=ray energies. Depending on the type and similarity of samples to be measured, it may be possible to measure/calculate this for a particular set of samples. However, when the sample quantities and configurations are widely varying, it is not possible to do so.

Conveniently, the materials that are being considered here already undergo gamma-ray measurements for the determination of isotopic ratios and specific powers (W/g). This is typically done using software called FRAM [11]. Over nearly 40 years, FRAM has been developed at LANL, primarily to meet the needs and requirement for measurements at the Los Alamos Plutonium Facility, but it has been used throughout the DOE complex and the world to provide highly accurate results from complex gamma-ray spectra.

In this particular case, FRAM is used to calculate the isotopic composition of plutonium, from which is derived the specific power of a particular plutonium oxide in Watts/g and which can be used to convert $^{240}Pu_{eff}$ into total Pu mass by isotope. It is intended that this particular measurement/calculation would be carried out, at minimum, for all "parent items" (material to be diluted and repackaged) and verified for, at least, a selection of "daughter" items (diluted material repackaged for disposition).

Throughout the last three or four decades, a significant effort has been put into improving the fitting codes that are at the heart of FRAM. It makes sense to use FRAM to provide the specific gamma-ray fitting results that are used as the basis for the Prompt Gamma Analysis (PGA) algorithm under consideration here.

A gamma-ray spectrum extending to approximately 1 MeV is acquired using the appropriate HPGe detector and electronics. This spectrum is then processed through FRAM using an appropriate parameter set. The isotopic results for the sample are obtained from FRAM, as well as the peak-fit results that are used by FRAM to calculate the efficiency and specific plutonium peak results that are used to normalize the data.

These peak-fit results are then fit to a standard efficiency model using a non-linear least-squares minimization implemented in Python. The exact method used depends on whether the fit parameters are bounded or not (Trust Region Reflective method for a bounded fit and Levenberg-Marquardt method for an unbounded fit). The fit can also be weighted based on uncertainty, though weighting the fit tends to favor the lower-energy region to the detriment of the higher-energy region of the efficiency curve.

The parameterization of the efficiency fit is based on the Duc Vo physical efficiency model within FRAM. The model allows for multiple absorbers and multiple efficiency curves. It is based on the physical reality of the measurement scenario and is more capable of being extrapolated to higher energies. This is very important for this application as establishing the absolute efficiency above about 1 MeV will be difficult, if not impossible. An example of this efficiency fit is shown in Figure 1.

