In-Situ Sample Analysis with Portable Gamma Spectrometers

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Abstract. This paper presents methods for conducting in-situ spectroscopic measurements using 3DCZT instruments. These devices are capable of excellent energy resolution on the order of 1% FWHM at 662 keV and around 5% relative photopeak efficiency compared to a 7.6mm cylindrical NaI device. They also provide list-mode data with the energies and interaction positions of each gamma-ray event that enable the use of event filters that would not be possible in a conventional spectrometer. This work looks at the use of the spectrum of events that interact only once and compares the spectral quality to the spectrum of all events. The impact on peak tails, pileup, and escape peaks is studied for the single interaction spectra for all the commercially available 3DCZT platforms. Dramatic improvements in pileup rejection are demonstrated for all platforms.

KEYWORDS: 3DCZT, CdZnTe, In-Situ Spectrometer, Gamma-ray Spectroscopy

1 INTRODUCTION

In-situ spectroscopic innovations related to CdZnTe [1] have enabled higher resolution embedded counting than was available to previous generations of nuclear scientists. This has enabled the nuclear industry to conduct monitoring programs in environments that would otherwise be impracticle. One example of the benefit of this work is the ability to monitor Ag-110m concentrations in the primary loop of a nuclear reactor. Historically the silver was detected via water radiochemistry but due to its strong desire to plate on pipes the water sampling typically indicates much lower levels of Ag-110m. Therefore, these in-situ tools proved a much more effective tool to monitor crud buildup which is a major source of radiation dose to plant workers.

This work is looking at the latest generation of high resolution in-situ spectrocopic monitoring tools in an attempt to take this work to the next logical step: determination of operationally relevant trace quantities of isotopes of interest. Large-volume high purity germanium detectors are a good choice for this application from a technology capability standpoint but it is challenging to package and maintain a reasonably large volume of HPGe for in-situ monitoring. The next closest thing to HPGe in terms of performance is 3D position sensitive CdZnTe [2], or 3DCZT, which is owned in one form or another by over 75% of the US commercial nuclear power generating stations so it is already pervasive in the field. The 3DCZT technology is much better suited for in-situ measurements from a practical standpoint due to its room temperature operation, compact size, and good performance at high count rates. With energy resolution of 1% FWHM at 662 keV or better [3] 3DCZT provides spectral performance than any HPGe alternative. The efficiency of the 3DCZT devices is greatly improved over previous generations of in-situ portable spectrometers, such as the Inspector 1000, due to the much larger crystal size. For example, the crystal volumes presented in this work range from 10 to 50 times larger than the crystal in the Inspector1000 which was the standard for portable in-situ measurements in nuclear power facilities [1].

This paper aims to understand the limits of commercially available solutions that could address the in-situ monitoring of relatively weak isotopes. One aspect of these systems that is unique compared to conventional spectrometers is that the readout configuration is much more complicated and a dominant factor in influencing the overall spectral performance. Therefore, the impact of using different readout electronics can be drastic in some applications. Furthermore, the 3D position information available for all interactions allows the rejection of event sequences that are detrimental to overall spectral quality.

One of the most common challenges in the quantification of multiple isotopes is the independent determination of the net area from two adjacent peaks. Examples include Ag-110m in the presence of

Cs-137, where the Cs-137 peak near 662 keV is influences by the Ag-110m peak near 657 keV. Likewise both the of the primary Fe-59 emissions are on the low energy tail of the Co-60 emissions. These are scenarios where it may be well worth the reduction in counts in order to more easily determine the net area of each peak or to at least to identify the presence of more than one peak. This work studies the tradeoffs between different readout configurations and filters to present the readers with best practices for applying these tools to their application.

2 MEASUREMENT DESCRIPTION

2.1 Detector Platforms Used for Studies

This work focuses on the commercially available 3DCZT detector architectures that are strong candidates for in-situ spectroscopic monitoring. These architectures are listed in Table 1. The "standard 3DCZT" technology is the most common configuration used at the time of this publication. The "high rate 3DCZT" architecture represents a minor iteration of the standard 3DCZT that is optimized for higher count rates but otherwise achieves very similar performance. The "high performance 3DCZT" is a more drastic departure because it leverages a different Application Specific Integrated Circuit (ASIC) based on a readout archicture where the entire preamplifier waveform is read out and passed to a digital signal processor [4] rather than relying on analog shaping and peak hold techniques [5].

