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Title: Application of Spectral Summing to Suspect Low Level Debris Drums at Los Alamos National Laboratory

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Abstract

The spectral summing technique developed by Pajarito Scientific Corporation (PSC) is a unique modeling technique that is being employed by the Waste Disposition Project – Low Level Waste Disposal (WDP-LLWD) Group at Los Alamos National Laboratory (LANL). This technique is being used to disposition low-level radioactive waste that has dropped out of the transuranic (TRU) category and has no place to go unless it can be proven to be LLW and not TRU. The TRU program at LANL run by Mobile Characterization Services (MCS) employs two High Efficiency Neutron Counters (HENC) with built-in gamma assay systems to assay radioactive waste for shipment and disposal as TRU waste at the Waste Isolation Pilot Plant (WIPP) at Carlsbad, New Mexico. As well as being certified for WIPP assays, the HENC systems can also be used for low-level waste assays for disposal at LANL or off-site disposal facilities, such as the Nevada Test Site (NTS). Some of the waste processed through the HENC systems cannot be confirmed TRU due to the absence of detected TRU alpha emitters above the TRU cutoff of 100 nCi/g. This waste becomes suspect low-level waste (SLLW). In many cases, the waste also can't be classified as LLW because the minimum detectable activity (MDA) of TRU radionuclides is above the 100 nCi/g level. These wastes that do not have enough detectable TRU activity to be classified as TRU waste and have too high a MDA to be classified as LLW enter a radioactive waste characterization limbo that prevents their dispositioning as either TRU waste or LLW. Spectral summing allows an experienced gamma spectroscopy analyst to add the HENC gamma spectra of a number of similar waste items together to form a consolidated (summed) spectrum. This summed spectrum contains the assay results of the group of items rather than the individual item, and gamma peaks that were not discernable in the individual spectra become quantifiable in the summed spectrum. The group of waste items can then be properly classified as LLW based on the summed spectrum and valid assay values can be assigned for disposal. This technique is being successfully used to dispose of LLW debris drums from LANL.

Introduction

The Waste Disposition Project (WDP) at Los Alamos National Laboratory (LANL) utilizes multiple Nondestructive Assay (NDA) techniques for the characterization of waste items for disposition as transuranic (TRU) or low level (LL) waste. One of the NDA techniques used is passive neutron counting with built in gamma spectroscopy. A NDA system employing this technique is called a High Efficiency Neutron Counter (HENC) with built in gamma spectrometer. A HENC NDA system is a cavity based system with Helium-3 (He3) detectors arranged in a 4-π geometry for neutron detection and a single High Purity Germanium (HPGe) detector directed at the measured item for gamma-ray detection. Two of these assay systems are used at LANL's Technical Area 54 (TA-54): HENC1 and HENC2. Both of these NDA systems are certified by the Carlsbad Field

Office (CBFO) for characterization of 55-gallon drums for disposal at the Waste Isolation Pilot Plant (WIPP).

Most of the drums measured by the two HENC systems at LANL's TA-54 are prescreened as likely TRU as defined by WIPP's waste acceptance criteria (WAC) and subsequently confirmed TRU once measured by a HENC system. Some drums measured by the HENC systems at LANL could not be confirmed as TRU because no α-emitting transuranic nuclides were detected. Furthermore, since the minimum detectable TRU alpha-activity concentration (MDC) was greater than 100nCi/g these drums could not be confirmed as low level waste. Thus, these drums are considered suspect low level waste (SLLW) drums and have no direct disposal option.

A technique for establishing these drums to be low level in compliance with the NTS WAC has been applied to a set of suspect low level debris drums measured by a HENC system at TA-54. The technique is called spectral summing and was used to combine the gamma spectra from multiple SLLW drums with the intent of reducing the MDC for the group of drums to below 100nCi/g. If the group average MDC is sufficiently below 100nCi/g and none of the individual spectra show evidence of TRU activity, then a reasonable argument can be made that the actual TRU alpha-activity concentration of any single drum in the group is unlikely to exceed 100nCi/g. The technique utilizes the existing NDA data for the drums so that no additional time or money is spent to re-count the individual drums for a longer duration on a HENC system or count the drums in a different NDA system to attempt to achieve individual TRU alpha MDCs less than 100nCi/g. A detailed description and technical basis for the spectral summing technique is available in [Reference 1.]

Selection of Debris Drums for Spectral Summing

For spectral summing to be most effective, it is preferable to select drums of similar waste types (waste streams) to be grouped together for the summing. The LANL WDP has identified several hundred SLLW 55-gallon drums that were measured on a HENC system from two primary waste streams: 1) debris and 2) sludge. From these SLLW drums, the debris drums with TRU alpha MDC values less than 120nCi/g (and greater than 100nCi/g) were identified and selected. There were fifty four drums in this category. Of these drums, fifty two were measured on the HENC1 system and two were measured on the HENC2 system. The drums measured on the HENC2 system were removed from the selection as the calibration for the two systems were not sufficiently matching. The remaining fifty two drums measured on the HENC1 system comprised the final set of drums to which spectral summing was applied.

Calibration Parameters

The calibration of a HENC system performs a matrix correction based on an average density calculated using the net weight and dimensions of the container. This matrix correction is performed by the system software, NDA 2000 [Reference 2], using a multi-curve efficiency correction. Multiple efficiency curves generated during calibration at different densities are interpolated by the measured density to generate an efficiency curve representative of the assayed container.

