Application of Asymptotic Uncertainties in Automatic Non-Destructive Analysis of Plutonium Isotopics

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The Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) software is widely used to analyze the isotopic compositions of plutonium or uranium. The uncertainties reported from FRAM have been studied as a function of the live time for plutonium samples with Pu-239 fractions ranging from 60% to 93%. For plutonium fractions, the specific power and other parameters calculated from FRAM, we have found that the associated uncertainties often converge quickly to some asymptotic value. An asymptotic behavior is present when the counting statistical uncertainty is no longer the dominant contributing factor. Thus, the asymptotic behavior can be used to determine when the measurement can be ended in an automated data acquisition and analysis process. With the process, the acquired interim data are saved and FRAM analysis is initiated in command mode without user intervention. Once the data have been analyzed, the FRAM analysis reports are parsed. The reported uncertainties are compared with results from prior analysis automatically. If the uncertainties are found to approach asymptotic behavior, then the data acquisition can be terminated. For samples with an unknown amount of plutonium or uranium, this method has the advantage in that satisfactory results can be obtained automatically without specifying a preset count time. With a conventional measurement method, the same measurement time is often predefined for all samples. The acquired data are analyzed and the results are reviewed manually. If the results are not satisfactory, the process is repeated. For small numbers of measurements, this manual process might be fine. However, to deal with a large number of measurements, it is inefficient. Thus, the proposed automated analysis process would be beneficial to obtain satisfactory results with greatly improved efficiency.

Keywords: Non-destructive analysis, FRAM, uncertainty vs. time, asymptotic uncertainty, power function

Introduction

The Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) software is widely used to analyze the isotopic compositions of plutonium and/or uranium samples. The total uncertainty has both the random and systematic uncertainties included by default with the latest version.[1] When the sample amount or the isotopic compositions are unknown, it is difficult to know how long to measure the sample. A preset real or live time is often the method used. If the uncertainty for the measured quantity is too high once the preset has been reached, then the measurement is continued to reduce the uncertainty. When there are many samples to be measured, this process could be time consuming. Thus, it is worthwhile to investigate a method for improved efficiency. At the French Institute for Radiological Protection and Nuclear Safety (IRSN), a stop-criteria software has been developed. [2] During data acquisition, the spectra were saved at a fixed time interval and analyzed with FRAM. The results were compared with the average of 10 previous results. If the difference was less than a predefined precision, then the data acquisition was stopped. At the International Atomic Energy Agency (IAEA), FRAM is also integrated with the MCA Touch (MCAT) software for enhanced analysis capabilities. [3]

We have investigated a different approach to determine when the data acquisition should be stopped. The method is based on analyzing the trend of uncertainty over time. During data acquisition, interim spectra are saved at a predefined interval (10 seconds). Those spectra are analyzed with FRAM in embedded mode. The FRAM reports are parsed to obtain the uncertainties of interest. Then the uncertainty data are fitted with a simple power function. After the fit, the uncertainty at any given time can be calculated. The uncertainty

calculated at a predefined maximum allowed data acquisition time is called the *asymptotic uncertainty*. The calculated uncertainty at the time that the data acquisition is stopped is called the *stop uncertainty*. The difference between the two calculated uncertainties is compared with a predefined value which is called the *stop-criteria*. If the difference is smaller than the stop-criteria, the data acquisition is stopped. If the difference is larger than the stop-criteria, the data acquisition continues. This process is repeated until the stop-criteria is met, or until the maximum allowed data acquisition time has been reached.

There are several advantages with our method over the one used in the IRSN software. Uncertainty is often a better indicator of the results desired. By curve fitting, it is easier to assess the true trend of the uncertainties. The range of uncertainties is usually from 0.1% to 100%, making it easier to define the uncertainty precision needed. With a fitted power function, the uncertainty at any given time can be calculated. Thus, it is easier to decide when the data acquisition should be stopped based on the trend curve.

Data Acquisition and Search for A Stop Time

Spectra with very low counting uncertainties are needed to study the asymptotic uncertainty behavior. Plutonium samples were measured with a coaxial P-type High Purity Germanium (HPGe) detector surrounded by a stainless steel clad 50mm thick lead shield and cooled by a mechanical cooler. The detector faced into a stainless-steel enclosure where the sample was placed on a platform central to the detector's field of view as shown in Figure 1. To maintain a gross count rate between 10000 and 50000 counts per second, the sample-to-detector distance was varied, and a cadmium (Cd) filter was used, as needed. Measured samples included metals, residues, and well characterized standards (Pu-239, Pu-238, and mixed oxide (MOX)). The ORTEC Maestro-32 MCA Emulator software was used for data collection. A total live time of 7200 seconds was maintained for all samples; except for the Pu-238 standards which ran for 14400 seconds. Those acquired spectra are used as master spectra to study the trend of uncertainty over time.

