ANALYSIS OF AMERICIUM-BERYLLIUM NEUTRON SOURCE COMPOSITION USING THE FRAM CODE

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ABSTRACT
The FRAM code was originally developed to analyze high-resolution gamma spectra from plutonium items. Its capabilities have since been expanded to include analysis of uranium spectra. The flexibility of the software also enables a capable spectroscopist to use FRAM to analyze spectra in which neither plutonium nor uranium is present in significant amounts. This paper documents the use of FRAM to determine the ^{239}Pu/^{241}Am, ^{243}Am/^{241}Am, ^{237}Np/^{241}Am, and ^{239}Np/^{241}Am ratios in americium-beryllium neutron sources. The effective specific power of each neutron source was calculated from the ratios determined by FRAM in order to determine the americium mass of each of these neutron sources using calorimetric assay. We will also discuss the use of FRAM for the general case of isotopic analysis of nonplutonium, nonuranium items.

BACKGROUND
There are a multitude of Americium-Beryllium (AmBe) and Plutonium-Beryllium (PuBe) neutron sources throughout the United States that are slated for disposal. The mass of each source must be known before it can be sent for disposal. The Solid State Calorimeter[1] was used to measure the heat produced by a selection of these neutron sources. High-resolution gamma spectroscopy was used to determine the isotopic content of the sources. Combining the heat measurement with the isotopic information enabled calculation of the masses of americium and plutonium in the selected neutron sources. Based on these measurements, the selected neutron sources became secondary working standards for calibration of other nondestructive assay technologies.

Calorimetric assay provides effectively bias-free, high-precision measurement of the neutron sources. Good information about the isotopic content of the items is required; the undetected presence of unexpected heat-producing isotopes in the neutron sources would bias the results. High-resolution gamma-ray spectroscopy was used to nondestructively determine the isotopic content of the neutron sources. Measurements took place between January 2001 and May 2001.

THE ISOTOPES OF INTEREST
For this project, isotopic analysis was being conducted to answer two questions as follows: were there other heat-producing isotopes in the spectrum, and if so, how much of the heat produced by the item was due to those isotopes. The isotopes of interest were therefore restricted to heat-producing isotopes, or those whose gamma rays would interfere with (or appear to be) heat producing isotopes. The heat producing isotopes we included in the analysis were ^{239}Pu, ^{243}Am, ^{237}Np, and ^{239}Np. These isotopes, along with the peaks discussed below, were entered into a FRAM[2,3] parameter set developed for the analysis of the AmBe neutron sources. We also checked for the following isotopes—^{228}Ac, ^{234}Pa, ^{59}Fe, and ^{105}Ru—all of whose gamma rays might interfere with the analysis of the previously mentioned heat-producing isotopes. The measurements were

performed in a location with a ^{137}Cs background, so all spectra included a strong 662keV peak from ^{137}Cs.

^{241}Am produces a wide range of gamma rays in addition to the very abundant 59-keV energy. For this analysis, regions of interest were established to analyze ^{241}Am peaks between 123 keV and 652 keV. The 662-keV peak from ^{241}Am was obscured by the ^{137}Cs peak from area background. ^{243}Am content was evaluated at 142 keV.

Plutonium-239 content was analyzed with the strong 129-keV peak. Additional regions of interest could have been established for other strong peaks (such as 375 keV or 414 keV), but the absence of the 129-keV peak was sufficient to demonstrate that no measurable plutonium was present in the item.

Neptunium-237 was evaluated using 300-keV, 312-keV, 340-keV, and 416-keV peaks. Neptunium-239 was evaluated using 228-keV, 278-keV, and 316-keV peaks.

These peaks were chosen to maximize the absolute branching ratios and minimize the number of potential interferents. The choice of these isotopes of interest and gamma energies meant that several non-heat-producing isotopes could artificially increase the peak areas attributed to the isotopes of interest. These potential interferents included ^{228}Ac, ^{234}Pa, ^{59}Fe, and ^{105}Ru. The spectra were inspected to verify that these isotopes were not evident, using peaks other than those listed for the isotopes of interest.

DATA ACQUISITION

Spectra were acquired using an 8% efficient Canberra germanium detector. The crystal was a 49-mm x 28-mm coaxial crystal, positioned behind a 16-mm x 1-mm planar crystal in a common housing. The planar crystal was not used in these measurements. Data was acquired through an Ortec Dspec spectrometer system controlled by Maestro™ version 5.1 and FRAM version 3.2.