Table 1: These are the architectures that were considered for this study.

Architecture	Max Count Rate	Max Dynamic Range	ASIC Output
Standard 3DCZT	30,000	3 MeV	Peak Amplitude & Time
High Rate 3DCZT	150,000	3 MeV	Peak Amplitude & Time
High Performance 3DCZT	10,000	9 MeV	Preamplifier Waveform

The "standard 3DCZT" architecture was the only commercial-off-the-shelf (COTS) 3DCZT architecture available between 2015 and 2018 and is still commercially available. It is the foundation of the commercial S, H, A, and P series detectors from H3D, Inc, therefore it was necessary to include in this work as it is the most common configuration a reader will encounter. The high rate 3DCZT system is included because in many applications the maximum count rate of 30,000 CPS is a major barrier to attaining better spectroscopic data on weak signals so the 150,000 CPS readout capability is expected to pull in a sizable user base over time. The high performance system is included because it is currently the only COTS option for 3DCZT systems that can examine higher energy gamma-rays such as the emissions from N-16. Also, the high performance system can achieve better energy resolution which is important for distinguishing weak signals and neighboring peaks.

The sensitivity of the architectures has been studied academically over the years, with the most recent focus on the high performance system [5,6]. One common theme even in modern work is that it is possible to get close to the expected efficiency given the gamma-ray interaction rate in the device but only if a relatively broad region of interest is used for the photopeak. In the case of the reasonably narrow photopeak regions of interest that are used in the commercial software the intrinsic efficiency is only about half of what it should be based on gamma-ray interaction rate at Co-60 energies. Therefore, the reader should consider that for all the detectors used in this work the relative photopeak absolute efficiency at 1333 keV (compared to a 7.6cm diameter and length cylinder of sodium iodide) is expected to be 5-10% based on the detector volume but is actually in the range of 2-5% after considering count losses due to charge collection problems in certain detector regions as well as the influence of the photopeak tail.

2.2 Count Rate Stability Studies

A representative detector was chosen for each of the three platforms shown in Table 1. For the standard 3DCZT measurements an H420 model from H3D was used which contains 19cm^3 of CdZnTe. For the High Rate 3DCZT measurements an M400 model from H3D was used which also contains 19cm^3 of CdZnTe. For the High Performance 3DCZT an S400X model from H3D was used and the largest possible crystals were inserted for a total detector volume of 29cm^3.

Each detector model was kept stationary throughout the tests. A Mn-54 source was kept stationary and unshielded throughout the analysis such that the count rate should be stable in the case of a perfect detector system. A strong Cs-137 source was then moved between each measurement to vary the amount of radiation interfering with the Mn-54 source. These sources were used to generate count rates high enough such that the live time of each system was less than half of the real time. The resulted in dead times that varied from a <10% to >50% for each system and the impact on spectral quality and stability was measured by looking at the net area of several peak regions of interest (ROIs) as well as the overall shape of the spectrum based on the influence of pileup.

2.3 Event Rejection Studies

Conventional spectrometers will report data in the form of a spectrum that shows the total number of counts per bin integrated over a period of time. The 3DCZT technology is based on a platform that was primarily developed for gamma-ray imaging purposes and records the data for each individual gamma-ray interaction in addition to the spectrum in order to enable the reconstruction of radiation images. This interaction information includes the energy deposited as well as the 3D location and time. A measurement can be reprocessed after the initial acquisition based on this list mode data and it is possible to omit events that are not of interest.

