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PLUTONIUM ISOTOPIC ANALYSIS IN THE 30 keV TO 210 keV RANGE

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Abstract

Low-Energy Gamma-ray Spectroscopy (LEGS) is a nondestructive assay (NDA) technique developed in the 1980s. In 1999, it was modified to include a physical-based model for the energy dependent efficiency. It uses the gamma rays in the energy range from approximately 30 keV to 210 keV, except the 100-keV region. This energy region provides intense, well-separated gamma rays from the principal isotopes of plutonium. For applications involving small quantities (mg to g) of freshly separated plutonium in various chemical forms, it is ideally suited for accurate real-time or near real-time isotopic analysis. Since the last modification, LEGS has been incorporated into the FRAM code (Fixed-energy Response-function Analysis with Multiple efficiency), version 4. FRAM v4 is capable of analyzing the peaks in the whole energy range from 30 keV to 1 MeV, including the X-ray region. The new capability of analyzing the peaks in the 100-keV region greatly enhances the plutonium analysis in the 30 keV to 210 keV ranges of the traditional LEGS. We now can analyze both the freshly separated and aged plutonium with greater accuracy.

I. INTRODUCTION

Many nondestructive gamma-ray techniques have been developed to determine plutonium isotopic ratios. In all of those techniques, the determination of the relative efficiency of the data points at different energies is a fundamental part of the analysis. In some methods, the efficiency is interpolated or extrapolated with simple linear *ln*(eff) vs. *ln*(E) or quadratic methods between a small number of relative efficiency points.[1] In some other methods, the relative efficiency data points are empirically fitted to a polynomial function in log of energy.[2,3] A third method uses the knowledge of the physical processes involved in the relative efficiency curve (detector efficiency, external absorbers, and plutonium self-absorption) to fit the relative efficiency data.[4]

Different techniques use different methods and energy regions to analyze the data. The traditional LEGS,[1] developed at Los Alamos National Laboratory (LANL), uses close-lying gamma-ray pairs in the ranges from 38 keV to 65 keV and 120 keV to 208 keV in the determination of the plutonium isotopic ratios. The current LEGS[5] uses a physical-based efficiency curve to fit the peaks in the ranges of the traditional LEGS. The Multi-Group Analysis (MGA),[6] developed at Lawrence Livermore National Laboratory (LLNL), employs a physical-based efficiency curve to determine the plutonium isotopic ratios in the energy range from 94 keV to 208 keV. The Fixed-energy Response-function Analysis with Multiple efficiency (FRAM),[3] version 3.x or earlier, developed at LANL, uses the empirical efficiency curve method in the energy range from 120 keV up to 1 MeV for the plutonium isotopic analysis. Each different method has some advantage and disadvantage compared with the other methods. Depending on the application, one may choose one method over the others.

For a very small quantity of freshly separated plutonium (μg to mg), LEGS appears to be the appropriate choice. The 40-keV region contains peaks of most isotopes in plutonium samples (except ^{241}Pu), and those peaks represent the most intense gamma rays for each of the respective isotopes. However, if too much ^{241}Am is present (such as in aged plutonium), its 60-keV gamma ray will overwhelm all other peaks in the region, making this region useless for isotopic measurements.

For samples inside thin containers (freshly separated or aged), MGA's use of the 100-keV region usually gives better measurement precision than other codes. The 100-keV region contains the second most intense group of gamma rays for the isotopes in plutonium. This region is also the most complex and difficult to analyze of all the regions of the gamma-ray spectrum of plutonium.

For samples inside thick or lead-lined containers, FRAM is probably the choice. Most low-energy gamma rays will be absorbed by the thick container that will prevent the use of LEGS or MGA. FRAM has demonstrated analysis over a wider range of attenuations than any other published data. It has been shown to analyze the plutonium isotopic from plutonium inside 25-mm-thick walls of lead.[7]

The new version of FRAM (v4) added the capability of analyzing the X-ray peaks and using the physical-based efficiency curve in addition to the empirical efficiency curve of the previous versions. It is now capable to analyze plutonium and uranium (or the combination of both plutonium and uranium) in the whole energy range from 30 keV to 1 MeV.