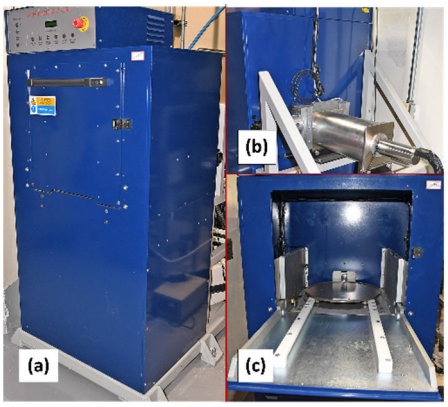

Figure 1. (a) shows the full enclosure set up, (b) shows where the detector interfaces with the shielded enclosure, and (c) is the chamber where the sample is placed.

Process for the Study of the Uncertainty Trend

To investigate the trend of uncertainty over time, a process shown in Figure 2 has been developed. In the process, interim spectra are saved after every 10 seconds of live time has elapsed. The maximum allowed data acquisition time (live time) was 7200 seconds for the majority of the measurements. The live time was doubled for the Pu-238 samples.

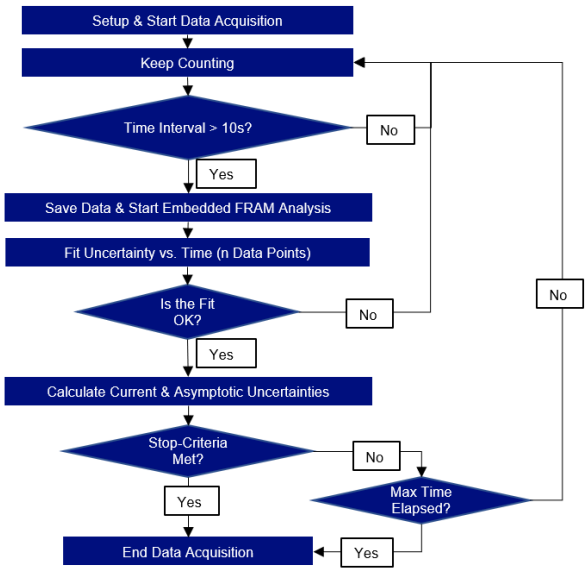

Figure 2. Flow Chart for Searching for a Data Acquisition Stop Time.

In our studies, the first spectrum saved has a live time of only 10 seconds. The spectrum is analyzed with FRAM in command-line mode. After the FRAM analysis, the uncertainty of interest (e.g., the uncertainty of the specific power) is parsed from the report. That uncertainty is the first data point for trend analysis. Once there are 5 data points available, they are fitted with a power function representing the uncertainty trend. After the fit, the uncertainty at the last data point is calculated based on the fitted curve. That uncertainty is compared to the asymptotic uncertainty calculated at the maximum allowed data acquisition time. If the uncertainty difference is smaller than a predefined constant value (the stop-criteria), then a stop time is declared as found. Otherwise, the data acquisition continues for 10 more seconds. After 10 seconds, one more point is added to fit the power function. The calculated uncertainty difference is compared to the stop-criteria again to determine if the data acquisition can be stopped. Those steps are repeated until a stop time is found, or until the maximum data acquisition time is reached.

It is time consuming to acquire data to find a stop time since the stop time is often longer than an hour. Thus, simulated spectra based on the acquired master spectrum are used instead. It takes insignificant time to generate a simulated spectrum with any given live time, helping to speed up the algorithm development tremendously. Thus, data acquisition of the master spectrum can be performed only once. The live time of the master spectrum should be longer than the stop time. Then, only one master spectrum is needed for each measurement configuration.

