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MULTI-ISOPTOPE PROCESS MONITOR: A COMPARISON OF MODEL AND EXPERIMENTAL VALIDATION

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Introduction

The MIP Monitoring concept was demonstrated through a series of simulations as well as experiments. While qualitatively these studies generated similar results, there were significant differences between the simulated and experimentally determined spectra. This chapter discusses differences between modeled and experimentally determined CZT gamma spectra and subsequent PCA analysis generated from each.

Comparison of Model to Experiment

Modeled and experimental spectra representing aqueous raffinate from similar spent nuclear fuel sources are provided in Figure 1. The raffinate stream was from a liquid-liquid extraction of dissolved spent fuel (16 MWd/kgU), separated at an acid concentration of ~2.5 M acid. Apparent from Figure 1, model simulations were unable to fully reproduce the structure of the experimentally derived spectrum. Limitations likely exists in all three models used to generate the simulated spectrum including ORIGEN-ARP, AMUSE and Synth.
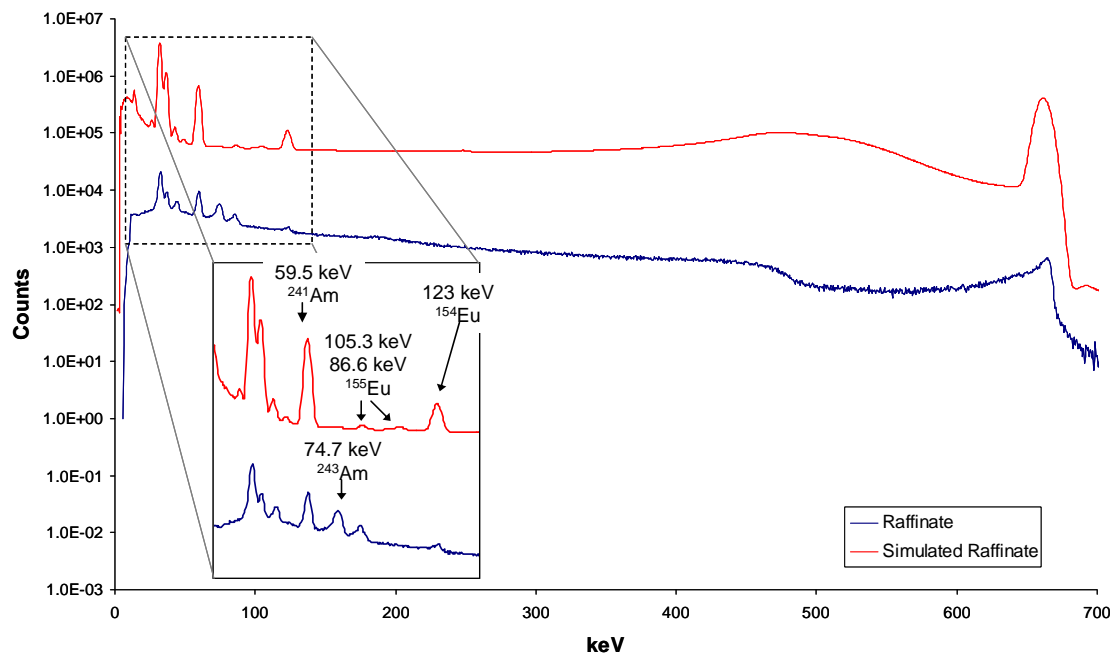
Figure 1. Modeled and experimental CZT spectrum of ~2.5 M raffinate of the 16 MWd/kgU fuel.

Discrepancies due to the potential inaccuracy in ORIGEN output were considered as potential

sources of error. Though it appears that the model misrepresents several nuclide activities, it was not

clear whether this was an error in the input file or the calculation methods employed by ORIGEN. A few

active elements produced by ORIGEN were left out of the liquid extraction model and subsequently, the

simulated spectra. These elements were omitted because the constants used to calculate their distribution

by AMUSE were determined to be either missing or inaccurate. However, none of the omitted nuclides

had gamma lines in the areas where discrepancies were identified, so these emissions would not have

caused the discrepancies identified below.

Another possible, but less probable, explanation for model versus experimental peak

discrepancies is the omission of peaks by the gamma library accessed by Synth to populate the spectra.

This possibility would suggest Synth is mishandling major gamma lines of various isotopes in order for

this to be significant within the Compton region of the spectra. Most gamma libraries are relatively

accurate with respect to major gamma lines, typically only lacking accurate information for low branching ratio peaks or very high energy lines that are difficult to detect.

The most pronounced difference between modeled and experimentally derived spectra is the Compton scatter region of the 661 keV gamma energy peak from [137]Cs. Compton edge differences in the simulated spectra were due to limitations of the Synth code. These limitations have been previously identified and discussed for Ge detectors[1] and attributed to neglected scatter from the shielding material. Synth creates the Compton region by using the theoretical shape of the Compton spectrum, based on a peak-to-Compton ratio appropriate for the volume of the detector. The code then adds these counts to the spectrum for each peak.