The sources were located inside a hot cell during the measurements. Each source was moved to a dedicated hot cell for measurement and positioned before an opening (approximately 30cm in length) during the measurement. The detector was positioned at the outside of the opening. The distance between source and detector was varied to obtain the desired count rate, but most often the detector was positioned as close to the opening as possible. The crystal housing was positioned roughly parallel to the hot cell wall to allow the gamma rays a direct path to coaxial detector and prevent additional attenuation by the planar detector.

Count rates varied between 200 counts per second and 1100 counts per second. The spectra indicated that the sources were not heavily shielded. The low count rates were due to the distance from the sources and the low efficiency of the detector. Measurement times varied between 2 and 18 hours. All measurements could have been made in less than 8 hours, but when scheduling required overnight measurements, input rates were reduced to prevent unnecessary neutron exposure of the germanium crystals.

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THE DATA

A representative spectrum appears in Fig. 1. This is a 16- hour (true time) spectrum. The input rate was 215 counts per second. As discussed above, the count rate was reduced for this overnight measurement to reduce neutron exposure of the detector.
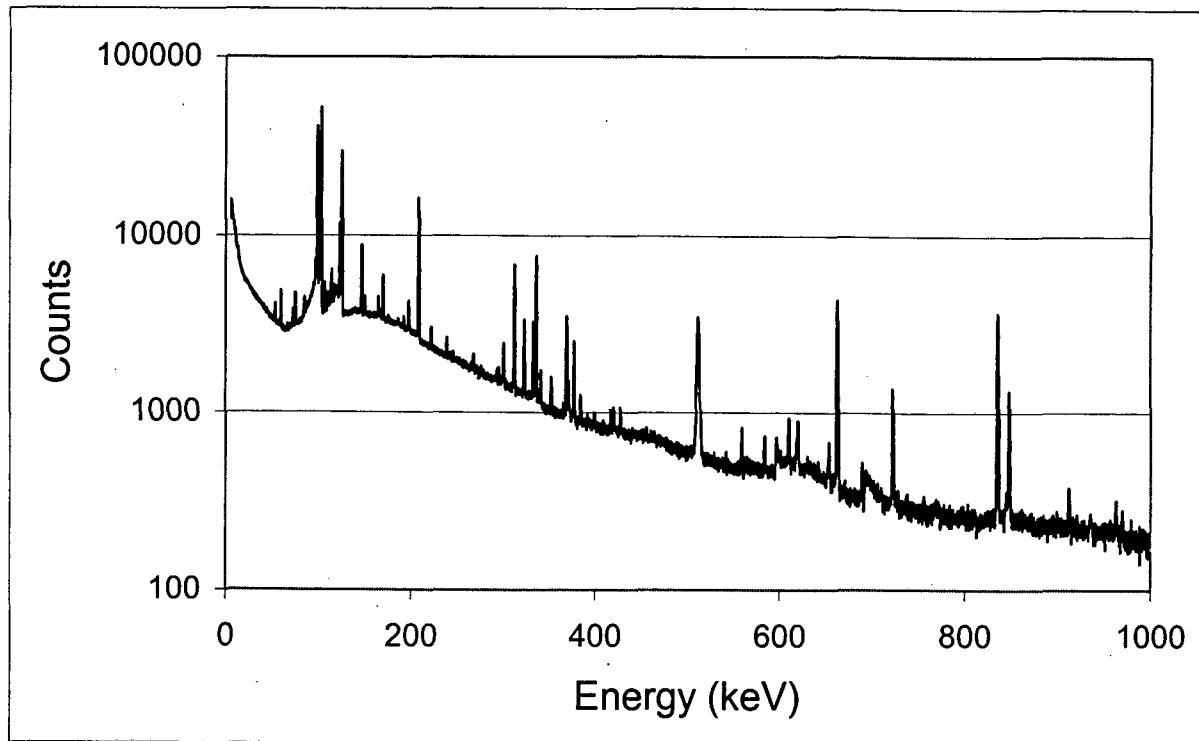
Fig. 1. AmBe neutron source spectrum.

ANALYSIS

A typical FRAM plutonium analysis output is shown in Fig. 2. These results are from FRAM analysis of a spectrum from a 454-g PuO_2 item with approximately 6% ^{240}Pu. The major plutonium isotope fractions are prominently displayed, with effective specific power and effective ^{240}Pu content information appearing at the bottom of the summary.