With this new capability, FRAM now can be used to determine the isotopic composition of plutonium and uranium in most different measurement conditions and situations mentioned above, from freshly separated to aged samples, and from bare source to source inside a thick wall.

II. IMPROVEMENT OF FRAM v4

Many new and unique features have been added to FRAM v4 in response to new measurement requirements and to meet user needs and requests. Some of those improvements are mentioned in reference 8. Some improvements are worth mentioning here but will not be described in detail:

- Automated parameter file selection to speed the measurement process for robotic applications or applications for samples with largely unknown contents.
- Separated engine structure and user interface making it easier to accomplish derivative applications.
- Uranium analysis enhancements, which include the corrections for the loss of peak areas due to summing, the ^{236}U correlation prediction, and the decay correction for non-equilibrium ^{238}U/^{234}Th.
- Capability of isotopic analysis data taken with the Peltier-cooled CdTe detectors.[9]
- Interfacing with the Canberra's Genie-2000 programming library in addition to the Canberra S100 system and various Ortec Multi-Channel Buffers (MCB) which already existed in version 3.x and earlier.

The two improvements to be discussed in this paper are the new physical-based efficiency curve and the fitting of the X-ray peaks.

A. New Relative Efficiency Curve Option

All versions 3.x and earlier FRAM used an empirical relative efficiency curve first proposed by Fleissner.[2]

$$ln(\text{Area/BR}) = c_1 + c_2/E_2 + c_3(lnE) + c_4(lnE)^2 + c_5(lnE)^3 + c_i + c_j/E,$$

where E is the energy in MeV; c_i is associated with additional isotopes beyond the first one, and each c_j is associated with an efficiency function beyond the first one.

This empirical relative-efficiency curve has been very successful for many measurement situations. However, its empirical nature and polynomial structure make it behave unphysically in some situations, notably when extrapolated outside its range of definition or when used with very weak data.

In FRAM v4, we added new efficiency curve formalism based on the physical properties of the analyzed material and surrounding materials. The new efficiency curve is constructed as

$$\text{Area/BR} = \left[\frac{1-e^{-\mu_{Pu}x_{Pu}}}{\mu_{Pu}x_{Pu}}\right] * \left[e^{-\mu_1 x_1} e^{-\mu_2 x_2} e^{-\mu_3 x_3}\right] * [I_i] * \left[e^{c_j/E}\right] * [\text{Det eff}] * [\text{Correction factor}]$$

where the term inside the first square bracket associates with the U/Pu attenuation; the term inside the second square bracket associates with the attenuation due to the absorbers (up to three different absorbers can be used); I_i is associated with the activity of the isotope i; c_j associates with an efficiency function beyond the first one; "Det eff" is a generic detector efficiency parameterized in the software; and "Correction factor" is to correct for the detector efficiency and the attenuation of the measured materials and the absorbers.

This formula is very much the same as the one in the widely used MGA. (The MGA code was the first to use a physics-based model for the relative efficiency). The factor that makes this different from the MGA is the "correction factor," where in the MGA is expressed as a quadratic ($1 + bE + cE^2$). In our formula we use the modified Hoerl formula ($E^b * c^{1/E}$) where E is the peak energy and b and c are some variables.