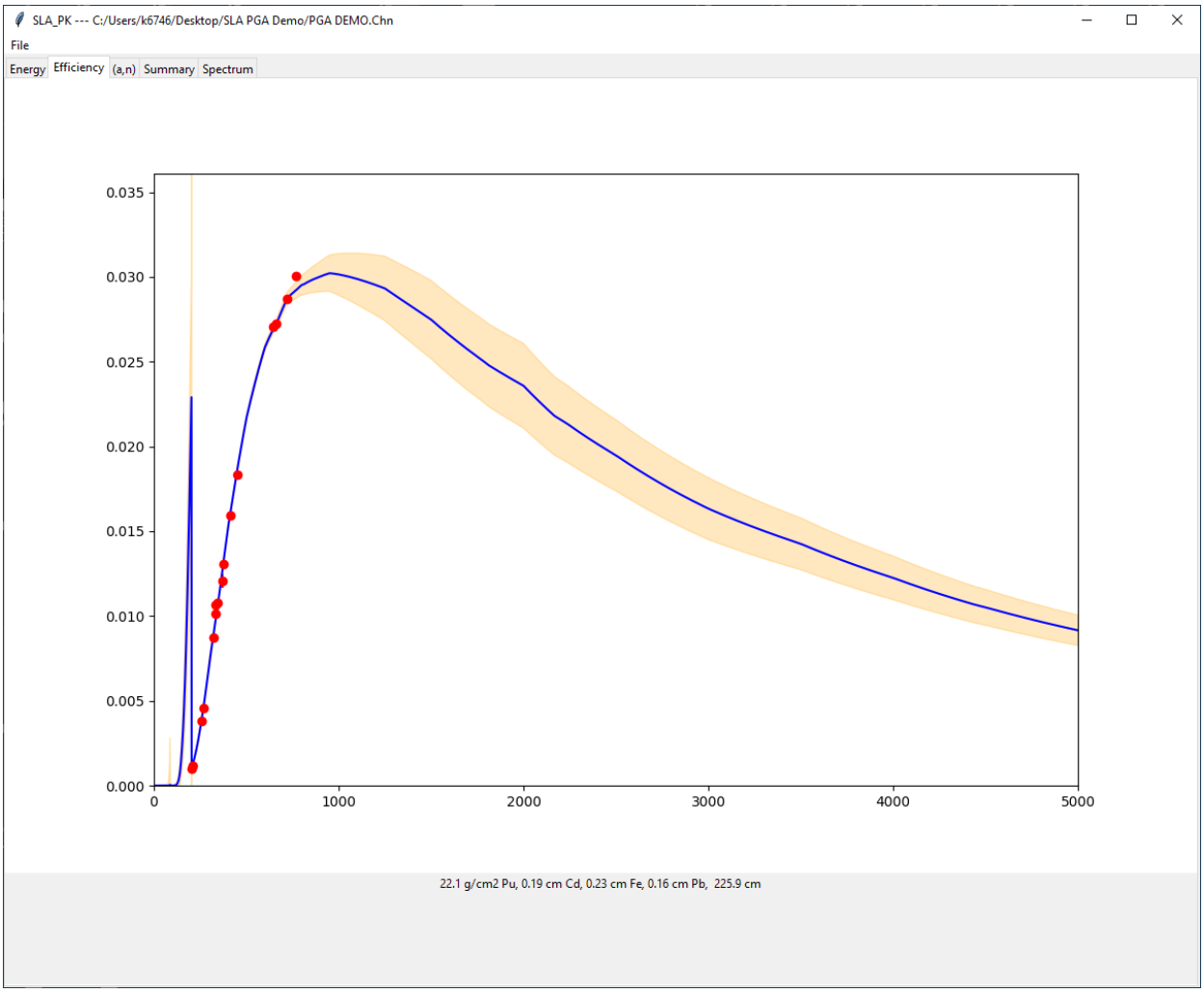

Figure 1: Parameterized efficiency fit for a typical sample measurement of a 3013 container

Peak Fit Results

Once the efficiency of a particular detector/measurement combination has been calculated, the prompt-gamma-ray peaks of interest are fit. These are as follows:

1. Be 4439 keV
2. F 891 keV
3. Cl 2168 keV
4. Na 1809 keV
5. MG 1779 keV
6. Al 2236 keV

This is shown below in Figure 2.