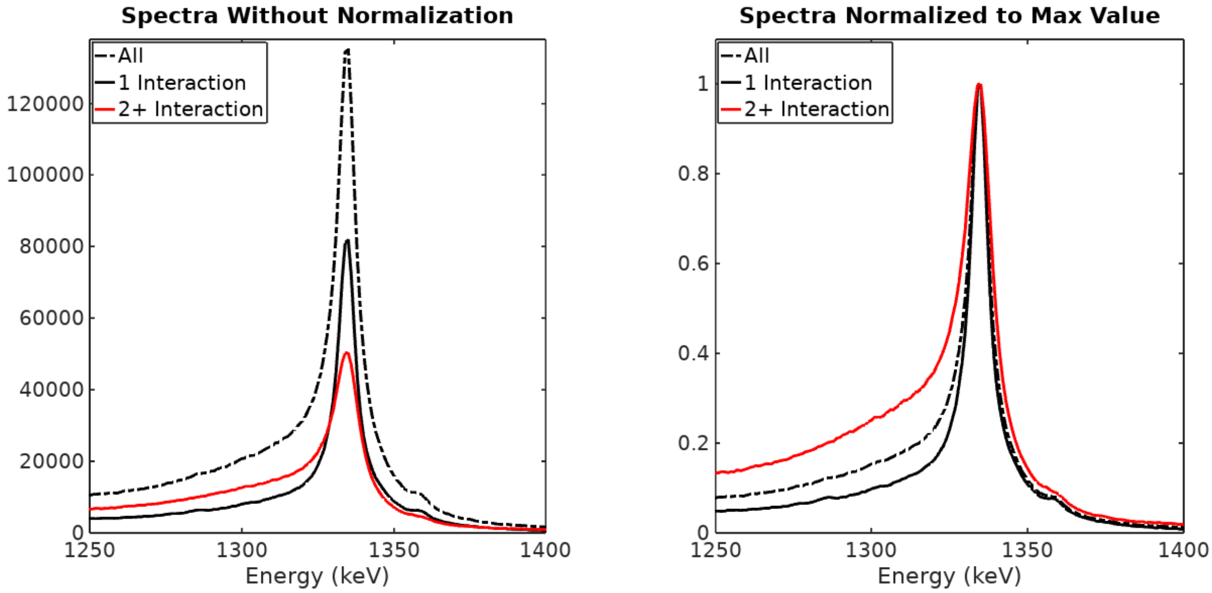
The list mode data files from various detectors are processed and are filtered out based on different parameters. The primary filter of interest is to eliminate events that involve more than one interaction. These multiple interaction events tend to have poorer signal quality than the single interaction events so the overall quality of the spectrum might actually improve by omitting the multiple interaction events. Another filter that is considered is the elimination of the signal from the pixels on the detector periphery which tend to have a poorer peak-to-continuum ratio than the central pixels.

3 RESULTS

3.1 Peak Tail Minimization

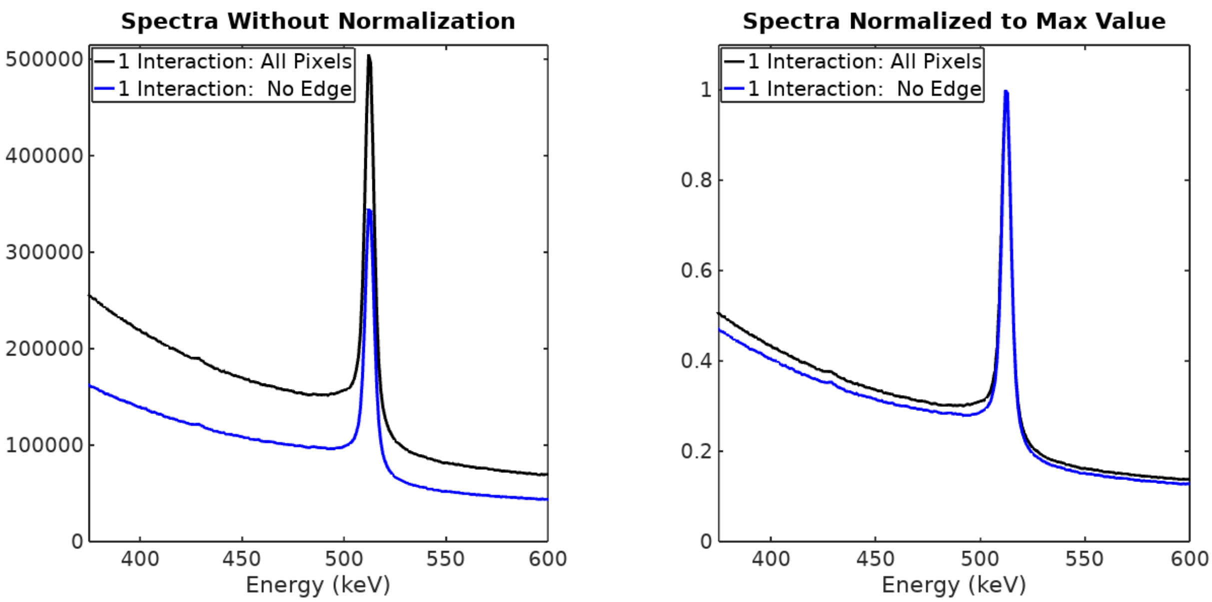
Figure 1 shows an example of the 1333 keV peak from Co-60 broken down by single interaction and multiple interaction events. The figure on the left shows the absolute number of counts and it is clear that the single interaction peak is at least 50% taller than the multiple interaction peak while the low energy tail from multiple interactions is at least 50% greater than the single interaction tail. Thus it is unsurpising to see a ratio of more than 2:1 across the entire low energy tail region between the multiple interactions and the single interactions when we normalize by peak height. In scenarios where the uncertainty due to counting statistics is secondary compared to systematic errors due to peak interference it is guaranteed that the user will get lower uncertainty if they use the single interaction spectrum due to the improvement in peak-to-tail ratio.