The HENC system stores all of the calibration data from the instrument utilized in performance of the assay and calculation of the results in the raw data files. This makes it possible to extract values

for the multi-curve efficiency calibrations from each debris drum assay. In order for the spectral summation of multiple assays to be possible, there must be no significant changes in the instrument response during the period of operation. This operational consistency is evaluated by reviewing the multi-curve efficiencies, stored in the raw data files, of all the measurements for which a summation analysis is required.

Prior to performing the summation, a detailed review was performed on an individual container basis. The gamma spectra were reviewed manually, ensuring that no peaks were missed by the automated peak location algorithms used in the original data analysis. After a thorough review of all the calibration data for the instrument during the time span of the assays performed on the debris drums, it was determined that there were no major changes performed to the instrument affecting data quality.

Initial Spectral Summing

Spectral summation was performed on the gamma spectra for the fifty two (52) debris containers and an average density calculated based on the debris drum information. Following initial summation of the data, it was found that there were peaks present at higher energies consistent with ^{241}Am. The 662 keV and 722 keV energy peaks were present in sufficient quantity and quality to determine they were indeed associated with the presence of ^{241}Am. Further review of the sum spectrum showed at lower energies the ^{241}Am peaks were not consistent with the expected values based on branching ratios of the isotope and detector efficiencies. The lower energy peaks were more than an order of magnitude smaller than expected. The detailed review performed on the sum spectrum indicated that it was highly likely that the source of the ^{241}Am was external to the instrument (i.e., background); this assessment is based on the excessive attenuation at lower energies.

Background Subtraction (Stripping)

The spectral summation technique is not limited to analyzing assays of containers. It can also be used to perform summations of backgrounds to establish any additional contributions due to background. A peak consisting of a total of ten counts, over a thirty minute background, would be nearly indistinguishable based on a single measurement. But when a summation of fifty backgrounds is performed, this peak will be nearly five hundred counts above the spectral continuum. This is what was found by performing a summation of the background spectra that were acquired on the days that the debris drum assays were taken. This background summation showed the same characteristic high energy ^{241}Am peaks present in the summation of the debris container data with the previously mentioned attenuated lower energy peaks.

In order to properly account for the background in the data analysis, it was necessary to strip the debris drum spectra of the corresponding background and perform a spectral summation of the stripped spectra. Some of the debris drums were measured on the same day as each other and are therefore represented by the same background spectrum. In these cases, the background for that day was only subtracted once (for only one of the debris drums measured on that day) in order to prevent spectral features present in a single background from being unduly magnified in the final summation of data. Thus, the final background corrected summation does not entirely account for all the “bleed through” of the ^{241}Am seen in the background summation. But this background stripping was enough to completely eliminate the lower energy peaks from ^{241}Am in the background

corrected summation. The higher energy peaks for ^{241}Am were significantly reduced and it was determined that their contribution is entirely due to background external to the instrument. The only other (than ^{241}Am) major TRU radionuclide present in the final background subtracted summation was ^{239}Pu. Based on what appeared to be significant low energy attenuation, the resulting ^{239}Pu could be partially attributed to background. However, this was not as conclusive for ^{239}Pu as it was for ^{241}Am. So, as a conservative measure, the ^{239}Pu was treated as originating from the debris drums and was therefore quantified in the final spectral summing analysis.

Final Spectral Summing

After the final summation of the background corrected debris data, the summary results were calculated using weapons grade isotopics for plutonium (Pu), and assuming an aged in-growth of ^{241}Am at 30 years. These final results were then used to calculate the average TRU alpha activity concentration of the population using the total net weight of the containers. This resulted in a measured average TRU alpha concentration of 37 nCi/g.

Disposition and Status of Debris Drums

The results produced by the spectral summing application to the fifty two debris drums were presented to Perma-Fix® representatives for consideration to accept these drums for treatment as low level waste (LLW). After review, this application of spectral summing for characterization of LLW was approved by Perma-Fix®.

For shipping manifest purposes the drums are required to be characterized on an individual basis. To achieve this, the group average TRU alpha activity was equally distributed amongst the fifty two drums (assigning the same activity to each drum) and then the individual measured weight of each drum was used to calculate a TRU alpha activity concentration for each drum. The resultant individualized TRU alpha activity concentrations were all still well below 100nCi/g. The fifty two debris drums have now been dispositioned as LLW, are staged at LANL's waste facility and are in the queue for shipment and disposal as LLW.

Conclusion

The WDP considers that the application of spectral summing to the fifty two SLLW debris drums identified in this report was successful. The group average MDC was reduced sufficiently below 100nCi/g that the final spectral summing was able to determine a measured TRU alpha concentration for the group of 37 nCi/g. This is well below the TRU/LL waste threshold of 100nCi/g. This analysis provides a reasonable argument that the actual TRU alpha-activity concentration of any single drum in the group is unlikely to exceed 100nCi/g. Thus, making each of these drums a reasonable candidate for disposition as LLW. The application data was submitted to Perma-Fix® for consideration and was approved as an acceptable technique to characterize LLW for treatments at Perma-Fix®. TRU alpha activity concentrations were calculated for each drum by dividing the spectral sum activity equally amongst the drums and using the individual measured drum weights. The drums are staged as LLW and pending shipment.

References

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2. NDA 2000: Technical Reference Manual, Canberra Industries, 9231595D V3.2, 2003.

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