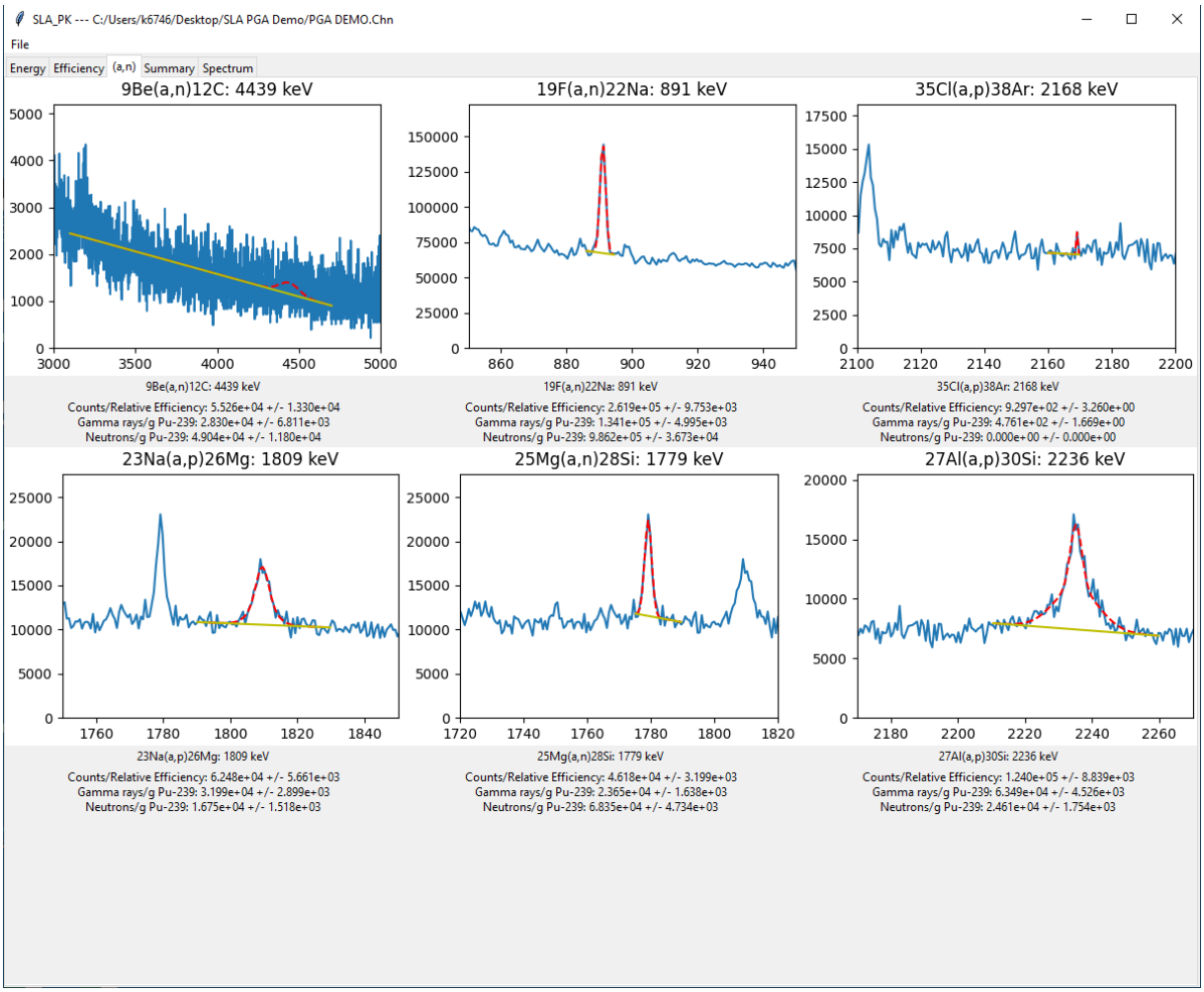

Figure 2: Gamma-ray fits for the six lines of interest for the PGA algorithm for a typical sample

Further algorithmic processing

Using the calculated efficiency and the PGA peak-fit results, the PGA peaks are normalized to Pu peaks retrieved from the FRAM output. These PGA gamma yields are then converted to neutron yields using the appropriate γ/n ratios (calculated or measured, as available).

These neutron yields, normalized to the Pu, are then used to calculate a total α value for the sample based on standard Pu fission neutron yields. This value is summed with a calculated value for the oxide based on ideal conditions and an overall "known" α value is determined.

Using this "known" α value, the NMC singles and doubles values, and equations (1) through (9), the multiplication, M, and $^{240}Pu_{efff}$ are calculated for the sample. A sample output is shown below in Figure 3.