Data Simulation

To create a simulated spectrum with any given live time based on an acquired master spectrum, the averaged counts at each channel is calculated from the master spectrum by scaling of the counts by the ratio of the two live times. Then, a random sample for the counts at that channel is drawn from a Poisson distribution. The scaled count at that channel is taken as the true mean value for the Poisson distribution. Thus, the counts at each channel in the master spectrum should have very high statistical precision. As a result, all the master spectra have been acquired with long live times only once for each master spectrum. A measured spectrum and a randomized spectrum with the same live time of 60 seconds are compared in Figure 3. The two spectra are almost indistinguishable. To access the quality of simulated spectra quantitatively, 120 simulated spectra with 60 seconds of live time were generated and analyzed with FRAM. The specific powers and the associated 2-sigma uncertainties are plotted for 20 selected spectra. On the graph, all the specific powers are well within the 2-sigma uncertainties. The averaged specific power for the 20 spectra is 2.627 mW/g Pu, well

within the known value of 2.630 mW/g Pu for the Pu sample. The averaged value is the orange horizontal line on the graph.

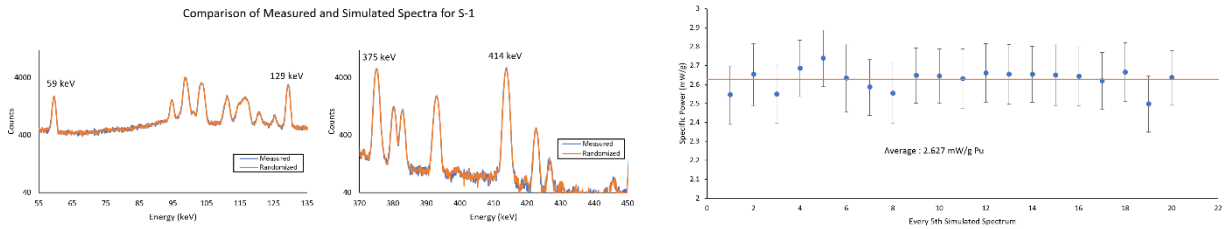

Figure 3. (a) Simulated and measured spectra. (b) Specific powers with 2-sigma uncertainty from simulated spectra.

Trend of Uncertainty Over Time

In Figure 4, simulated spectra were analyzed with FRAM. The total uncertainty for the specific power as a function of time was shown for up to 3000 seconds of live time. After about 500 seconds of live time, the total uncertainty was close to 1%. After about 1000 seconds, the total uncertainty decreased slowly. The dotted line in the graph is a fit of the data points with a power function. The FRAM parameter set used was GeCoax_Pu_120-420, and all the parameters were at the default values. A similar uncertainty trend has been found for Pu-239 fraction, effective Pu-240 fraction, and other quantities of interests.

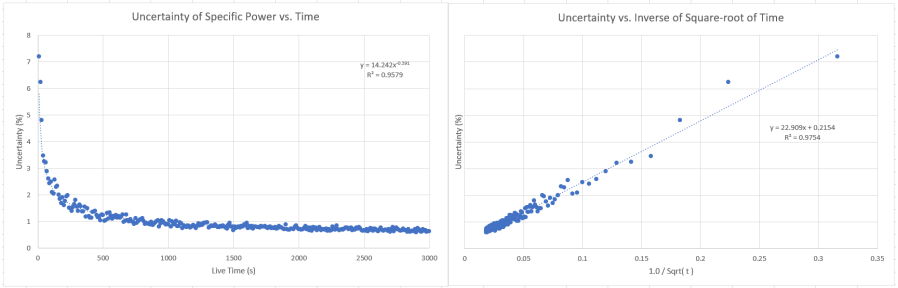

Figure 4. (a) Uncertainty of specific power vs. time (t) for a Pu standard. (b) When x-axis is 1/sqrt(t), all data are close to a line.

In Figure 4(a), we see that the uncertainty vs. time data can be fitted with a power function. Two power functions are shown in Figure 5. For an ideal counting uncertainty curve, the exponent for the power function should be at 0.5. This is shown as the blue curves in the figure. Since FRAM uncertainty calculations are very complicated, we have found that the actual exponents are often between 0.25 and 0.5.