For the purpose of testing the MIP approach, these issues in the simulated Compton edge are thought to be of minor importance. Here, spectra were used for relative comparisons in the MIP monitor approach. Since the Compton edge (correctly or incorrectly simulated) would be relatively consistent between data sets, inaccurate, but consistent, simulations would not contribute significantly to the overall variance between spectral groups.

Modeled spectra were also lacking a few key peaks at 75 and 86 keV, and over-emphasized the peak at 123 keV. Since these discrepancies were also apparent on the feed spectra for the 16 MWd/kgU fuel (before separation), these issues likely originated from either ORIGEN-ARP or Synth, rather than AMUSE. The spectral region beyond the [137]Cs (661 keV) peak was not shown, as the counts are too low on the experimental spectrum to produce any meaningful peaks.

The missing gamma line at 75 keV in the simulated spectrum was due to either inaccuracies in handling [243]Am or the Compton contribution from [137]Cs. Upon further investigation, [243]Am contributes a significant number of counts in the 75 keV region of the modeled spectrum. However, this contribution did not rise above the modeled Compton background. It is unclear whether the modeled [243]Am activity is the result of this missing peak, an overestimated Compton effect from the 661 keV gamma line, or both.

The simulated spectra were also missing a gamma line at 86 keV seen in the experimental spectra. At the 86 keV line, the model identified [155]Eu as the main contributor. However, [155]Eu also has a

105 keV gamma ray, with a similar branching ratio (21% for 105 keV versus 31% for 86 keV), for which a peak was not apparent in the experimental spectrum. This does not rule out [155]Eu as the cause, as the detector has a sharp drop of efficiency with energy. Synth does not model the efficiency curve perfectly, leaving the root cause of the missing 86 keV peak questionable. [243]Am contributes a small fraction of its decay gammas to the 86 keV line as well, though its low branching ratio (0.34%) makes its activity an unlikely source of error. Additional analysis is necessary to discover or confirm a reason for the discrepancy.

The relatively oversized simulated peak at 123 keV is primarily produced by [154]Eu. One possible explanation for the overemphasis is the inability of Synth to accurately model the rapid decline of efficiency of the CZT crystal with energy. It is possible that this decrease in efficiency is sufficient to account for the smaller peak in the experimental spectrum.

The spectra from both simulations and experiments were analyzed using an unsupervised Principal Component approach. CZT spectra of the organic fraction of the liquid extraction were analyzed, following normalization by area and mean centering. A scores plot of the unsupervised PCA of the simulated spectra is found in Figure 2.
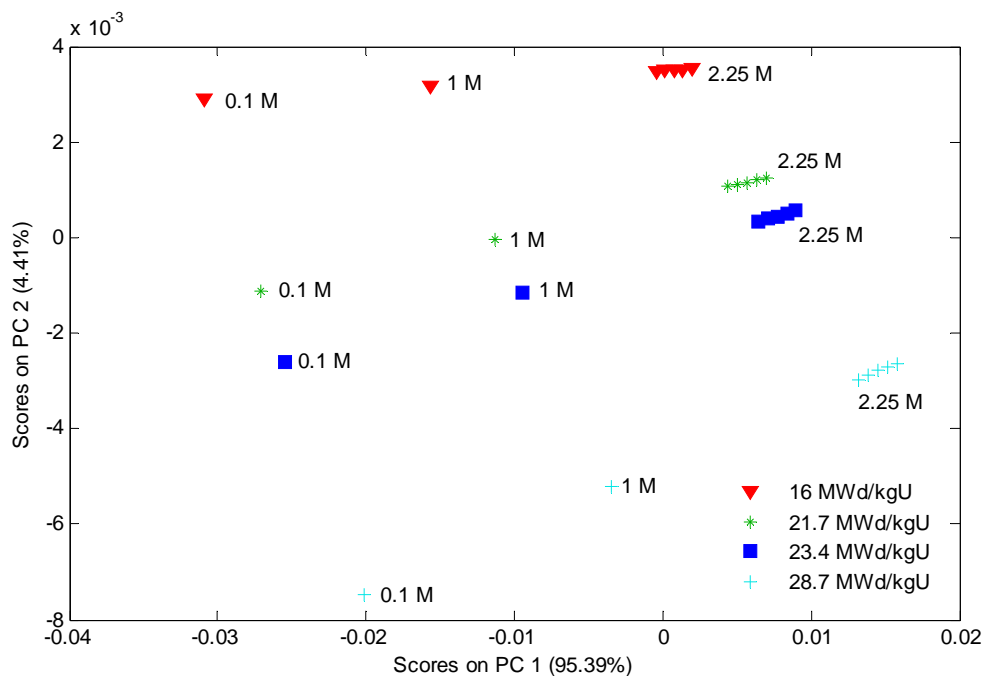
Figure 2. Scores plot from PCA of simulated organic extract spectra using a CZT detector