The advantage of using the Hoerl equation for the correction factor is that all the individual deviations can be corrected and all those corrections can be combined together and still retain the Hoerl form

$$\left(E^{b_1} c_1^{1/E}\right)\left(E^{b_2} c_2^{1/E}\right) = E^{(b_1+b_2)}\left(c_1 c_2\right)^{1/E} = E^b c^{1/E},$$

where the leftmost side of the equation shows the individual corrections (such as detector efficiency and

self-attenuation in the solution, etc.), and the rightmost side of the equation shows the combined correction where

$$b = b_1 + b_2 \text{ and } c = c_1 c_2 .$$

The detector efficiencies are parameterized in the software. FRAM v4 can analyze data taken with three different types of detectors: high-purity germanium (HPGe) planar, HPGe coaxial, and Peltier-cooled CdTe detectors. The user can select the detector type in the data analysis dialog box.

The "analyze plutonium data" dialog box in Fig. 1 shows the options such as ^{242}Pu correlation, auto analysis, print out the results, relative efficiency model, etc. that the user can enter. If the physical-efficiency model is chosen, the user can also choose the detector type and set ranges of the material and absorbers' thickness. Up to three different types of absorbers at all different ranges can be set. There are eight different types of parameterized absorbers in the code: aluminum, steel, cadmium, erbium, lead, uranium, water, and concrete. The elemental absorbers were chosen to about evenly cover the range of all different material types. If a different type of absorber not in the list is used in the data acquisition, one may choose the one closest to it from the list for the analysis. As an example, one may choose steel to substitute for copper or cadmium for tin, etc. The two compound absorbers, water and concrete, are chosen to represent the most common compound absorbers in field measurements.

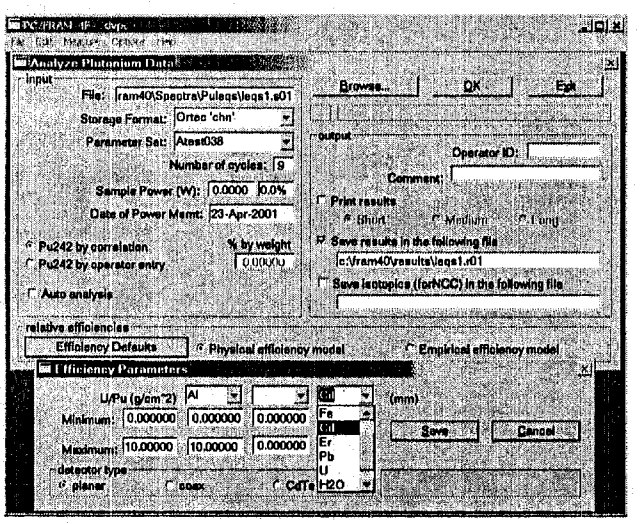

Figure 1. FRAM's plutonium analysis dialog box.

B. Fitting The X-Ray Peaks

FRAM fits a background-subtracted gamma-ray peak with a Gaussian function plus exponential tailing function as

$$Y_i = Y_o exp[\alpha(x_i - x_o)^2] + \text{Tail}(x_i)$$

where

Y_i = the net counts in channel x_i,
Y_o = the peak height at centroid x_o,
α = -4 $ln2$/FWHM^2, is the peak-width parameter.

The tailing parameter is given by

$$\text{Tail}(x_i) = Y_o exp[(T_1 + T_2 E) + (T_3 + T_4 E)(x_i - x_o)] \\ [1 - exp(0.4\alpha(x_i - x_o)^2)],$$

where E is the energy of the peak and T_i are constants that determine the shape of the tail.

These equations can fit the gamma-ray peaks well but would not accurately fit the X-ray peaks. The intrinsic X-ray peaks are Lorentzian distributed. When this energy distribution of radiations is convoluted with the instrumental dispersion, which is the Gaussian plus exponential tailing, the resulting peak shape is very different from that of an equivalent energy gamma ray.

To fit the X-ray peaks, we fit the Voigtian line shape, which is a convolution of the Gaussian and Lorentzian distribution, plus the exponential tailing function. The tailing function is the same as that of the gamma-ray peaks described above. As for the Voigt function, fitting it directly would consume enormous computing power and be very inefficient. Instead, we adopted the approximation employed by Czosnyka and Trzcinska.[10] The authors claimed that the accuracy of this approximation of the Voigtian line shape is better than 0.001, which is sufficient for X-ray peak fitting.