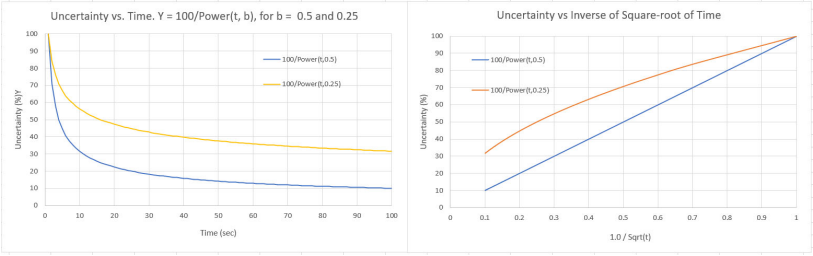

Figure 5.Two power functions. (a) X-axis is time (t). (b) X-axis is 1/sqrt(t)

Asymptotic Uncertainty and Stop Time

The trend of uncertainty over time can be presented well with a power function, as shown in Figure 4. The rate of uncertainty decreases slowly with time after the initial quick drop. It almost approaches a constant after a large enough live time. Thus, it is possible to terminate the data acquisition earlier without significant

sacrifice in the final uncertainty obtained. To do so, the asymptotic uncertainty is defined as a calculated uncertainty from the power function for a predefined maximum data acquisition time, which is also the live time of the master spectrum. For example, we use 7200 seconds of live time for most recently measured Pu samples. The time interval between consecutive data points was fixed at 10 seconds except for the Pu-238 samples.

Curve Fitting for Uncertainty Over Time

The uncertainty over time is modeled by the following simple equation:

$$f_{unc} = h \cdot t^b \qquad (1)$$

where t is the live time, h is the height for the uncertainty, and b is the power exponent. Taking the natural logarithm of both sides:

$$Ln(f_{unc}) = Ln(h) + b \cdot Ln(t) \qquad (2)$$

Variable substitutions used in Eq. (2) are: $y \equiv Ln(f_{unc}),\ x \equiv Ln(t),\ and\ a \equiv Ln(h)$. It is easy to see that the result is a linear equation:

$$y = a + b \cdot x \qquad (3)$$

For linear-fit of Eq. (3), the linear-correlation coefficient is calculated as: [4]

$$r \equiv \frac{(N\sum x_i y_i - \sum x_i \sum y_i)}{\sqrt{N\sum y_i^2 - (\sum y_i)^2}\sqrt{\sum x_i^2 - (\sum x_i)^2}} \qquad (4)$$

The value range for r is from -1 to 1. If it is 0, there is no correlation among the data points at all. For complete correlation and complete anti-correlation, the values are 1 and -1 respectively. A probability distribution for r can be derived. For the null hypothesis test, the probability that any random sample of uncorrelated data would yield a linear-correlation coefficient equal to r is given by: [4]

$$P_r(r;\upsilon) = \frac{1}{\sqrt{\pi}}\frac{\Gamma\left[\frac{(\upsilon+1)}{2}\right]}{\Gamma\left(\frac{\upsilon}{2}\right)}(1-r^2)^{(\upsilon-2)/2} \qquad (5)$$

Where $v = N - 2$ is the degrees of freedom and Γ is the gamma function. The two-sided cumulative probability for having a linear-correlation coefficient as large as r, and larger than r, is given by: [4]

$$P_c(r;N) = 2\int_r^1 P_\tau(r;\upsilon)\, d\tau \qquad (6)$$

The factor of 2 in Eq. (6) is due to the two tails of the distribution. A small value is for the case that the observed data are highly correlated. Thus, for all the results we presented, a value of 1% has been used. The equation cannot be integrated analytically, thus requiring numerical integration by Simpson's rule. If the correlation probability is larger than 1%, the fitted curve is ignored. This helps to ensure the quality of the asymptotic uncertainty calculated.

Results

For all the data analyzed, a stop-criteria between 0.1% and 0.5% was used. For many Pu samples, uncertainties of the specific power are often analyzed to search for a stop time. It is possible to analyze the trend based on the uncertainties of Pu-238, Pu-239, and effective Pu-240 fractions. In addition, the uncertainties of U-235 can be analyzed as well.

Format of the Tables

Analysis results are presented in the following tables. The live time of the master spectra can be found in the table caption if identical for all samples. Otherwise, the live times are shown in the table. The stop-criteria is also in the caption. Sample information can be found in the first two columns. The stop times are listed under 'Live Time @ Stop'. The calculated uncertainty corresponding to the stop time is listed in the next column. The asymptotic uncertainty calculated is listed under the 'Asymptotic Uncertainty' column. In the next column, the FRAM uncertainty from analyzing the master spectrum is shown. In the last column, the difference between the calculated asymptotic uncertainty and the FRAM uncertainty can be found. The relative difference is the difference shown in the last column divided by the FRAM uncertainty. The relative difference is shown as a percentage in the table caption. It is related to the accuracy of the asymptotic uncertainty calculated. Smaller differences indicate better results.