Figure 2 clearly illustrates how the samples grouped according to burn up and acid concentration in PCA

scores space. Simulated aqueous samples also grouped in a like manner.
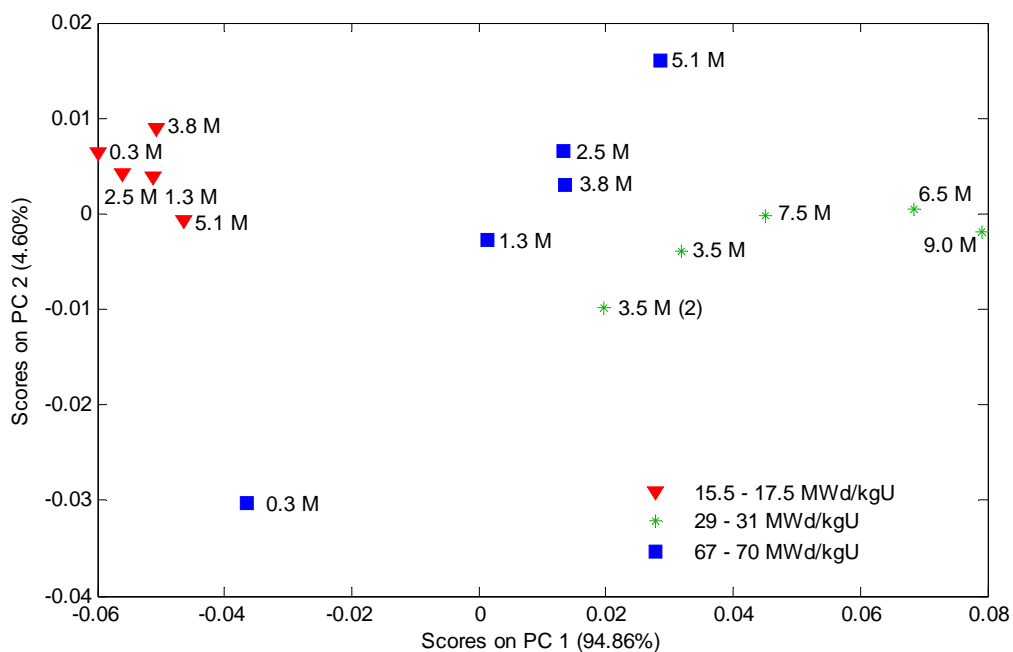

Figure 3. Scores plot from PCA of experimental organic extract spectra using a CZT detector

Similar PCA, as conducted using corresponding experiment spectra, is shown in Figure 3. From this figure it is clear that experimental samples do not group as distinctly in PCA scores space as the simulated spectra. Due to resource constraints, multiple samples at each acid concentration were not available, which would have enhanced PCA of the experimental spectra. However, even considering the rather small amount of data available, the experimental spectra still group according to burnup. The PCA results also suggest there may be potential for grouping according to the extraction acid concentration with advanced preprocessing of the data. The close proximity of spectra taken from duplicate samples at 3.5 M acid concentration for the ATM 109 fuel (67-70 MWd/kgU) suggests process variations in replicate spectra were relatively small. Further analysis of both the experimental and simulated spectra will be necessary to delineate these pattern shifts.

While comparisons of model and experimental spectra have identified important differences and shortcomings of simulations, it is important, also, to acknowledge the similarity of the model spectra to the experiment. For instance, several prominent peaks are visible in both the simulated and experimentally derived spectra including the 661, 123, 60, 43, 36, and 32 keV lines. Also, simulations correctly predicted the capability of compiling multivariate analysis and gamma spectroscopy as a promising technique for safeguards technology development. Further study is necessary to determine the root causes of the minor differences between model and experiment and to test and validate the MIP approach with more realistic, continuous flow systems.

Summary and Conclusion

The results of the experimental validation of the MIP modeling concept were compared to model simulations. PCA scores plots generated from modeled spectra were more distinguishable by both burnup level and acid concentration. Several peaks were missing from the simulated gamma spectra, and potential causes of this discrepancy were hypothesized, including limitations in Synth, ORIGEN-ARP and AMUSE. Nonetheless, the PCA based on the experimentally derived spectra were able to correctly

identify off-normal conditions at a 99% confidence interval. Overall, the model represented the

experimental observations for the purpose of testing the MIP monitoring concept.

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[1] W.K. Hensley, A.D. McKinnon, H.S. Miley, M.E. Panisko, and R.M Savard. "Synth: A Spectrum Synthesizer." *Journal of Radioanalytical and Nuclear Chemistry, Articles,* Vol. 193, No. 2, p. 229-237, 1995.