A Comparison Between Two Methods of Gamma Spectrum Analysis for FDA Food Emergency Response Network Food Samples

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Abstract

To safeguard nation's food supply against radioactive contamination in the event of a nuclear or radiological incident, Food Emergency Response Network (FERN) needs to detect and identify gamma-emitting radionuclides in foods. Proficiency test (PT) samples are routinely sent to FERN member laboratories for analysis of food samples containing multiple gamma-emitting radionuclides. FDA's Winchester Engineering and Analytical Center (WEAC) also participates in these proficiency studies.

Gamma spectra of various samples were analyzed using Canberra APEX and VRF (Visual RobFit) software to compare their performance characteristics. Traditionally, gamma spectrum analysis begins with simple peak search followed by subtraction of environmental background. If the net peak area for a given peak is statistically significant, it is considered as detected and further analyzed for radionuclide identification. When there are unidentified peaks in the spectrum, adjustments to peak search parameters or energy recalibration will be required, which can be tedious and time-consuming.

Identification and quantification of gamma radionuclides are crucial for radiological risk assessment. In order to identify gamma radionuclides quickly and reliably, an alternative approach applying non-linear least-squares minimization of chi-square to fit full-spectrum and continuum shapes was studied, which can be used to provide additional confirmation for critical sample analysis. A comparison study of Canberra Apex and VRF software was performed for analyzing the gamma spectra of samples containing mixed gamma radionuclides of interest, natural Th and U decay chains, and fresh fission products.

The results indicate that VRF is capable of identifying gamma-emitting radionuclides qualitatively using an alternative method of peak processing. However, Apex software is superior in that it can reliably identify and quantify gamma-emitting radionuclides of interest simultaneously.

Introduction

Gamma-rays, or gamma radiation, are high-energy, short-wavelength, electromagnetic radiation emitted from the nucleus of an atom. Some radioactive materials emit gamma radiation during their decay. Gamma radiation is a radiation hazard for the entire human body as they can pass completely through the body and cause ionization that damage tissue and DNA.

Since various radiological incidents may pose food safety and public health concerns, FERN Radiological Proficiency Testing program was established to improve and evaluate method performance and laboratory proficiency among member laboratories. The analysis of gamma-emitting radionuclides in foods has been performed using Canberra APEX software at WEAC. Canberra APEX is a spectrometric analysis with simple peak search followed by subtraction of environmental background. The advantage of the software is its ability to identify and quantify gamma-emitting radionuclides. Quantification requires significant processing of the data.

Samples were analyzed with Canberra APEX and VRF software to compare their results. VRF adopts a new and different method for analysis of high-resolution gamma-ray spectra using non-linear least-squares minimization of chi-squared fitting techniques to fit full-spectrum radionuclide shapes. A background spectrum is never needed and never subtracted, and the original data is never altered.

Materials and Methods

Instrument: High purity germanium detector (HPGe)

Figure 1. High purity germanium detector (HPGe) (Left) Inside gamma detector shielding (Right)

Application Software:

- Canberra APEX
- VRF (Visual RobFit)

Test Samples:

- 2020 FERN PT samples
  Containing mixed radionuclides of interest
- EZA103294 Fission Product
  Containing mixed radionuclides typically from nuclear power plant accident
- Tank top sample
  Containing mixed radionuclides associated with natural Th/U decay chains

Each type of sample was homogenized. A 400-mL portion of the sample was transferred into a cylindrical polypropylene container, and then the container was filled to 400-mL fill line in order to maintain a constant geometry for all samples countered on the detector. Each sample was weighed. The detector is calibrated using mixed gamma radionuclide standards in an identical cylindrical polypropylene container filled to the 400-mL line. Each sample was placed on detector and counted for 100 minutes. All required QC checks for the detector were performed before sample analysis.

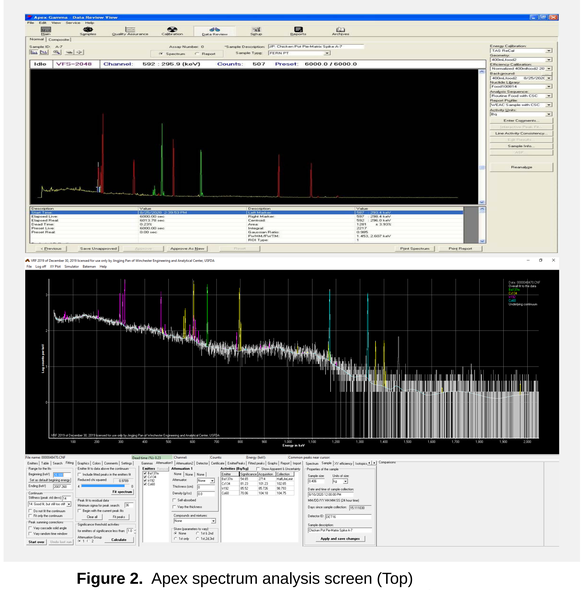

Figure 2. Apex spectrum analysis screen (Top) VRF spectrum analysis screen (Bottom)