Results for Pu Samples with Different Total Masses

In Table 1, the total Pu masses are from 20 to 874 grams and the live time is 7200 seconds for all samples. For the first Pu sample (S-3) in Table 1, the stop time found was 3710 seconds. If the data acquisition is terminated at a live time of 3710 seconds, the uncertainty of the specific power is as low as 0.47%. If data acquisition continues to the maximum allowed time of 7200 seconds, the final uncertainty would be only about 0.1% smaller. The difference between the stop time and the maximum data acquisition time is the time saved. For this sample, the time saved is 3490 seconds, almost an hour of time saved. Similar results were found for the other three Pu samples in the table. Furthermore, the differences between the asymptotic uncertainty and those from analyzing the master spectra were all less than 0.1%. Thus, the fitted trend curves are good representations of the true uncertainty trend. For all samples, data acquisition could be stopped much earlier on instead of at the maximum allowed data acquisition time of 7200 seconds. The simple search process for a stop time works well for these Pu samples with a wide range of total Pu masses (from 20 to 874 grams).

Table 1. Samples with different Pu masses. Live time: 7200s, stop-criteria: 0.1%, averaged relative difference: 15%.

Sample Information		Curve Fit for Specific Power		Asymptotic Uncertainty for Specific Power		FRAM Uncertainty & Asymptotic Uncertainty
Sample	Total Pu (g)	Live Time @ Stop (s)	Stop Uncertainty (%)	Asymptotic Uncertainty (%)	FRAM Uncertainty (%)	Absolute Difference (%)
S-3	20	3710	0.47	0.38	0.46	0.08
S-4	30	3290	0.45	0.35	0.43	0.08
S-1	400	4290	0.58	0.48	0.44	0.04
S-2	874	4190	0.66	0.57	0.67	0.1

It is interesting to note that the stop times found for Table 1 samples do not show correlation with the total Pu mass. When the samples were measured, the sample-to-detector distance was adjusted to limit the dead time. So, the total mass of Pu is not the only parameter changed from sample to sample. However, no matter how the samples were measured, stop times could be found without any changes to the search process.

Results for Two MOX Samples

Results for two uranium-plutonium mixed oxide (MOX) samples are listed in Table 2. The live time is 7200 seconds for all samples. The U/Pu ratios for the samples are 2.5 and 0.25 respectively. Since it is highly

enriched uranium, those ratios are about the same as the U-235/Pu ratios. The stop times found are longer than those for the corresponding Pu samples in Table 1. The presence of U-235 gamma rays could make the Pu analysis more complicated, leading to larger final uncertainties for the specific power. Thus, it takes longer for the uncertainties to decrease to a precision of 0.1%.

For the MOX samples, the trend of U-235 uncertainty over time could be used to find a stop time as well. The results are 4730 seconds and 4940 seconds respectively for the two samples. Sample S-7 has more U-235 mixed with Pu. Thus, the stop time found based on U-235 is shorter than that for sample S-8. However, if the specific power is analyzed, the stop time is longer for S-7 than that for S-8, as shown in the table below.

Table 2. MOX samples with known U-235/Pu ratios. Live time: 7200s, stop-criteria: 0.1%, averaged relative difference: 12%.

Sample Information		Curve Fit for Specific Power		Asymptotic Uncertainty for Specific Power		FRAM Uncertainty & Uncertainty @ Stop
Sample	U-235/Pu	Live Time @ Stop (s)	Uncertainty @ Stop (%)	Asymptotic Uncertainty (%)	FRAM Uncertainty (%)	Absolute Difference (%)
S-7	2.5	5050	0.7	0.6	0.54	0.06
S-8	0.25	4590	0.64	0.54	0.58	0.04

Results for Pu Samples with Different Pu-239 Fractions

Results for four Pu samples with Pu-239 fractions from 64.8% to 93.7% can be found in Table 3. The Pu-240 fractions are listed in the table since they are more relevant to the specific power than the Pu-239 fractions. The total Pu mass is 6 grams for all samples. Those spectra were acquired with a planar detector, and the spectra can be found in the data folder after FRAM version 6.1 software has been installed. The real time preset was at 1800 seconds. So, the live times were different for those sample. The ADC gain was set at 75 eV per channel. For all the stop times found, the differences in the last column are also well within 0.1%. From the results in the table, it seems that the stop times found do not depend strongly on the Pu-239 fractions.