Figure 2 shows an example of the peak fitting in the X-ray region.

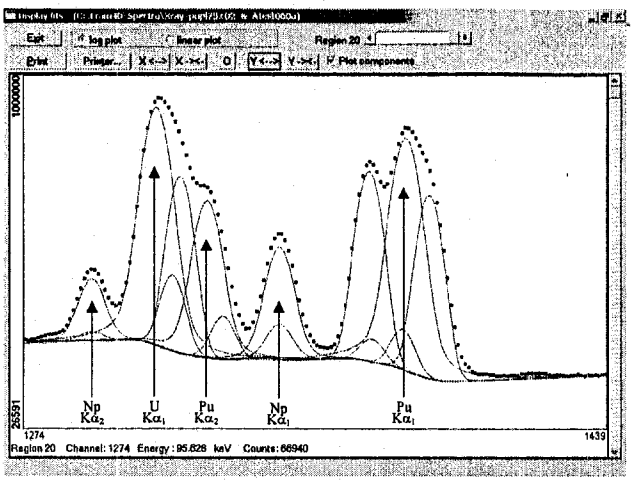

Figure 2. An example of the peak fitting in the 96- to 104-keV region. The scale is logarithm. Neptunium X-rays are from the decay of ^{241}Pu-^{237}U and ^{241}Am. Uranium X-rays are from the decay of all the plutonium isotopes. Plutonium X-rays are from alpha- and gamma-ray-induced fluorescence.

III. ANALYSIS

As mentioned in the introduction, FRAM v4 can analyze peaks in the range from 30 keV to 1 MeV. The user can use any peak in this energy range for the determination of the efficiency and activity of the isotopes. In theory, one can use all the peaks in this wide energy range to analyze the data. In practice, we found that the results from the analysis of a very wide range, even though it employs more data and peaks, may not be better than the analysis in a smaller energy range. The main reason is that when the peak energy range is very large, the deviations in the efficiency curve may become large and the "correction factor," used to correct for small deviations in the efficiency, may not be able to correct them.

A. Freshly Separated Plutonium

For the freshly separated samples, all the peaks above 30 keV can be used for analysis. The peaks in the 38- to 65-keV range represent the most intense gamma rays for most of the plutonium isotopes, except ^{241}Pu. It, therefore, is not sufficient to use this region alone for plutonium isotopic analysis. This region will need to be used together with another region that has a ^{241}Pu peak.

The X-ray region, from about 90 keV to 104 keV, contains intense peaks from all the plutonium isotopes except ^{239}Pu. Pu-239 actually has one fairly intense peak at 98.8 keV. However, it is sitting on the shoulder of a much more intense uranium X-ray peak and its area cannot be accurately extracted so it is not used. As in the 50-keV region, we cannot use this region alone in obtaining all the isotopic information and have to combine it with another region.

The region above the X-ray region contains peaks from all the plutonium isotopes so by this region alone, we can determine the isotopic ratios of all the plutonium isotopes. However, the gamma rays in this region are about two and one orders of magnitude less intense than the peaks in the 50-keV and X-ray regions, respectively. It is best if used together with another region in the determination of the plutonium isotopic.

We created three different FRAM parameter files at three different energy ranges to use with these samples. One with energy from 38 to 106 keV, one from 38 to 208 keV, and one from 60 to 208 keV.

Six high-burnup samples are analyzed with FRAM v4 using these three parameter files. The results, together with those from mass spectrometry, are shown in Table I. Assuming the results from mass spectrometry are most accurate, the results from other methods are then divided by the results from mass spectrometry for easy comparison. Because there are no measurable gamma rays from ^{242}Pu, FRAM cannot accurately determine ^{242}Pu. Therefore, the ^{242}Pu values are taken from mass spectrometry for use in the normalization of other isotopes.