Results and Discussion

Unknown radionuclides in FERN PT samples (A-7 and B-7) were analyzed for radionuclide identification using Canberra Apex. An analysis report was generated automatically without printed spectrum. In both types of software, the user has an opportunity to set up different libraries for analysis of data depending on the radionuclides of interest. Our laboratory's library for routine analysis in Apex was applied to the results as well as VRF software. The fact that Ir-192 was not originally identified using Apex is a result of it not being included in our routine library. This stresses the importance of reviewing the energy spectrum generated during analysis to make sure all peaks are identified. This process requires extra time and expertise.

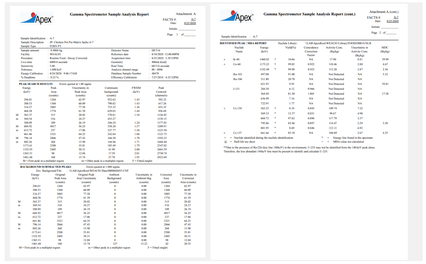

Figure 3. Apex gamma report for FERN PT A-7 sample

VRF was also used for identifying unknown radionuclide in FERN PT A-7 and B-7 samples, which allowed this chemist to perform interactive-spectrum analysis by commanding VRF to pick peaks and check fit of the combination of spectrum-wide emitter shape. With VRF software, all radionuclides including Co-60, Cs-134, Cs-137, and Ir-192 were correctly identified.

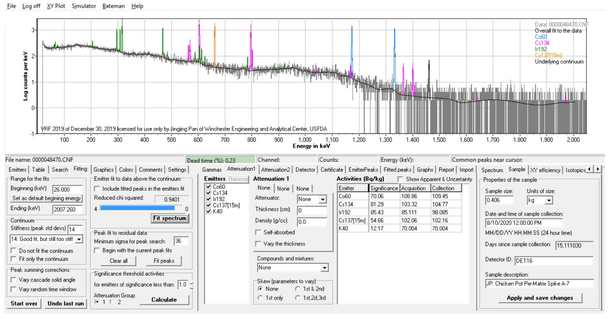

Figure 4. FERN PT A-7 sample identified with VRF

Table 1. Comparison of radionuclides identified by Apex and VRF

Sample ID	Radionuclides	Apex Identified	VRF Identified	Ratio of Measured Activity Concentration/Known Activity Concentration		
				Apex Value	VRF Value	Known Value
A-7	Co-60	✓	✓	1.00	0.99	1.00
	Cs-134	✓	✓	1.02	0.92	1.00
	Cs-137	✓	✓	0.97	0.99	1.00
	Ir-192	First, it was not identified, then was reprocessed to identify	✓	0.96	0.90	1.00
B-7	Co-60	✓	✓	1.00	0.99	1.00
	Cs-134	✓	✓	0.97	0.93	1.00
	Cs-137	✓	✓	1.01	1.05	1.00
	Ir-192	First, it was not identified, then was reprocessed to identify	✓	0.97	0.89	1.00

Table-1 shows that identification of Ir-192 was missing on the Apex gamma analysis report. because Ir-192 was not listed in the nuclide library for the method chosen. Later Ir-192 was added to the library and Apex gamma report was reprocessed to make Ir-192 show up on the report. It took about 30 minutes. With VRF, it took less than 3 minutes to analyze the VRF spectrum.

Table 2. Radionuclide identification – Sample (EZA103294 fresh fission product)

Radionuclide	Identified by VRF
Ba-140	✓
Ce-141	✓
Ce-144	✓
I-132	✓
La-140	✓
Mo-99	✓
Nb-95	✓
Nd-147	✓
Ru-103	✓
Tc-99m	✓
Te-132	✓
Zr-95	✓

Table 3. Radionuclide identification – Sample (Tank top)

Radionuclide	Identified by VRF
K-40	✓
Tl-208	✓
Bi-212	✓
Pb-212	✓
Bl-214	✓
Pb-214	✓
Ra-223	✓
Ra-224	✓
Ra-226	✓
Ac-228	✓
Th-231	✓
Pa-234m	✓
U-235	✓
Rn-220	✓
Th-234	✓
Th-228	✓

Figure 5. Tank top sample printed with gel dots containing natural Th and U

Conclusion

VRF method has an ability to quickly and accurately identify unknown radionuclides in FERN PT foods qualitatively. VRF never alters the original data with background subtraction.

While use of Apex Software may require the establishment of a new library in the software to address unique samples, it is still the ideal choice for food analysis as it can very accurately identify and quantify gamma-emitting radionuclides in food products simultaneously.

VRF gives confidence in qualitative results, but needs to be fine-tuned, especially for quantification analysis.

Acknowledgement and Disclaimer

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