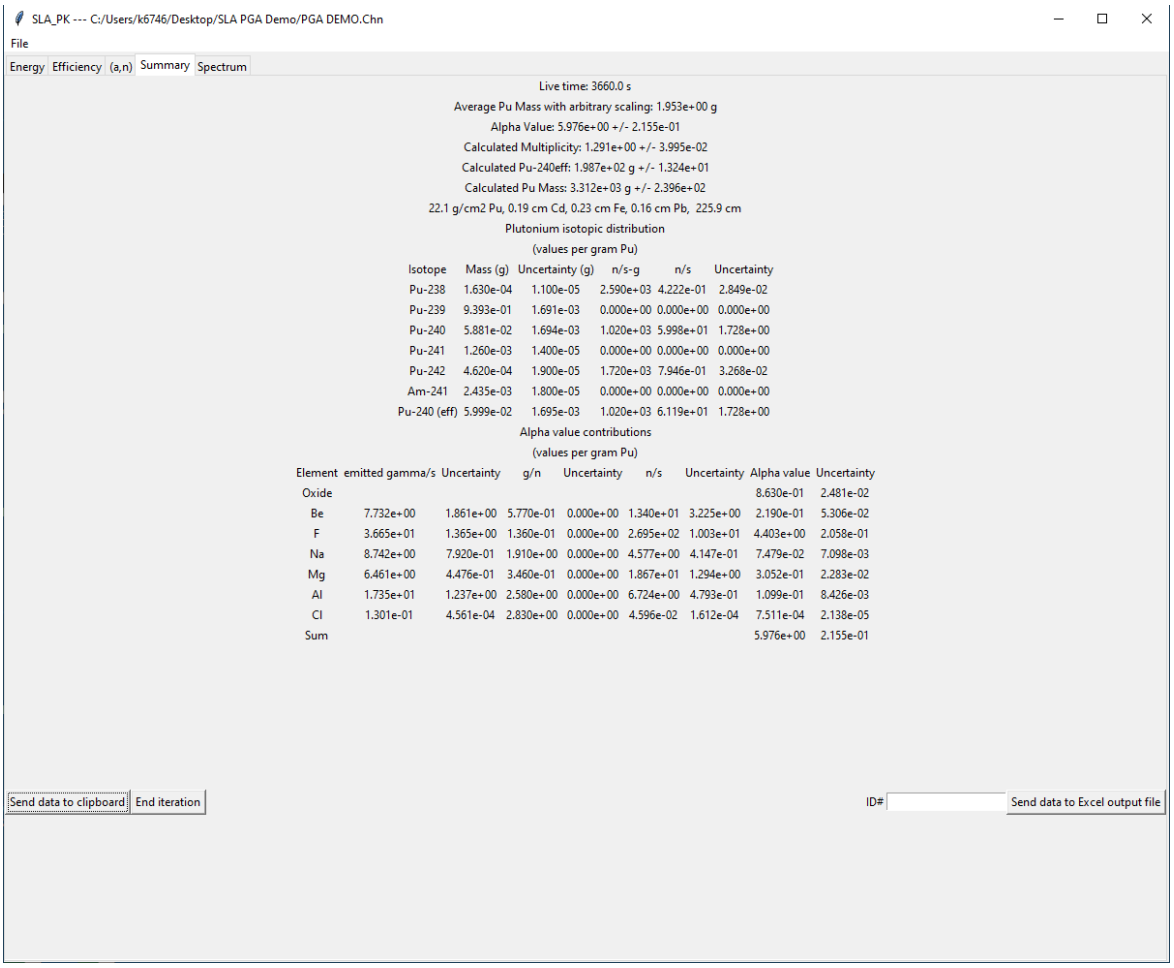

Figure 3: Sample output showing the results of the algorithm

Results

Various other programs related to plutonium storage and disposition have resulted in a small group of samples being analyzed by gamma-ray spectroscopy, neutron multiplicity counting, calorimetry, and, in some cases, destructive analytical techniques. These are fairly well-characterized and represent a small data set on which to test the PGA algorithm.

When these samples are analyzed and the calculated Pu inventories are compared to the declared value, the results shown in Figure 4 are produced.

