Project 95009

Radioanalytical Chemistry for Automated Nuclear Waste Process Monitoring

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RESULTS TO DATE:

1) Comparison of different pulse shape discrimination methods for BC400/BGO phoswich and CsI:Tl detectors.

Comparison of different pulse shape discrimination methods was performed under two different experimental conditions and the best method was identified. Beta/gamma discrimination of 90Sr/90Y and 137Cs was performed using a phoswich detector made of BC400 (2.5 cm OD x 1.2 cm) and BGO (2.5 cm O.D. x 2.5 cm) scintillators. Alpha/gamma discrimination of 210Po and 137Cs was performed using a CsI:Tl (2.8 x 1.4 x 1.4 cm3) scintillation crystal. The pulse waveforms were digitized with a DGF-4c (X-Ray Instrumentation Associates) and analyzed offline with IGOR Pro software (Wavemetrics, Inc.). The four pulse shape discrimination methods that were compared include: rise time discrimination, digital constant fraction discrimination, charge ratio, and constant time discrimination (CTD) methods. The CTD method is the ratio of the pulse height at a particular time after the beginning of the pulse to the time at the maximum pulse height. The charge comparison method resulted in a Figure of Merit (FoM) of 3.3 (9.9 % spillover) and 3.7 (0.033 % spillover) for the phoswich and the CsI:Tl scintillator setups, respectively. The CTD method resulted in a FoM of 3.9 (9.2 % spillover) and 3.2 (0.25 % spillover), respectively. Inverting the pulse shape data typically resulted in a significantly higher FoM than conventional methods, but there was no reduction in % spillover values. This outcome illustrates that the FoM may not be a good scheme for the quantification of a system to perform pulse shape discrimination. Comparison of several pulse shape discrimination (PSD) methods was performed as a means to compare traditional analog and digital PSD methods on the same scintillation pulses. The X-ray Instrumentation Associates DGF-4C (40 Msps, 14-bit) was used to digitize waveforms from a CsI:Tl crystal and BC400/BGO phoswich detector. The CsI:Tl was characterized for alpha/gamma-ray discrimination with 210Po and 137Cs, while the BC400/BGO phoswich detector was characterized for beta/gamma-ray discrimination with 90Sr/90Y and 137Cs. The digitized waveforms were analyzed offline using IGOR Pro (Wavemetrics Inc). The four PSD methods investigated were rise time discrimination, constant fraction discrimination, charge ratio and constant time discrimination (CTD). The CTD method uses the normalized digitized value of the pulse at a fixed instant from the start of the pulse as the pulse shape parameter. The pulse shape data was binned to obtain a pulse shape spectrum. Each method was optimized to obtain the best PSD parameter setting for the highest figure-of-merit (FOM) and lowest spillover. The pulse shape data was also inverted to investigate if there were any improvements to the PSD. The FOM and spillover were obtained from the optimized pulse shape parameters and used to compare the performances of the different methods. The pulse shape parameter obtained for each method is plotted against energy deposited resulting in a dual-parameter spectrum. A typical dual-parameter spectrum for beta-gamma discrimination using the phoswich detector and the rise time discrimination method is shown in figure 1a. The corresponding dual-parameter spectrum obtained from the inverted pulse shape discrimination data is shown in figure 1b. The corresponding single parameter pulse shape spectra are shown in figure 2a and 2b, respectively. Note that although the resolution of the inverted spectral data is greater (larger FOM) the ability to perform PSD is the same, i.e. the spillover remains the same. The CTD method provided the best performance in most instances, with charge comparison coming in a close second. Table 1 summarizes the experimental findings.