The results of ^{239}Pu, ^{240}Pu, and ^{241}Pu from all three parameter files at three different energy ranges appear to agree well with the mass spectrometry results. The ^{238}Pu results don't appear to match the values from alpha counting very well. The ^{238}Pu values determined from the traditional LEGS method also have similar bias when comparing to the alpha counting values. We believe the ^{238}Pu values determined from the alpha counting have large bias, which leads to large differences between the alpha counting and the gamma-ray measurements.

On close examination of the results, we see that the parameter file with energy ranges from 38 to 208 keV gives the best results. Next comes the 38-104-keV parameter file. The 60-208-keV parameter file is least accurate of the three.

Table I. Comparison of the results of the plutonium analysis by various methods.

Sample	Method/ Parameter	Isotopic Percent				Isotopic Ratio to Mass Spec.			
		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu
1	Mass Spec	0.691	66.616	22.694	7.101				
	38keV-208keV	0.732	66.645	22.687	7.038	1.0591	1.0004	0.9997	0.9911
	38keV-104keV	0.728	66.773	22.625	6.976	1.0540	1.0024	0.9969	0.9824
	60keV-208keV	0.729	66.610	22.732	7.031	1.0551	0.9999	1.0017	0.9901
2	Mass Spec	0.525	69.682	21.793	5.797				
	38keV-208keV	0.498	69.695	21.827	5.776	0.9490	1.0002	1.0016	0.9964
	38keV-104keV	0.500	69.564	21.935	5.798	0.9519	0.9983	1.0065	1.0001
	60keV-208keV	0.493	69.655	21.894	5.755	0.9382	0.9996	1.0047	0.9928
3	Mass Spec	0.510	70.220	21.211	5.915				
	38keV-208keV	0.473	70.325	21.171	5.887	0.9272	1.0015	0.9981	0.9953
	38keV-104keV	0.474	70.227	21.290	5.865	0.9293	1.0001	1.0037	0.9915
	60keV-208keV	0.466	70.349	21.188	5.853	0.9134	1.0018	0.9989	0.9894
4	Mass Spec	1.246	58.455	26.498	8.900				
	38keV-208keV	1.252	58.478	26.512	8.856	1.0050	1.0004	1.0005	0.9951
	38keV-104keV	1.256	58.413	26.596	8.833	1.0081	0.9993	1.0037	0.9925
	60keV-208keV	1.244	58.509	26.532	8.813	0.9985	1.0009	1.0013	0.9903
5	Mass Spec	0.353	70.756	23.158	3.981				
	38keV-208keV	0.339	70.725	23.208	3.976	0.9610	0.9996	1.0021	0.9987
	38keV-104keV	0.342	70.545	23.370	3.991	0.9681	0.9970	1.0092	1.0024
	60keV-208keV	0.338	70.439	23.469	4.003	0.9563	0.9955	1.0134	1.0054
6	Mass Spec	0.756	59.950	28.015	7.375				
	38keV-208keV	0.760	59.977	27.999	7.359	1.0054	1.0005	0.9994	0.9979
	38keV-104keV	0.766	59.891	28.106	7.334	1.0128	0.9990	1.0032	0.9944
	60keV-208keV	0.754	60.048	27.961	7.332	0.9973	1.0016	0.9981	0.9942

B. Aged Plutonium

For aged material, the 60-keV peak of ^{241}Am is very intense, and its corresponding Compton distribution would overwhelm all other peaks below it, making the region below 60 keV useless for isotopic measurements. Therefore, for aged plutonium, we can only use the gamma rays in the X-ray region and the region above it. One advantage of using aged material is that the ^{237}U has already become secular equilibrium so its gamma rays can be considered to decay directly from ^{241}Pu. This would increase the accuracy of the ^{241}Pu determination.