Figure 3 shows the FRAM output obtained from the analysis of an AmBe source spectrum, using the parameter set created to analyze these sources. The valid output of interest is the list of relative mass values at the bottom of the window; other output in the window should be disregarded because the parameter set did not include the other isotopes of plutonium in the analysis. The other plutonium isotopes were omitted from the parameter set because we neither expected nor detected significant peaks from any plutonium isotope in the spectra.

```
■ Analysis Results                                              _ |□| x|

********************************************************************
PC FRAM (4F)    Isotopic Analysis    14-May-2001 17:10:34
(Fixed energy Response function Analysis with Multiple efficiencies)
System ID: My System

spectrum source:  C:\Program Files\fram40\Calx30.chn
spectrum date:  15-Jan-1995 14:07:24
live time:  2817 s
true time:  3600 s
num channels:  8192

parameter set:  coax8k125.3 (2001.05.02 15:38)
    Coax .125 kev/ch, Homo. Am/Pu, Equ., 3-25% Pu240,<460 keV
********************************************************************
********************************************************************
diagnostics passed.

                                              (By Corr)   (ug/gPu)

            Pu238      Pu239      Pu240      Pu241      Pu242      Am241
mass%     0.00673   93.71608    6.07432    0.17711    0.02576    1976.1
sigma     0.00110    0.03037    0.16516    0.00020    0.00070       6.5
%RSD      16.31%      0.03%      2.72%      0.12%      2.73%      0.33%

%TotPwr:   1.5233   72.0804   17.1551    0.2409    0.0012    8.9991

Specific Power (W/gPu):   (    2.5077 +/-    0.0133)e-003  (   0.53%)

Effective Pu240 fraction:  (    6.1346 +/-    0.1652)e-002  (   2.69%)

Time since chemical separation:     5722.31    +/-   14.05   days   (   0.25%)

********************************************************************
```

Fig. 2. Typical FRAM summary output.

```
■ Analysis Results                                              _ |□| x|

********************************************************************
PC FRAM (4F)    Isotopic Analysis    15-May-2001 07:59:42
(Fixed energy Response function Analysis with Multiple efficiencies)
System ID: My System

spectrum source:  C:\AmBe.Chn
spectrum date:  18-Apr-2001 15:02:19
live time:  57385 s
true time:  57600 s
num channels:  8192

parameter set:  AmBe sources (2001.05.14 16:54)
    Coax .125 kev/ch, 120-800 keV, AmBe source analysis for ca
********************************************************************
********************************************************************
diagnostics passed.

                                              (Default)   (ug/gPu)

            Pu238      Pu239      Pu240      Pu241      Pu242      Am241
mass%     0.00000    0.00000    0.00000    0.00000    0.00000       0.0
sigma     0.00000    0.00000    0.00000    0.00000    0.00000       0.0
%RSD     >99.99%    >99.99%    >99.99%    >99.99%    >99.99%    >99.99%

%TotPwr:   0.0000    0.0000    0.0000    0.0000    0.0000    0.0000

Specific Power (W/gPu):   (    1.0000 +/-    0.0000)e-003  (<   .01%)

Effective Pu240 fraction:  (    0.0000 +/-    0.0000)e-002  (>99.99%)

** cannot compute time since chemical separation!

Relative mass (Np237 / Am241):   4.814939e-002  (   0.58%)
Relative mass (Pl239 / Am241):   5.914710e-002  (  81.10%)
Relative mass (Am243 / Am241):   7.772742e-004  (  67.04%)
Relative mass (Np239 / Am241):   3.601135e-012  (  41.73%)
********************************************************************
```

Fig. 3. FRAM output for AmBe source analysis.

In the AmBe source analysis output, the plutonium fraction columns across the center of the summary output are blank because no plutonium isotopes have been entered as isotopes of interest. The isotopes of interest appear at the bottom of the summary output, reported as ratios to ^{241}Am, the reference isotope. The number in parentheses at the end of each row is the one-standard-deviation uncertainty in the ratio. The uncertainty in the ^{241}Am content can be found in a table in another portion of the output, not shown in Fig. 3.

The "Pl239" isotope listed in Fig. 3 is ^{239}Pu. Mislabeling the isotope in this fashion causes the ratio to be displayed as a ratio to ^{241}Am with the others, instead of showing up in the "Pu239" column in the center of the output window.

The ^{241}Am uncertainties achieved in this investigation ranged from 0.22% to 0.3%.