Table 3. Samples with Pu-239 fractions from 64% to 93%. Stop-criteria: 0.1%, averaged relative difference: 7.3%.

Sample Information		Curve Fit for Specific Power		Asymptotic Uncertainty for Specific Power			FRAM Uncertainty & Uncertainty @ Stop
Sample	Pu-240 Fraction (%)	Live Time @ Stop (s)	Uncertainty @ Stop (%)	Live Time (s)	Asymptotic Uncertainty (%)	FRAM Uncertainty (%)	Absolute Difference (%)
P-1	26.3	680	0.93	989	0.84	0.92	0.08
P-2	18.8	660	0.78	993	0.69	0.76	0.07
P-3	14.3	670	0.51	1132	0.42	0.45	0.03
P-4	6.3	780	0.52	1232	0.42	0.44	0.02

Pyrochemical Processing Samples

Samples discussed so far are all certified Pu standards. To test the uncertainty trend analysis with non-standard samples, several metal and residue samples from pyrochemical processing were measured and analyzed. The results are shown in Table 4. The live times were all at 7200 seconds. As seen from the table,

stop times much shorter than 7200 seconds were found for all samples, even with a stop-criteria at 0.1%. For the last two samples (Residue-1 and Residue-2), the time saved is about 50%. The asymptotic uncertainties are small because the count rates are high for the Pu-239 peaks. That is likely due to less attenuation for the residues. The final differences are well within the stop-criteria of 0.1% for the last two samples as well. The differences are higher (about 0.2%) for the first two samples with Pu metals. The asymptotic uncertainties are also higher, likely because of strong attenuation of Pu-239 peaks by the metals. Nevertheless, stop times can be found for unknown samples, just as for the standards.

Table 4. Pyrochemical processing samples and residues. Live time: 7200s, stop-criteria: 0.1%, averaged relative difference: 15%.

Sample Information		Curve Fit for Specific Power		Asymptotic Uncertainty for Specific Power		FRAM Uncertainty & Asymptotic Uncertainty
Sample	Pu-239 Fraction (%)	Live Time @ Stop (s)	Stop Uncertainty (%)	Asymptotic Uncertainty (%)	FRAM Uncertainty (%)	Absolute Difference (%)
Metal-1	93	5520	1.04	0.94	1.17	0.23
Metal-2	93	4680	0.63	0.53	0.73	0.2
Metal-3	93	4570	0.59	0.49	0.52	0.03
Residue-1	93	3690	0.48	0.38	0.45	0.07
Residue-2	93	3560	0.67	0.58	0.63	0.05

For the results presented in the four tables discussed so far, stop times were found under various measurement conditions. Furthermore, the stop times were found with the same set of input parameters. In an automated analysis of a large number of samples, it is important to have a process which is simple to setup and works under various conditions. If the set-up is too complicated and time-consuming, then it will not be efficient.

Results from the Uncertainty Trend of Effective Pu-240

For Pu samples, the effective Pu-240 is also often a quantity of interest. It is used together with neutron measurements to determine the total amount of Pu. The set of Pu samples with different masses shown in Table 1 were used to obtain the results in the table below. Stop times were found for all samples, as shown in the table, with a stop-criteria of 0.5%. Although the uncertainties are larger, the absolute differences are still less than about 20% of the FRAM uncertainties reported.

Table 5. Results for Effective Pu-240. Live time: 7200s, stop-criteria: 0.5%, averaged relative difference: 17%.

Sample Information		Curve Fit for Effective Pu240		Asymptotic Uncertainty for Effective Pu-240		FRAM Uncertainty & Asymptotic Uncertainty
Sample	Total Pu (g)	Live Time @ Stop (s)	Stop Uncertainty (%)	Asymptotic Uncertainty (%)	FRAM Uncertainty (%)	Absolute Difference (%)
S-3	20	3560	2.14	1.64	2.04	0.4
S-4	30	3100	1.95	1.46	1.84	0.38
S-1	400	4520	3.1	2.61	2.29	0.32
S-2	874	3400	1.95	1.46	1.7	0.24

In Figure 6, how the stop time varies with the stop-criteria is shown for sample S-1. On the graph, when the stop-criteria is 0.5%, the stop time for sample S-1 is 4520 seconds, as shown in Table 5. As expected, the higher the stop-criteria, the shorter the stop time found.