2) CsI:Tl temporal luminosity dependence on radiation type and energy

The primary objective of this research was the investigation of the dependence of light intensity and decay time constants on the radiation type and the energy deposition within a CsI:Tl crystal. A 1.4 x 1.4 x 2.8 cm3 CsI:Tl crystal was coupled to an Burle, 8575 photomultiplier tube. The PMT anode output was digitized at 40 MSPS with a GaGe 8012A/PCI (12-bit) CompuScope digital oscilloscope and data processing through National Instruments LabVIEW software. The relative intensities and decay time constants were determined following fitting the signal decay to two exponential decays. For alpha radiation, the relative intensities were in the range of 0.8 and 0.2, and decay time constants were 0.3 μs and 2.0 μs for the fast and slow components, respectively. For gamma-ray radiation the relative intensities were in the range of 0.56 and 0.48, and decay time constants were 0.5 μs and 3.0 μs for the fast and slow components, respectively. The relative intensity and decay time constants were independent of alpha and gamma-ray energies in the range of 3.182 to 5.49 MeV, and 0.512 to 0.83 MeV, respectively. The results were used to develop a model for determining pulse shape discrimination capability given the luminosity temporal distribution.   The response of a scintillator to energy deposition from ionizing radiation is in the form of photon emission. The photon yield for certain crystals is observed as an exponential increase in the number emitted followed by decrease through a fast and slow mode. The output light pulse as a function of time thus consists of the summation of a single exponential rise and single or multiple exponential decay modes. CsI:Tl is an inorganic scintillator well suited for the application of pulse shape discrimination techniques, since it has the ability to produce scintillation pulses with varying light intensities and distinct timing characteristics according to the incident radiation. It can thus differentiate alpha particles from beta or gamma radiation on the basis of light contained within the fast and slow modes and the decay constants for the same. The primary objective of this research is characterization of the temporal luminosity of CsI:Tl both with respect to radiation type and energy variation. The second objective is then to apply the correlation determined between the radiation type and energy with luminosity to the pulse shape spectrum. A Monte Carlo model was developed to investigate the correlation of the temporal luminescence of the scintillation pulse with the pulse shape spectrum.   The experiments were performed with 1.4x1.4 x 2.8 cm3 CsI:Tl crystal coupled to the photomultiplier tube (Burle, 8575) using silicone oil. Alpha and gamma sources with diversified energies as mentioned in Table 2 were used. The entire assembly is placed inside an aluminum box and connected to the data acquisition system. The data acquisition is conducted with a GaGe 8012A/PCI (12-bit) CompuScope digital oscilloscope at 40 MSPS and data processing is achieved through digital signal processing software LabVIEW (version5.1), developed by National Instruments. Filtering the pulses for particular energy of interest is an essential requirement for these experiments. The data is average over 10,000 pulses collected corresponding to a set energy window. Solver provided by Microsoft Excel is used to determine the fractional luminosities and decay time constants, using weighted least square principle. Figure 3 shows the model fit applied to an average pulse. The weighted difference between the average and the model is minimized to obtain an optimal solution for the unknown parameters. The results for alpha and gamma decay constants and the ratios of luminosities for fast and slow decay are presented through Table 2. These results corroborate published values for γ- ray excited CsI:Tl.

A Monte Carlo model was developed in order to generate a pulse shape spectrum of the luminosity ratios and decay constants corresponding to alpha and gamma radiation over a range of energies. Figure 4 is theoretical pulse shape spectrum. Crystal Ball provided by Decisioneering is used to obtain the probable distribution of luminosity ratios in the individual decay modes. The model was used to correlate the relative intensity and the decay time constants to the pulse shape discrimination capability.

3) Development and characterization of an automated radiochemical separation scheme utilizing SuperLig 620

Successful analysis of 90Sr in nuclear waste processing streams is critically dependent on rapid and reliable separation of 137Cs and 137mBa from 90Sr. HLW process streams will have high activities of 137Cs/137mBa that will interfere with quantification of 90Sr, unless high decontamination factors are achieved by chemical separation. IBC Advanced Technologies (American Fork, Utah) has developed a strontium-specific solid-phase extraction material available under the trade name Superlig 620. This material is comprised of a strontium-selective macrocyclic functionality covalently attached to the silica-gel support. The loading and elution characterization of the SuperLig 620 for HLW separation has not been documented in the literature and was the focus of this study. Batch test uptake studies of strontium/cesium/barium onto SuperLig 620 determined that the optimum uptake of the 90Sr was in 2 M nitric acid. Ammonium citrate was discovered to be a suitable complexant for the removal of strontium and barium from the SuperLig 620. Using a combination of forward flow and reverse flow in a small column packed with SuperLig 620, chromatography separation of strontium and barium in a high level waste tank stimulant was demonstrated.

4) Development of the ZnS:Ag scintillation detector for quantification of alpha-emitters in liquids

A comparison and contract of the silicon diode and ZnS:Ag detection methodologies for the purpose of total alpha analysis in process matrices was investigated. The goal is to be able to selectively quantify the alpha radiation in the presence of high levels of beta- and gamma-emitting radionuclides, which are likely to be present in HLW process monitoring applications. A thin ZnS:Ag scintillation detector was investigated to determine if there is any advantage of that detector over the silicon diode that was investigated in Phase I. Specific issues include the detection efficiency as a function of alpha energy, the selectivity of the detector to only alpha radiation, and the minimum detectable activity. The disadvantage of the ZnS:Ag over the silicon detector is that the ZnS:Ag detector does not provide individual isotopic information.

DELIVERABLES: Ayaz, B., DeVol, T.A., “Experimental-Theoretical Response of ZnS(Ag) Scintillating Discs for Gross Alpha-Measurements of Aqueous Radioactivity,” IEEE Transactions on Nuclear Science, Vol. 51, No. 4, 1688-1692 (2004).

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