WEAKNESSES OF THIS APPROACH

The results in Fig. 3 show a ^{239}Pu content of 6%, when it is apparent from visual inspection of the 129keV region that there is no appreciable plutonium in the spectrum (see Fig. 4).
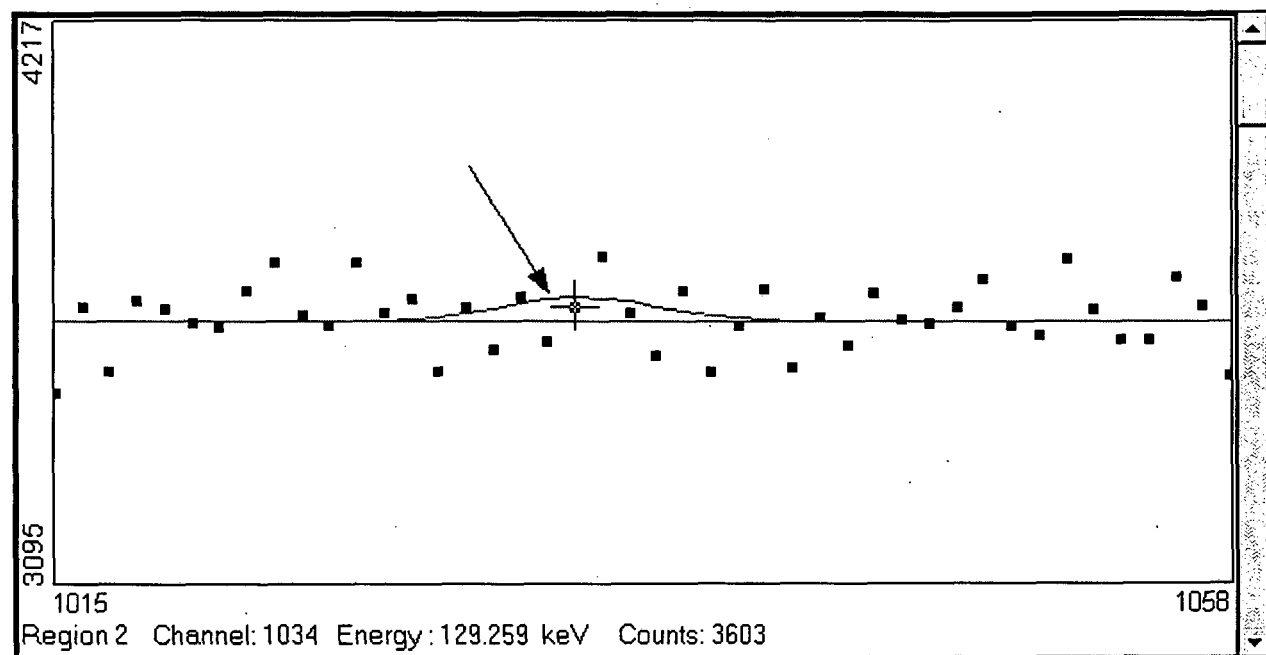

Fig. 4. 129keV region of the spectrum. The curve designated with the arrow denotes the 129keV peak as fit by the software.

This demonstrates one of the weaknesses in this analytical approach. Even when no peak is present, the net counts in a region of interest will often be a positive value. In this situation, the isotopic ratio calculated can be unrealistically high if the isotopes in the ratio have significantly different specific activities. When performing the first several analyses with this procedure, it is best to carefully visually inspect the spectrum. Not only will this reveal issues such as that demonstrated here with ^{239}Pu, it will also allow the spectroscopist to verify that the energy calibration is correct, that the regions of interest are well selected, and that no other problems are apparent. Figure 4 also

demonstrates that one can also visually inspect the peak fitting performed by FRAM. It is always a good idea to visually inspect the peak fitting, especially when developing a new parameter set.