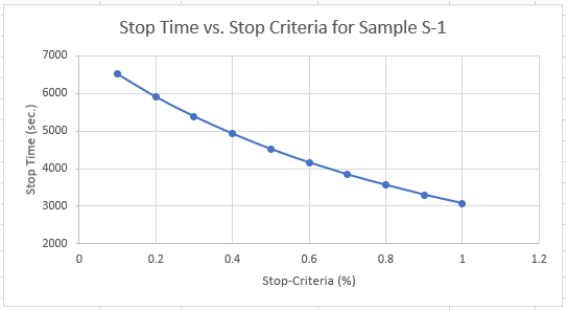

Figure 6. Stop time vs. stop-criteria for sample S-1. The higher the stop-criteria, the shorter the stop time found.

Results for Five Uranium Standards with Different Enrichment

For uranium samples, the trend of U-235 uncertainty could be used to search for the stop time. Results for five uranium standards with different amounts of U-235 can be found in Table 6. The total weight for all standards is 200 grams. Sample U-1 is depleted uranium (DU), sample U-2 is natural uranium and the other three are low-enriched uranium samples (up to 4.46%). The live times for the master spectra are listed in the table. A stop time was found for all samples with a stop-criteria of 0.1%. For this group of samples, the stop time is smaller for samples with more U-235. Such a trend is not present for the Pu samples shown in other tables. Due to the relatively large uncertainties in the U-235 results, the stop times found are all not that far from the maximum data acquisition time.

Table 6. Uranium standards with different enrichment. Stop-criteria: 0.1%, averaged relative difference: 9.3%.

Sample Information		**Curve Fit for U-235**		**Asymptotic Uncertainty for U-235**			**FRAM Uncertainty & Asymptotic Uncertainty**
Sample	Enrichment (%)	Live Time @ Stop (s)	Uncertainty @ Stop (%)	Live Time (s)	Asymptotic Uncertainty (%)	FRAM Uncertainty (%)	Absolute Difference (%)
U-1	0.31	2980	2.91	3265	2.82	2.93	0.11
U-2	0.71	2830	1.89	3257	1.8	1.98	0.18
U-3	1.94	2670	1.42	3245	1.32	1.49	0.17
U-4	2.95	2590	1.43	3186	1.34	1.5	0.16
U-5	4.46	2520	1.33	3145	1.23	1.39	0.16

Results for Two Pu-238 Samples

Finally, results for two Pu-238 samples can be found in Table 7. The Pu-238 fractions are close to 80% for both. The live times were set to 14400 seconds for both samples (twice the live time for a lot of other samples). In the last column, the differences between the asymptotic uncertainty and the FRAM uncertainty are the largest among all the samples we have investigated. Thus, the stop-criteria used for the trend analysis was set to 0.5%. The large uncertainties are due to high background from Pu-238.

Table 7. Two Pu-238 standards. Live time: 14400s, stop-criteria: 0.5%, averaged relative difference: 25%.

Sample Information		Curve Fit for Specific Power		Asymptotic Uncertainty for Specific Power		FRAM Uncertainty & Asymptotic Uncertainty
Sample	Pu-238 (%)	Live Time @ Stop (s)	Stop Uncertainty (%)	Asymptotic Uncertainty (%)	FRAM Uncertainty (%)	Absolute Difference (%)
S-5	78.44	10040	2.82	2.33	1.69	0.64
S-6	80.29	10800	5.5	5.32	6.11	0.79

Summary

The trend of uncertainty over time has been studied for plutonium, uranium, MOX, pyrochemical processing and other types of samples. All the uncertainty data have been fitted with a simple power function and the function is a good representation of the actual uncertainty trend. With a stop-criteria between 0.1% to 0.5%, some count times have been reduced by up to 50%. If a stop-criteria larger than 0.5% is used, the stop time would be even shorter. The asymptotic uncertainties calculated at the maximum allowed data acquisition times match well with the actual results from analyzing the master spectra. The difference between the asymptotic uncertainty and the actual uncertainty is often close to the stop-criteria used. In addition, the corresponding relative difference is found to be less than 20% for most of the measurements performed. The method we investigated is automated and works well for samples of different types and under various measurement conditions. It is also easy to setup, with the stop-criteria as the only key input parameter needed. Thus, we believe the method could be used in other types of measurements as well.

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