Coincidence methods in gamma-ray spectrometry for radioecological applications - DTU Orbit (07/04/2019)

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Gamma spectrometry is one of the most powerful radiometric techniques available. The nondestructive method enables both quantitative determination and identification of the majority of radioisotopes. Compared to other radiometric techniques, it has a great advantage in being able to detect minor isotopes, even in the presence of a large background from a multitude of other radioactive elements without any need for separating the isotopes. This has enabled the technique to be used as the standard tool in nearly all disciplines where radioisotopes are analyzed.

The technique plays an important role in environmental radioactivity, nuclear safety and reactor monitoring, nuclear medicine, isotope geology… Gamma spectrometric analysis of artificial radioisotopes in man (whole body counting) has improved the understanding of human metabolism, while the analysis of the very same radioisotopes in sea water has shed light on Arctic Ocean water circulation.

All of this is thanks to a deliberate and continuous effort to improve the technique over the years. With improvements in energy resolution, detector size and performance, coupled to better background reduction, studies of new phenomena in environmental radioactivity have become possible. Not seldom have these improvements occurred suddenly though discrete events. The introduction of digital signal processing in gamma spectrometry is definitely one such event, and it will, in the next coming years, revolutionize the way in which we acquire information through environmental gamma spectrometry.

Up till now, gamma spectra were measured in a way that the energy deposited in a detector was measured with an analogue chain (preamplifier, amplifier, ADC), events were saved in computer memory with its energy and arranged into a histogram called spectrum. With digital list-mode systems each event is saved with its energy and time-stamp when the event happened. In simple words, the difference between the standard gamma spectrum and time-stamped list-mode file can be compared to a difference between the long exposition photography and a video.

Coincidence gamma spectrometry exists from the early-days of nuclear research, but the complexity of such systems usually limited its use to large experiments or highly specialized applications. The systems needed delicate tuning for each particular experiment, and once the system was set-up and working, making changes was a cumbersome procedure. Often a change of a single cable (in a fast signal branch) made the system not working. With digital list mode gamma spectrometry, once the acquisition parameters are adjusted and the sample is measured, all the coincidence settings can be tested in post-processing. That means if different coincidence timing or energy gating is needed, it takes only minutes to generate a new spectrum, in contrast to ‘standard’ approach where the new measurement needed to be done (often a long time measurement if we’re dealing with low activity levels). That significantly simplified the process of setting up coincidence experiment. Widespread use of the field-programmable gate array (FPGA) technology led to reduction in size and price. A single digital unit replaced a whole set of special analogue units (like CFD, TAC, Coincidence unit, delay unit, shaper…). Small physical size enables integration of coincidence system to mobile (or hand-held) instruments. All that will make changes to gamma spectrometry in coming years, which cannot even be foreseen. Even now some producers are completely stopping the manufacturing of standard analogue NIM bin modules, although the use of time-stamping is still at its start. Developments of IEC standard for list-mode data acquisition will certainly speed-up the things by making the implementation of the new technology into laboratory even easier. The standard will make the implementation easy, enabling universal coincidence acquisition and analysis software, in contrast to current approach where the most groups are developing its own software.

This thesis reveals some promising aspects of digital-list mode acquisition systems when applied to gamma spectrometry, from low-level measurements, where it can be used with veto detectors or multiple HPGe detectors for background reduction and efficiency enhancement, to measurements of high activity levels where, in some cases, coincidence signals with narrow energy gating, enable extraction of weaker signal hidden in high activity matrix.

The use of a sum-coincidence mode resulted in 17% efficiency increase. Summing of coincident events energies reconstructed the full energy of a photon Compton scattered between two detectors. Applying anticoincidence setting enables better sensitivity for 210Pb determination by reducing background continuum for ~15%. The two abovementioned methods can be applied also for low or ultra low-level measurements. For high activity samples, narrow energy window gates combined with coincidence gating resulted in almost complete background reduction, revealing the 605 keV 134Cs peak under high 137Cs background. This approach seems promising for determination of impurities in radiopharmaceuticals or characterization of decommissioning samples.

Application of digital systems in activity standardization measurements with liquid scintillation counting (LSC) has become a standard, but its introduction to photon-photon coincidence techniques is still pending full recognition. The last chapter gives some reasoning on possible ways how it could be done. Primary standardization method for 125I using two NaI(Tl) detectors has been set-up at the Radioecology Section. New standardization method for 125I based on two HPGe detectors has been developed. The method, although inferior in precision compared to NaI(Tl) method, has an advantage of not relying on total count rate measurement allowing 125I activity standardization in the presence of impurities. Review of 60Co standardization method is presented with a theoretical solution for extension to 134Cs gamma-gamma standardization.

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