Measurement of Radionuclides Using Ion Chromatography and Flow-Call Scintillation Counting with Pulse Shape Discrimination - Tasks 1.11, 1.12 and 1.13

Topical Report

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Office of Environmental Management
Office of Technology Development
1000 Independence Avenue
Washington, DC 20585

U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
P.O. Box 880
Morgantown, West Virginia 26507-0880

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By
South Carolina Universities
Research and Education Foundation
Strom Thurmond Institute
Clemson, South Carolina 29634-5701

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SUMMARY

Evaluation of Alpha/Beta Pulse Shape Discriminating Flow-Cell Radiation Detectors Using BGO, GSO:Ce, YAG:Ce, CsI:Tl, CaF₂:Eu and ZnS:Ag

The objective of Task 1.1 is development of a flow-cell scintillation detector with pulse shape discrimination. When coupled with an existing ion chromatography, the resulting system will be used to measure non-gamma-ray emitting radionuclides in environmental samples.

Several flow-cell radiation detector systems are commercially available for quantification of aqueous radioactive solutions. These systems do not use the technique of pulse shape discrimination to identify the incident radiation and therefore are limited in environmental characterization application when coupled to an ion chromatography system. The advantages of the pulse shape discriminating flow-cell detector over the commercially available systems include: 1) lower minimum detectable activity for alpha radiation, 2) reduced radiological interferences that may exist between co-eluted alpha- and beta- emitting radionuclides, and 3) possible isotopic information from the ion chromatography system. For Tasks 1.1.1 and 1.1.2 of this project, several scintillation materials were investigated for pulse shape (alpha and beta) discrimination capabilities and the best candidate material was optimized. In addition, the following detector properties were also optimized: scintillator particle size, flow-cell tubing type, and electromagnetic as well as optical crosstalk between the photomultiplier tubes.

The flow-cell detection system was modeled after commercially available flow-cell radiation detectors and pulse shape discriminating liquid scintillation counters. The system was constructed of dual photomultiplier tubes which were used to convert the scintillation photons (induced from the radiation) into electronic signals. Fast and slow signals from the photomultiplier tubes were processed. The "fast" signals were required to be in coincidence before the slow signals were recorded. The slow signals were split to allow simultaneous quantification of the pulse height (energy deposited) and pulse shape (type of incident radiation) data. These slow data could be recorded separately to yield the single parameter pulse height or pulse shape spectra, or simultaneously to yield a dual parameter spectrum.

The flow-cells typically contained 63-90 μm inorganic scintillator particles packed into a translucent tube. The aqueous radioactive solution flows through the interstitial pore spaces of the scintillation material. The scintillator must have high luminosity and produce pulse shapes that are conducive to alpha/beta discrimination. In addition since the scintillator comes in direct contact with the aqueous solution, it must also be inert, insoluble, and non-hygroscopic. Lastly, the scintillator should be commercially available. Of the 21 different scintillation materials (organic and inorganic) [Table I; Branton, 1996] that were evaluated with the above criteria, seven flow-cell configurations were investigated using five different scintillators [Table III; Branton, 1996]. The seven flow-cells that were investigated in detail were: BGO, GSO:Ce, YAG:Ce, uncoated CaF₂:Eu, fluorescent coated CaF₂:Eu, combination flow-cells of coated and uncoated CaF₂:Eu with ZnS:Ag, and a non-fluorescent polymer coated CsI:Tl. The single scintillator flow-cells of BGO, GSO:Ce, YAG:Ce and uncoated CaF₂:Eu had poor to moderate pulse shape resolution, with a figure of merit (FOM) ranging from 0.18 to 0.44, respectively. Their pulse shape resolution were evaluated quantitatively using the FOM which is a parameter used to determine the resolution of alpha and beta events in the pulse shape spectrum [Equation 2; Branton, 1996]. A FOM = 2.0 indicates near optimum separation of alpha and beta events that are gaussian distributed. With the single scintillator flow-cells not having adequate pulse shape discrimination, a flow-cell constructed of a fluorescent polymer coated CaF₂:Eu was evaluated. This "phoswich" flow-cell did not perform as

