

Analysis of Long-lived Isotopes in the Presence of Short-lived Isotopes Using Zero Dead Time Correction

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Abstract-- High Purity Germanium (HPGe) detector systems are routinely used in counting laboratories in many types of nuclear facilities such as nuclear power plants and fuel production sites. These systems generally consist of a lead-shielded HPGe detector, Multi Channel Analyzer (MCA), and analytical software. These systems are used to analyze a wide variety of sample types for many different isotopes. Analysis of certain sample types, such as those from the reactor coolant or the off-gas extraction system, in nuclear power plant radiochemistry laboratories is complicated by the presence of short-lived isotopes. With these isotopes present, the sample count rate begins at a higher value than the ending count rate with a rapid change in count rate often observed. This decaying of the sample count rate causes the true count rate of the peaks to be unknown for those MCAs that use the traditional Live Time Clock extension methods. The current method of compensating for these short-lived isotopes is simply to delay starting the acquisition until these isotopes decay (typically 45-60 minutes). This has the effect of reducing the throughput capacity of the laboratory meaning fewer samples can be counted in any given period. The use of "loss free counting" methods in radiochemistry laboratories has been unacceptable because these methods do not provide the uncertainty in the measurement which must be reported with the activity calculation from the counting laboratory. An innovative MCA with a zero dead time (ZDT™) correction method will be presented which (1) compensates for the decaying count rate caused by the short-lived isotopes, thus eliminating the need for delaying the start time of the acquisition; and (2) calculates the uncertainty in the activity determination, thus satisfying the reporting requirements of the counting laboratory. Data from the analysis, including the uncertainty, of long-lived isotopes in

reactor coolant samples both in the presence and absence of short-lived isotopes will be presented.

I. BACKGROUND

In traditional Multi Channel Analyzers (MCA), a Live Time Clock (LTC) is used to determine the amount of time associated with the counting of pulses from, among other things, High Purity Germanium (HPGe) detectors. The MCA and HPGe detector system is widely used in counting laboratories for Health Physics, Environmental, and Radiochemistry sample analyses at nuclear facilities including nuclear power plants. These systems are used to collect information from a wide variety of sample types, typically dependent on the purpose of the facility or on governing regulations. Some sample types in the counting laboratories contain a mixture of long-lived and short-lived isotopes. A persistent analytical problem exists when attempting to quantify either the long-lived or short-lived isotopes in these samples. The problem is due to the inaccuracy in the LTC. In its simplest form, the activity of an isotope is determined by Equation 1.

$$Activity = \frac{GrossCounts - Background}{LiveTime * Efficiency * Yield} \quad (1)$$

If the Live Time is not accurately known, then the activity can not be determined. It has previously been shown that LTC-based MCA's do not yield accurate results for the system live time for samples that have rapidly changing count rates during the acquisition period such as would be the case for short-lived isotopes. There have been MCAs developed by Harms and others that correct for the dead time losses and are known as Loss Free Counting modules. These methods, however, produce spectral data which no longer obey Poisson counting statistics. Therefore, these methods can not be applied to the routine analysis of counting laboratory samples since the uncertainty associated with the analysis can not be determined from the spectra collected by Loss Free Counting modules.

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To compensate for the changing dead times in the system, then, the counting laboratory operators allow the short-lived isotopes to decay prior to acquiring the spectrum. There are three problems with this solution when applied to counting rooms: (1) the sample can not be counted immediately, (2) the operator must be trained to know how long is “long enough” to decay the sample, and (3) the short-lived isotopes can not be quantified.

In 2000, ORTEC® introduced the first integrated MCA which uses a loss free counting method called Zero Dead Time (ZDT™). It has been previously shown that the ZDT mode of the DSPECPLUS accurately correct for dead time losses. ZDT overcomes the deficiencies in both the LTC-based MCAs and previous loss free counting modules by dynamically correcting for dead time losses and accumulating a variance spectrum. In this way, the uncertainty of the analysis can be accurately quantified.

The use of this technique (patent pending) in the DSPECPLUS extends the concept of loss free counting to the routine analyses in counting laboratories and in particular for samples such as those described above. In this work, we will analyze spectra taken with the DSPECPLUS in both ZDT and LTC modes for a variety of input count rates from samples taken from the reactor coolant water of the Olkiluoto nuclear power station in Finland. Comparison of the ZDT and LTC analytical results will show that the ZDT method improves the results from these laboratories.