An HPGe planar detector was used to take a series of spectra of the well-known CBNM standard sources. Each spectrum was 5 minutes and 24 spectra for each of the four sources.

We created three different FRAM parameter files at three different energy ranges to use with these samples: one with energy from 94 to 208 keV, one from 94 to 414 keV, and one from 125 to 414 keV. Table II shows the average results from each source divided by the accepted value.

IV. DISCUSSION

FRAM allows the user to modify a parameter file for specific measurement conditions without any time-consuming, labor-intensive main code changes. With this capability, the parameters can be modified to include ^{235}U, ^{238}U, ^{239}Np, etc. or any isotopes mixed in with plutonium. For some difficult measurements such as samples containing impurity or inside a thick wall container, the user can either use a different parameter file suitable for

* The ^{238}Pu values for Mass Spectroscopy in the table are actually determined from alpha counting.

Table II. Average results from each source divided by the accepted values.

Source/ %239Pu	Parameter/ Energy		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Specific Power	^{240}Pu Effective
CBNM93 93.5%	94 - 414	Ave	0.9525	1.0015	0.9775	0.9989	0.9747	0.9936	0.9777
		STD	0.0899	0.0008	0.0125	0.0076	0.0115	0.0028	0.0124
	94 - 208	Ave	0.9284	0.9999	1.0018	0.9965	0.9899	0.9976	1.0015
		STD	0.0888	0.0015	0.0221	0.0073	0.0191	0.0047	0.0218
	125 - 414	Ave	0.9942	1.0003	0.9960	0.9954	0.9967	0.9990	0.9961
		STD	0.1006	0.0025	0.0363	0.0075	0.0240	0.0056	0.0358
CBNM84 84.8%	94 - 414	Ave	0.9851	0.9990	1.0056	1.0025	0.9993	0.9995	1.0052
		STD	0.0230	0.0012	0.0072	0.0040	0.0071	0.0030	0.0069
	94 - 208	Ave	0.9873	0.9975	1.0151	1.0002	0.9988	1.0014	1.0142
		STD	0.0241	0.0017	0.0100	0.0055	0.0101	0.0042	0.0096
	125 - 414	Ave	0.9934	0.9988	1.0072	0.9988	1.0027	1.0013	1.0068
		STD	0.0358	0.0022	0.0134	0.0065	0.0140	0.0048	0.0127
CBNM70 75.5%	94 - 414	Ave	1.0034	1.0005	0.9980	1.0001	1.0059	1.0034	0.9987
		STD	0.0109	0.0040	0.0165	0.0064	0.0088	0.0062	0.0127
	94 - 208	Ave	0.9996	0.9997	1.0020	0.9941	1.0000	1.0000	1.0015
		STD	0.0106	0.0044	0.0177	0.0075	0.0077	0.0056	0.0137
	125 - 414	Ave	1.0065	0.9982	1.0062	1.0058	0.9918	0.9998	1.0053
		STD	0.0191	0.0066	0.0275	0.0144	0.0085	0.0065	0.0207
CBNM61 64.8%	94 - 414	Ave	1.0077	1.0017	0.9955	1.0005	1.0118	1.0072	0.9973
		STD	0.0106	0.0049	0.0119	0.0072	0.0071	0.0055	0.0088
	94 - 208	Ave	1.0031	1.0012	0.9974	0.9954	1.0051	1.0029	0.9984
		STD	0.0096	0.0057	0.0139	0.0073	0.0072	0.0055	0.0101
	125 - 414	Ave	1.0117	1.0002	0.9984	1.0055	0.9979	1.0039	0.9997
		STD	0.0137	0.0130	0.0325	0.0123	0.0171	0.0077	0.0232

the measurement or can easily create a new file for the measurement.

Due to the lack of time, we did not properly create good parameter files for these energy ranges. We believe that in a very short time, we can develop much better parameter files and thus, much better results than those in Table II.

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