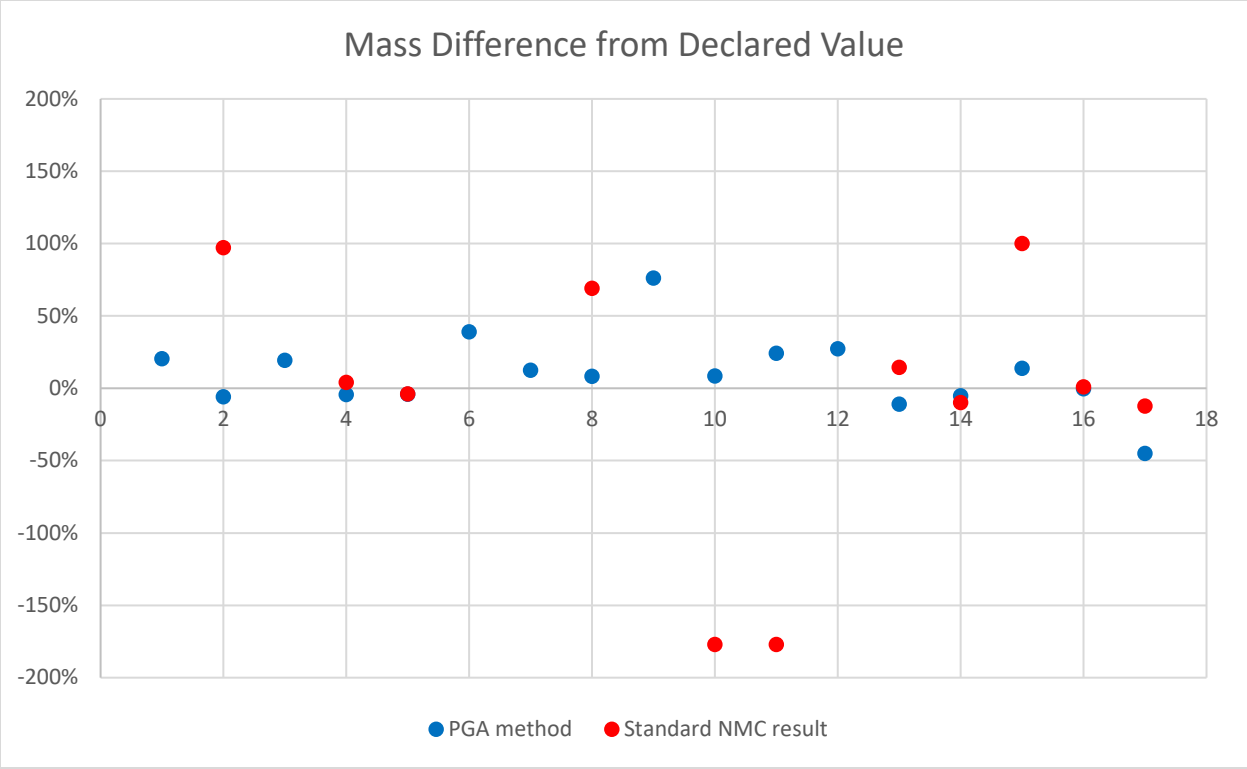

Figure 4: Comparison of PGA algorithm results to standard NMC analysis for well-characterized samples (missing NMC results are far off-scale or altogether impossible to calculate)

The results show that in nearly every case, the PGA algorithm provides improved results and less bias versus the traditional NMC analysis.

Conclusions

The test data set samples showed good agreement between the calculated Pu mass and the known or "declared" Pu mass for many single contaminant samples. For example, samples showing significant fluoride contamination show a deviation from the known value of just under 10% on average. Some of the other samples, particularly pyro-processing and metal samples perform much worse.

Initial indications are that there are whole families where the PGA algorithm performs well. However, mixed contaminant cases seem to perform less well, in general. This is probably explained by the fact that the impact of the (α,n) neutrons and the induced bias varies with the energy of the neutrons. Current research aims to couple neutron spectral measurements with the PGA algorithm in order to account for this effect.

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