Determination of the efficiency curve for the spectrum/detector requires an isotope with widely distributed peaks. For the spectra analyzed in this investigation, the reference isotope, ^{241}Am provided these peaks. We know the branching ratios of the ^{241}Am peaks; comparing their calculated areas to their branching ratios allows us to fit an efficiency curve, as shown in Fig. 5.
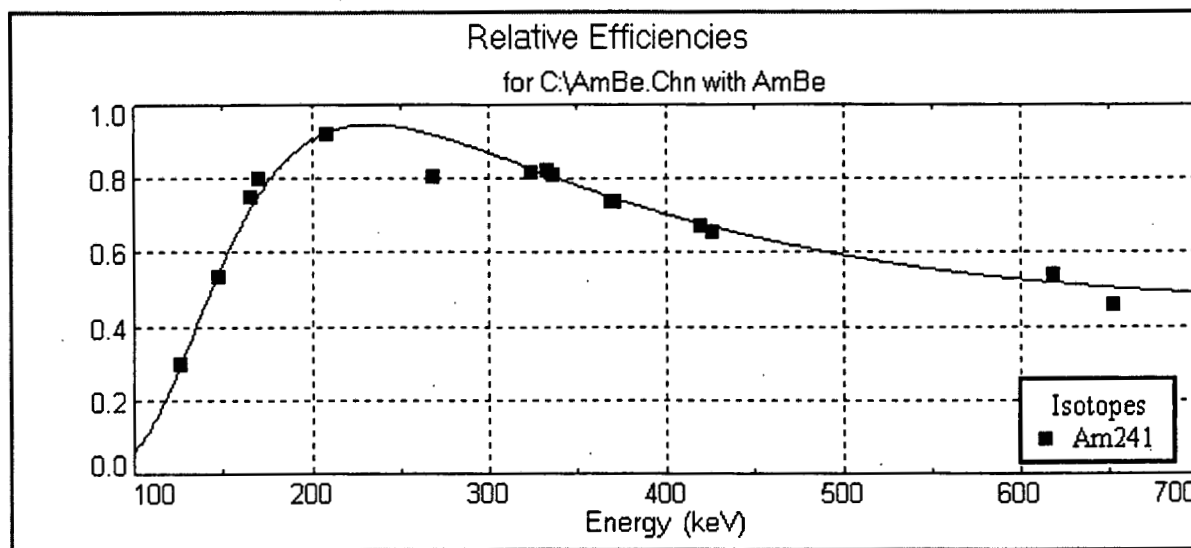

Fig. 5. Efficiency curve calculated for a representative AmBe source.

The task of developing parameter sets should only be undertaken by a capable, experienced spectroscopist. Those unfamiliar with high-resolution gamma-ray spectroscopy should not attempt to create parameter sets for the atypical FRAM analyses discussed in this paper (or other FRAM analyses). Errors in the parameter set can cause incorrect analysis, and the errors may not be apparent.

STRENGTHS OF THIS APPROACH
This approach is very flexible. Any significant peak from an isotope of interest can be used for analysis. Any isotope with suitable gamma rays can generally be analyzed. Different types of germanium detectors can be used, and representative standards are often not necessary.

Once a good parameter set has been created, the analysis of appropriate spectra is greatly simplified. Even a user who is not an experienced spectroscopist can get good results. With a little instruction, such a user can also evaluate the quality of the analysis by checking the peak fits and monitoring trends in uncertainty values. The analysis is also reproducible and easily documented for auditing.

As with typical FRAM analysis, the technique is not sensitive to changes in input rate or source geometry. The presence of background peaks can often be accommodated. The only requirement is that the isotopes be homogenously distributed. The importance of this requirement depends on the self-attenuation properties of the matrix and the source thickness.

The design of the software makes it easy for the experienced spectroscopist to create parameter sets. Parameter set editing takes place within the FRAM software; no programming abilities are required. All parameters are entered in a very intuitive manner through standard windows. To ensure clarity, the users' manual explains the information fields in the windows.

The software automatically corrects the analysis for the shape of the efficiency curve. This enables the use of widely separated peaks, detectors with different efficiencies, and very significant levels of shielding and self-attenuation.

CONCLUSION
It has been demonstrated that FRAM can be used to analyze nonplutonium/nonuranium spectra for isotopic content. A specific example (AmBe neutron sources) was discussed. The weaknesses and strengths of using FRAM to analyze nonplutonium/nonuranium spectra were discussed.

ACKNOWLEDGEMENT

REFERENCES
1. David S. Bracken and Philip A. Hypes, "Solid-State Calorimeter," *Nucl. Mater. Manage.* **XXIX** (Proc. Issue) CD-ROM (2000).

2. Thomas A. Kelley, Thomas E. Sampson, Dorothea DeLapp, "PC/FRAM: Algorithms for the Gamma-Ray Spectrometry Measurement of Plutonium Isotopic Composition," Proceedings of the Fifth International Conference, Facility Operations-Safeguards Interface, Jackson Hole, WY, USA, September 24–29, 1995.

3. Thomas E. Sampson, Thomas A. Kelley, Teresa L. Cremers, T.R. Konkel, and R.J. Friar, "PC/FRAM: New Capabilities for the Gamma-Ray Spectrometry Measurement of Plutonium Isotopic Composition," Proceedings of the Fifth International Conference, Facility Operations-Safeguards Interface, Jackson Hole, WY, USA, September 24–29, 1995.