II. EXPERIMENTAL SETUP

The radiochemistry laboratory in the Olkiluoto nuclear power plant has a DSPECPLUS and ORTEC p-type (GEM-series) HPGe detector. The Windows NT computer system runs GammaVision 5.30 to acquire, store, and analyze the spectral data. A JOB automation file was created within GammaVision to perform the following functions:

1. Set the DSPECPLUS into ZDT Mode
2. Collect a spectrum for preset count time (e.g., 2000 seconds)
3. Store the spectrum to disk
4. Set the DSPECPLUS into LTC Mode
5. Collect a spectrum for the same preset Real Time (e.g., 2000 seconds)
6. Store the spectrum to disk

A reactor coolant water sample was then extracted from the sampling point and brought to the laboratory for analysis. The sample was immediately placed on the detector and the JOB file run. In previous sample analyses, the reactor water would be allowed to decay for 30 to 60 minutes prior to data being collected using the LTC mode. In this case, the “decay time” is utilized to acquire the ZDT spectrum.

Multiple samples are collected and analyzed which have varying levels of activity. The relative activity of the sample can be quantified by looking at “Input Count Rate” (ICR) recorded in each sample spectrum in each of the two modes (ZDT and LTC). The value of the ICR is recorded at the end of the acquisition period. The ICR for the ZDT spectra are always higher, in some cases by a factor of 2, than the LTC spectra because the short-lived isotopes have not decayed.

After the data is collected for both ZDT and LTC modes, the GammaVision analysis is run on each spectrum for a preset library of isotopes. The isotopes for analysis were chosen to be:

Isotope	Half-life
I-132	2.3 hours
Mn-56	2.6 hours
Co-60	5.3 years
Co-58	70.8 days
Cr-51	27.7 days
Sr-92	2.7 hours
Na-24	15.0 hours

These isotopes are chosen because they were positively identified and have sufficient activity in each sample to present low counting uncertainties for the acquisition period.

The results of the analyses were then compared. The analysis of the LTC spectrum, which has been decayed before acquisition, is considered the “true” activity since after the decay of the short-lived isotopes the spectrum collected with a live time clock is accurate. The activity of these isotopes is then shown to be the same as the results from analyzing the ZDT spectrum within the reported statistical uncertainties.

III. RESULTS

Tables 1-6 below show the results of the six (6) dual-spectra sample analyses.

TABLE 1

	LTC			ZDT			Δ
	Activity Bq/ml	ICR = 1407		Activity Bq/ml	ICR = 1575		
		Uncertainty (2 σ)			Uncertainty (2 σ)		
	Counting	Total	Counting	Total	Total		
CR-51	29.4	3.0%	3.8%	28.1	2.5%	3.4%	-4.3%
CO-58	6.5	1.9%	3.0%	6.5	1.9%	3.0%	0.3%
CO-60	5.0	2.0%	3.1%	5.1	1.8%	2.9%	1.8%
NA-24	1.5	5.9%	6.4%	1.6	3.9%	4.5%	1.1%
MN-56	19.5	5.0%	5.5%	19.6	4.6%	5.1%	0.1%
SR-92	26.6	2.0%	3.1%	26.4	1.8%	2.9%	-0.5%
I-132	27.0	1.5%	2.7%	26.8	1.3%	2.7%	-0.6%

TABLE 2

	LTC			ZDT			Δ
	Activity Bq/ml	ICR = 2914		Activity Bq/ml	ICR = 4603		
		Uncertainty (2 σ)			Uncertainty (2 σ)		
	Counting	Total	Counting	Total	Total		
CR-51	64.2	4.7%	5.3%	63.3	11.3%	11.6%	-1.5%
CO-58	13.8	2.3%	3.3%	13.3	2.9%	3.7%	-3.1%
CO-60	10.1	3.6%	4.3%	10.7	5.1%	5.6%	5.4%
NA-24	3.0	6.7%	7.1%	3.1	6.4%	6.8%	4.0%
MN-56	39.4	3.6%	4.3%	38.9	3.7%	4.4%	-1.2%
SR-92	56.1	1.8%	2.9%	57.9	1.9%	3.0%	3.2%
I-132	56.3	1.1%	2.6%	55.4	1.1%	2.6%	-1.7%

TABLE 3

	LTC			ZDT			Δ
	Activity Bq/ml	ICR = 1535		Activity Bq/ml	ICR = 2341		
		Counting	Total		Counting	Total	
CR-51	9.9	14.5%	14.7%	10.1	10.4%	10.6%	-1.4%
CO-58	5.2	2.0%	3.1%	5.3	2.2%	3.2%	-1.7%
CO-60	3.3	3.9%	4.6%	3.2	3.6%	4.3%	-3.7%
NA-24	2.7	4.3%	4.9%	2.7	4.9%	5.5%	-0.7%
MN-56	14.8	4.8%	5.3%	14.8	4.2%	4.8%	-0.2%
SR-92	6.4	3.4%	4.1%	6.6	3.1%	3.9%	3.9%
I-132	1.8	5.3%	5.8%	1.8	4.7%	5.2%	-3.0%