Figure 1: 1333 keV (Co-60) peak shape for all events combined, single interaction events, and multiple interaction events. Data was taken with an H420 (standard 3DCZT).



The events that occur near the edge of the detectors are also expected to show inferior performance compared to the average event because they may suffer charge collection issues from events interacting near the side surface and the escape probability of scattered gamma-rays is higher than for inner pixels. However, it would appear that these effects are generally small and it is unlikely that discarding approximately 1/3 of the counts in the detector (the edge pixels account for 40 out of 121 pixels in each crystal) will justify the small improvement in peak to continuum ratio. Figure 2 shows an example from a standard architecture detector and from the normalized data on the right it is clear that the peak shapes are very similar and the difference in tailing is closer to 10% compared to the factor of two that is seen in Figure 1. This coupled with the fact that the photopeak count rate loss is similar to the loss associated with moving from all events to one interaction strongly suggest that the best return on investment from an event filter will come from discarding multiple interaction events but retaining the edge pixels events.

Figure 2: 511 keV peak shape for single interaction events from all pixels (black) and from inner pixels only (blue). An H420 detector is used and the continuum is mostly due to Co-60.



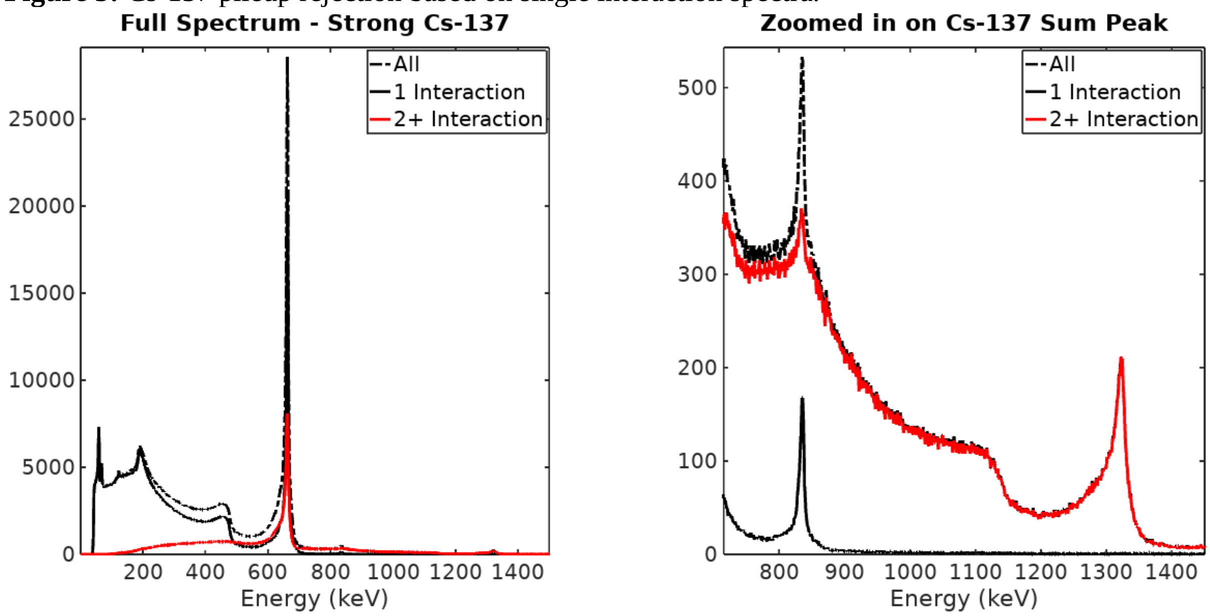
Note that all of these peak tail reduction results were repeated on the high performance and high rate platforms and similar behavior was observed: there is an approximate 2:1 improvement in peak height to tail ratio when looking at single interaction events and the returns on eliminating edge pixels are minimal.