expected for one or more of the following reasons: 1) the fluorescent coating was inefficient at producing light, 2) the coating was not thick enough to completely stop the alpha radiation, and 3) the temporal distributions of the scintillation light were insufficient to discriminate between the two scintillators. Another "phoswich type" flow cell was attempted by using two dissimilar scintillator materials and particle sizes. The concept behind the combination flow-cell was to have one scintillation material with a high efficiency for alpha particles and a low efficiency for beta particles and vice versa for the other scintillator. Although a "phoswich" style sensor is well grounded theoretically, the response of the coated and uncoated $\text{CaF}_2\text{:Eu}$ (the "beta only" scintillator) was too high for alpha particles, resulting in at least 10% spillover of alpha radiation into the beta channel. The 10% spillover which resulted from an alpha response from the "beta only" scintillator was considered too high for our application. The flow-cell design that was most successful was a thin polymer coated CsI:Tl . The coating was deposited onto the CsI:Tl to reduce the rate of its dissolution. The coating thickness was approximately 3 μm , which is sufficiently thin to allow the alpha radiation to pass and transparent enough to allow the scintillation light to escape. The pulse shape resolution from this flow-cell design was quite good ($\text{FOM} = 1.4$), and the spillover of alpha to beta and beta to alpha was less than 5%, one of the project goals. The detection efficiencies (for a 1 hour flow measurement) were 5%, 53% and 65% for ^{14}C , $^{90}\text{Sr}/^{90}\text{Y}$ and ^{233}U , respectively. The detection efficiencies for $^{90}\text{Sr}/^{90}\text{Y}$ and ^{233}U met project goals, but the ^{14}C efficiency was lower than the goal of 50%. The lower ^{14}C detection efficiency was the result of energy loss in the coating. The beta and alpha background count rates were 0.17 and 0.004 counts per second, and the corresponding minimum detectable activities for a 30 second counting time were 11, 1 and 0.2 Bq for ^{14}C , $^{90}\text{Sr}/^{90}\text{Y}$ and ^{233}U , respectively. Although the ^{14}C detection efficiency was lower than project goals, the alpha and beta backgrounds were lower also, resulting in the ^{14}C MDA being consistent with project goals and the ^{233}U MDA exceeding the goals by a factor of 2.

If pulse height information (recorded on the abscissa) is reported simultaneous with pulse shape information (recorded on the ordinate) the result is a dual parameter spectrum. Figure 1 illustrates a dual parameter spectrum obtained with aqueous $^{90}\text{Sr}/^{90}\text{Y}$ and ^{233}U in the polymer coated cesium iodide scintillator flow-cell. The upper region is the $^{90}\text{Sr}/^{90}\text{Y}$ and the lower thinner region is the ^{233}U . With the dual parameter analysis, the spillover is reduced to less than 1%. Figure 1 illustrates the first heterogeneous pulse shape discriminating flow-cell of its kind. These were very encouraging results midway through the project, but there is still work to be done. The polymer coating is not completely impermeable to water, causing the detector to degrade. This is reflected in a decrease in efficiency by a factor of two within the first five hours of operation. Additionally, the analysis of dual parameter data is difficult with existing software. The need to acquire in dual parameter data could possibly be circumvented by development of a digital pulse shape discrimination circuit which should reduce the alpha tailing into the beta window. This would allow a single discriminator level to separate alpha and beta events. And lastly, development of a homogeneous pulse shape discriminating flow-cell radiation detector should further reduce our already low minimum detectable activity to 1 Bq and 0.1 Bq for low energy beta, and alpha, respectively.

Upon completion of the pulse shape discriminating flow-cell system, the next step is to couple it with the ion chromatography system for simultaneous quantification of alpha- and beta- emitters that have been separated elementally by the ion chromatography system. The combined system will allow for simultaneous quantification of alpha- and beta-emitting contamination in environmental samples.

Branton, S.D., "Evaluation of Alpha/Beta Pulse Shape Discriminating Flow-Cell Radiation Detectors Using BGO, GSO:Ce, YAG:Ce, CsI:Tl, $\text{CaF}_2\text{:Eu}$ and ZnS:Ag ", M. S. thesis, Clemson University, 1996. (document attached)

Figure 1. Dual Parameter Spectrum of the polymer coated CsI:Tl pulse shape discriminating flow-cell acquired with an aqueous solution of $^{90}\text{Sr}/^{90}\text{Y}$ and ^{233}U . Single parameter Pulse Height and Pulse Shape Spectra are shown as projections.