TABLE 4

	LTC			ZDT			Δ
	Activity Bq/ml	ICR = 663		Activity Bq/ml	ICR = 834		
		Counting	Total		Counting	Total	
CR-51	11.2	5.9%	6.3%	11.3	5.2%	5.7%	-1.1%
CO-58	5.0	1.4%	2.7%	4.9	1.5%	2.8%	-1.2%
CO-60	3.2	1.8%	2.9%	3.1	1.6%	2.8%	-0.9%
NA-24	2.2	2.4%	3.3%	2.3	3.6%	4.3%	5.0%
MN-56	13.8	1.6%	2.8%	13.5	3.7%	4.3%	-2.4%
SR-92	6.2	3.1%	3.8%	6.3	2.7%	3.6%	-1.9%
I-132	2.2	4.6%	5.1%	2.3	3.5%	4.2%	4.0%

TABLE 5

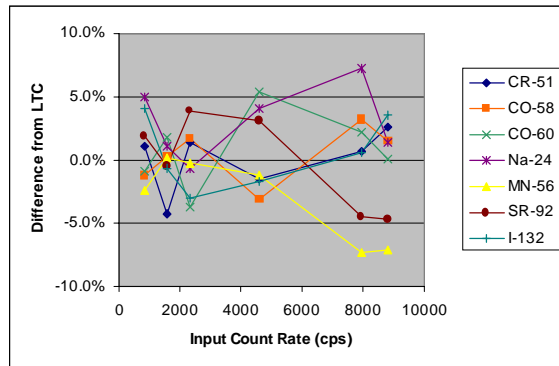
	LTC			ZDT			Δ
	Activity Bq/ml	ICR = 3772		Activity Bq/ml	ICR = 7977		
		Counting	Total		Counting	Total	
CR-51	75.4	4.3%	4.8%	75.9	9.4%	9.6%	0.7%
CO-58	3.5	4.1%	4.7%	3.6	4.6%	5.1%	3.2%
CO-60	2.8	4.2%	4.8%	2.9	5.1%	5.6%	2.3%
Na-24	1.6	5.6%	14.7%	1.7	7.2%	15.4%	7.2%
MN-56	37.2	1.1%	2.6%	34.4	1.4%	2.7%	-7.4%
SR-92	18.6	2.6%	3.5%	17.8	2.4%	3.3%	-4.4%
I-132	15.1	1.7%	2.9%	15.2	2.0%	3.1%	0.6%

TABLE 6

	LTC			ZDT			Δ
	Activity Bq/ml	ICR = 4619		Activity Bq/ml	ICR = 8797		
		Counting	Total		Counting	Total	
CR-51	21.5	9.8%	14.2%	22.0	13.7%	17.1%	2.7%
CO-58	1.8	5.7%	11.8%	1.8	6.0%	11.9%	1.5%
CO-60	1.8	6.6%	12.2%	1.8	7.7%	12.8%	0.1%
NA-24	0.9	7.6%	12.8%	0.9	9.1%	13.7%	1.4%
MN-56	4.3	9.8%	14.2%	3.9	10.5%	14.7%	-7.1%
SR-92	9.8	3.8%	11.0%	9.3	3.7%	10.9%	-4.6%
I-132	6.5	2.6%	10.6%	6.8	2.8%	10.6%	3.5%

Figure 1 below shows the ratio of the ZDT to LTC data from Tables 1-6 plotted by isotope against the ending value of the Input Count Rate in the ZDT spectrum acquisition.

Figure 1



IV. Discussion and Conclusions

In Tables 1-6 it is clear that the analytical results obtained from the ZDT-corrected spectra are in agreement with those in the decayed LTC spectrum. This shows that the ZDT acquisition mode in the DSPECPLUS can be confidently used to analyze, quantify, and report the activity of long-lived isotopes even in the presence of the fast-decaying, short-lived isotopes in the same sample.

In Figure 1 it is shown that the use of the ZDT-corrected spectrum is accurate across a wide range of input count rates. This system is configured with a relatively long rise time (12 μ sec) in order to give the best resolution possible in the spectra. In the LTC spectrum, an input count rate of 4600 cps is roughly 23% dead time in the spectrometer which is generally higher than normal for routine radiochemistry sample analyses. Some ZDT spectra were collected at count rates more than double this value.

All but four (4) of the data points show differences of less than 5% and all are less than 7.4% from the known value. There does not appear to be any bias in the analytical results either on a per isotope or per ICR basis.

The results obtained here for the specific case of the reactor water samples at a nuclear power plant is justification of the wider use of this technique for other sample types in other applications. In summary, it has been shown that the ORTEC DSPECPLUS in combination with the analysis capabilities in GammaVision-32 can be used to solve the complex analytical problem of the influence of short-lived, fast-decaying isotopes in counting laboratories.

V. REFERENCES

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