3.2 Pileup Rejection

The pixelated nature of 3DCZT devices make them especially well equipped to reject pileup events. The spectrum of all events will have the same pileup behavior as is expected in any conventional spectrometer. However, the spectrum of single interaction events is expected to have approximately two orders of magnitude less pileup. This is because pileup is due to two random independent interactions that could occur anywhere in the device. Therefore, the probability that any two coincidence events appear as a single interaction is very low because both events would need to be close enough to get interpreted as a single interaction. Based on the spatial resolution of the device and interaction determination algorithm the probability of a chance coincidence showing up as a single interaction is approximately 1/100 for a four CZT crystal device such as H400, A400, or S400 and approximately 1/25 for a single CZT crystal device such as H100, S100, or P100.

Figure 3 shows the relationship between number of interactions and pileup. In this scenario there is a relatively weak Mn-54 source present and a strong Cs-137 source interfering with it. On the right side of Figure 3 the Mn-54 photopeak and Cs-137 sum peak are shown and it is clear that the vast majority of the counts in the sum peak are due to 2+ interaction events while the Mn-54 photopeak is mostly due to the single interaction spectrum. The single interaction spectrum is much better than the all events spectra in this case because it is less likely to have interference between the pileup features and the true photopeaks. This includes interference both in the form of the sum peak as well as the overall shape of the sum continuum which is mostly non linear and relatively hard to compensate for when finding net area.

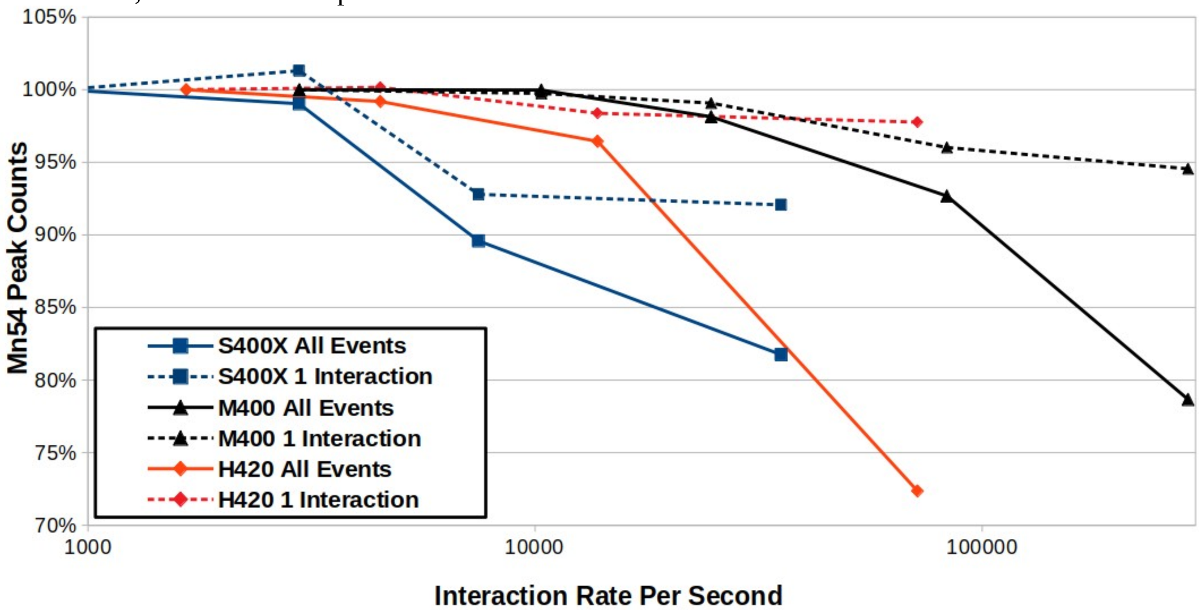
Figure 3: Cs-137 pileup rejection based on single interaction spectra.



The spectra shown in Figure 3 shows the impact of a strong Cs-137 source interfering with a Mn-54 source. This experiment was conducted at varying levels of Cs-137 source strength for all three of the architectures shown in Table 1. The net area of the Mn-54 photopeak was analyzed for each dataset for both the all interactions spectra ("all events" spectra) as well as the spectra of each interaction ("1 interaction" spectra). The net areas were determined based on the standard output from the software, H3D Visualizer, that is provided with the detectors (this net area is displayed when the user hovers over a peak). Each data curve is normalized based on the Mn-54 count rate at the lowest overall interaction rate for that detector, which is defined as 100% of the expected number of counts.

The results are shown in Figure 4. In the case of all three readout system types the single interaction spectrum provides measureably more stable net areas regardless of interaction rate, typically reducing the count rate deficit by a factor of two. The standard architecture (H420) showed the most significant improvement after switching to the single interaction spectrum which indicates this technique is relevant to many users of 3DCZT. The high rate system shows a similar pattern but the magnitude of the improvement is diminished. This is not surprising because the high rate system was not accounting for inter-module events (events that interact in multiple CZT crystals) when this data was taken which reduced the chance coincidence by a factor of four in the all events data. The rejection of intermodule events could be used as an intermediate step even in the standard architecture. This would be useful when the reduction of pileup by a factor of 4 is needed but the loss of up to half of the counts, which is the result of switching to single interaction spectra at some energies, is not acceptable. The intermodule events are less than 10% of the photopeak counts so this is a relatively small price to pay for a four times reduction in chance coincidence.

Figure 4: 834 keV (Mn-54) net area as a function of total gamma-ray interaction rate in the device. The S400X represents the high performance architecture, the M400 represents the high rate architecture, and the H420 represents the standard architecture.

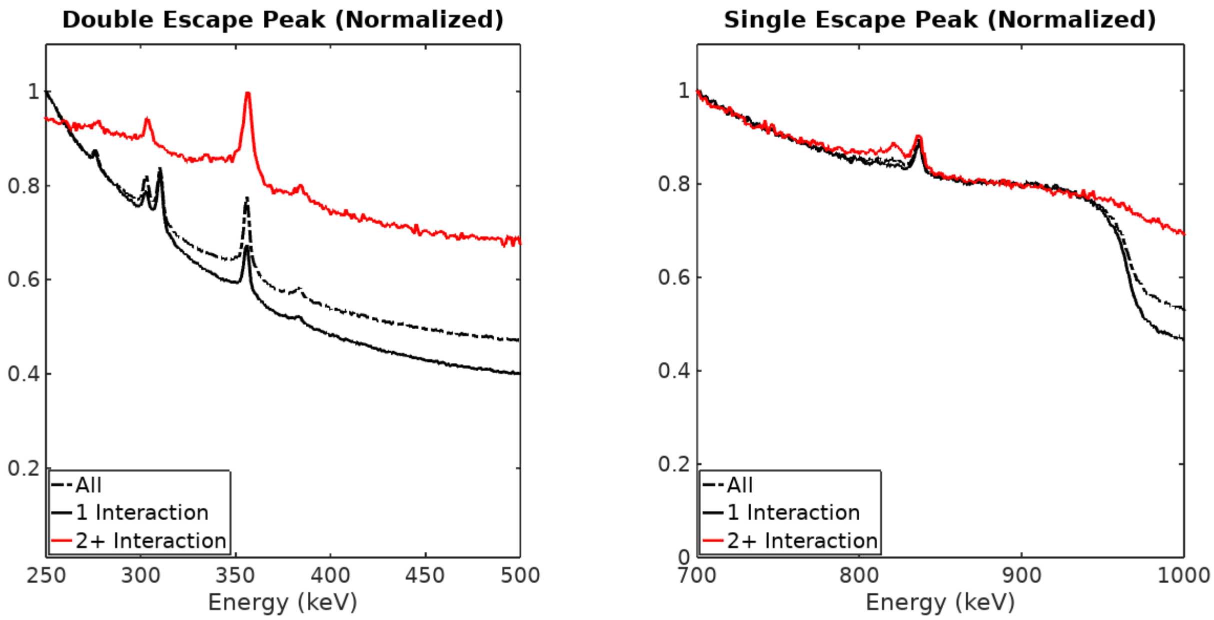


3.3 Escape Peak Behavior

One final issue that this work would like to bring attention to the attention of the reader is the impact of event selection on pair production spectral features. These are relevant because there is often a large quantity of Co-60 present in a nuclear facility relative to the quantity of Mn-54 present so the single escape peak at 822 keV can introduce error into the Mn-54 net area calculation. There is also a double escape peak at 311 keV which will interfere with the emissions from isotopes such as Cr-51.

Figure 5 shows the spectra of events from a strong Co-60 source with a relatively weak Ba-133 source present (left side) and a weak Mn-54 source present (right ride). In the case of the double escape peak there is a strong signal in the single interaction spectrum but there is almost no signal detected in the multiple interaction spectrum. This is because it is very unlikely that a Co-60 gamma-ray would undergo a Compton interaction first and then undergo pair production at a reduced energy. In the case of the single escape peak the behavior is reversed: the single interaction spectrum shows no hint of the escape peak that is strong in the multiple interaction spectrum. This is due to the fact that it is unlikely that the annihilation photon is absorbed close enough to the pair production location to get interpreted as a single interaction by the detector. This behaviour is repeatable across readout architectures.

Figure 5: Normalized spectra from an H420 in the presence of a strong Co-60 source. For the data on the left there is a Ba-133 source on top of the lower energy continuum region and on the right there is a Mn-54 source on top of the higher energy continuum region. The single and double escape peak detection probabilities are much different between single and multiple interactions.



4 CONCLUSIONS

This work has demonstrated that event filtration can be an effective tool for in-situ spectroscopy with 3DCZT devices. There are many in-situ counting scenarios where the count rates are high enough that counting statistics is a secondary effect compared to systematic errors due to interfering peaks or spectral features or count rate effects such as pileup. In those cases the 3D spatial resolution of 3DCZT detectors can be used to restrict the spectrum to the events that carry the information that will provide the most accurate and stable estimate of net area.

The elimination of multiple interaction events is a method that for the most part provides across-the-board benefits in high count rate scenarios. The downside of this approach is that for Co-60 energies about half of the photopeak counts are lost although these losses do decrease with decreasing energy; the multiple interaction probability for Co-57 is less than 10%. Additionally, the double escape peak is made stronger relative to continuum by the elimination of multiple interaction events. However, there are three main advantages to eliminating multiple interaction events:

1. The low energy tailing is reduced on photopeaks, especially at higher energies.
2. The chance coincidence probability is reduced by an order of magnitude or more.
3. The single escape peak magnitude is reduced by about an order of magnitude.

In many cases these advantages will outweigh the penalty from the poorer counting statistics and the reader should consider using the single interaction spectrum when using a 3DCZT device for in-situ measurements.

This work has also demonstrated that all of the 3DCZT architectures presented here can operate with an interaction rate greater than 50,000 per second while still achieving photopeak count stability within 10% of the expected value if the proper event filtration techniques are used. This is important for in-situ environments where the total count rate could change drastically during an event such as a crud burst in a nuclear facility and it is important that the individual isotopes are quantified correctly throughout the process. In this case the standard and high rate CZT architectures are the best choice because they operate with a much lower dead time than the high performance CZT which is optimized for best possible energy resolution and dynamic range.

5 ACKNOWLEDGMENTS

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6 REFERENCES

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