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Self-Correction of Lanthanum-Cerium Halide Gamma Spectra

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Abstract

Lanthanum-cerium halide detectors generally exhibit superior energy resolutions for gamma radiation detection compared with conventional sodium iodide detectors. However, they are also subject to self-activities due to lanthanum-138 decay and contamination due to beta decay in the low-energy region and alpha decay in the high-energy region. The detector's self-activity and crystal contamination jointly contribute a significant amount of uncertainties to the gamma spectral measurement and affect the precision of the nuclide identification process. This paper demonstrates a self-correction procedure for self-activity and contamination reduction from spectra collected by lanthanum-cerium halide detectors. It can be implemented as an automatic self-correction module for the future gamma radiation detector made of lanthanum-cerium halide crystals.

Keywords: Lanthanum bromide, cerium bromide, gamma spectrum, self-activity, computational removal, self-correction.

Introduction

Although superior energy resolution has been observed for gamma detectors made of lanthanum-cerium halides ($LaBr_3$ and $CeBr_3$) in many laboratory studies, the major barrier that has hindered the rapid commercial adaptations of lanthanum halides for gamma detection is the self-activity of these crystals, primarily due to the presence of isotope ^{138}La as well as other forms of contamination[1–3]. Lanthanum naturally contains about 0.09% of the radioactive isotope ^{138}La that emits gamma rays in predominantly two energy regions centered at ~789 keV and ~1436 keV, respectively. Apparently, to be able to use either $LaBr_3$ or $CeBr_3$ for gamma detection, it is

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necessary to have the self-activity and contaminations removed or reduced to an acceptable level. Complete physical removal of trace amounts of isotope ^{138}La from $LaBr_3$ through the material engineering process, if not impossible, can be extremely costly. All these reasons contribute to the delays of the next generation of high-energy resolution gamma spectrometers using lanthanum-cerium halides.

Most common techniques that remove the self-activity from the spectrum will also eliminate the background to within statistical precision and limitations. The method we propose here is a general method that will have the effect of removing the self-activity in the spectrum of the crystal, yet statistically preserving the background counts. This has important consequence in making certain high precision, high sensitivity, or gamma measurements involving large amounts of shielding. In our recent research[4,5], we developed two computational procedures for estimating the self-activity spectrum of a lanthanum-cerium halide crystal and for removing the self-activity spectrum from real-world gamma spectrum. By implementing these two computational procedures as internal data processing modules, a gamma spectrometer is able to automatically remove or reduce the self-activity from the real-time spectral measurements. The output spectrum is then compatible with the spectrum measured by a high-resolution spectrometer without self-activity. This paper presents the conceptual design and related engineering details for a gamma spectrometer made of lanthanum-cerium halide capable of automatically removing the self-activity from the detector crystals.

Key Technology Elements

The keys to this technology are the following two computational modules, with algorithm and procedures[5]:

(1) Computational module for estimating the self-activity of lanthanium-cerium halide crystals using a third reference detector, such as a NaI detector. The execution of this module can be performed during the manufacturig process of the detectors, and could be repeated as needed while the detectors are in the customer’s possession.

(2) Computational module for real-time self-activity removal. This module can be executed in automated mode during spectral acquisition, using the information related to the self-activity stored in the memory in the detector, and the information of the acquisition time. The output spectrum of the module would mimic the spectrum obtained by high spectral resolution detectors without self-activities.

All mathematical formulas and related notations used in this paper were defined in Yuan's work[5].

Detector Spectral Model

This technology adapts a simple gamma-spectral model[5] as follows:

$$S_R(e) = \varepsilon(e) \times [S_S(e) + S_B(e)] + S_A(e)$$

Where e is the energy level of the gamma spectrum, $S_R(e)$ is the detector-registered counts, or, more specifically, count rates per unit time. $S_S(e)$ is the true source count rate at the front of the detector crystal, $S_B(e)$ is the background count rate at the crystal, $S_A(e)$ is the count rate due to the internal self-activity, and $\varepsilon(e)$ is the detector-counting efficiency. The counting efficiency $\varepsilon(e)$ satisfies $0 \leq \varepsilon(e) \leq 1$. Although $\varepsilon(e)$ is usually energy dependent, for engineering purposes here we will assume that $\varepsilon(e) = \text{constant}$. However, the technology itself can be easily modified to accommodate situations where the counting efficiency $\varepsilon(e)$ is energy dependent.

Under the assumption that the detector-counting efficiency is independent of the energy levels, then the above model can be further simplified. For instance, for individual detectors made of $LaBr_3$ and $CeBr_3$ crystals, the detector spectral models are:

$$S_{LaBr3,R}(e) = \varepsilon_{LaBr3} \times [S_S(e) + S_B(e)] + S_{LaBr3,A}(e)$$

$$S_{CeBr3,R}(e) = \varepsilon_{CeBr3} \times [S_S(e) + S_B(e)] + S_{CeBr3,A}(e)$$

Conventional Scintillation Detector Data Processing Flow

Conventional scintillation detectors usually follow a conceptual design as shown in Figure 1. Since there is no internal data calibration process, the output spectra of the scintillation detector made of lanthanum-cerium halides ($LaBr_3$ and $CeBr_3$) are usually affected by the presence of the self-activity spectrum $S_A(e)$, as discussed in references[1,3].

Scintillation Detectors with Self-activity Removal Data Processing Flow

Based on the algorithm and procedures developed in our research and reported in the references 4 and 5, it is possible to design a scintillation detector with automated self-activity removal or reduction. Figure 2 shows the conceptual design of such detectors. The key component of the new design is an additional spectral processing unit (or self-activity isolation and reduction system) between the spectral digitizer and the output and display devices. The self-activity isolation and reduction system consists of two computational modules: the self-activity isolation module and the self-activity reduction module. The objective of the self-activity isolation module is to estimate the self-activity spectrum using a reference background spectrum measured by a relatively clean detector, such as a HPGe detector, or, in the worst case, a NaI detector. The estimated self-activity spectrum is then stored in the internal memory of the detector for future self-activity reduction purposes. The self-activity isolation module needs to be run at least once for each detector during the manufacturing process. This initial run will generate a “default” self-activity spectrum for future use. The self-activity reduction module can be automatically invoked during the operation of the detector, using the stored self-activity spectrum (in counts per channel per unit time).

Self-activity Isolation Module

The self-activity isolation module is used for estimating the self-activity spectrum using a reference spectrum measured by a relatively clean detector, such as a HPGe or a NaI detector. Figure 3 shows a procedure using a NaI detector as a calibration detector, where CPM stands for counts per minute. In industrial production mode, a better resolution HPGe detector is suggested. Figure 3 outlines the procedures for computing the self-activity spectrum using two spectra measured by both $\mathrm{LaBr_3}$ and NaI. The initial estimate of the self-activity spectrum must be performed by the manufacturer and stored in the internal memory for future use. If the crystal is free of contamination of other radioactive isotopes, the self-activity spectrum due to ^{138}La estimated by the manufacturer should be quite consistent, since the isotope ^{138}La has a long half-life of $t_{1/2} = 1.05 \times 10^{11}$ years and a small decay constant:

$$\lambda = \frac{\ln 2}{t_{1/2}} = \frac{\ln 2}{1.05 \times 10^{11}} = \frac{\ln 2}{1.05} \times 10^{-11} \approx 6.6 \times 10^{-12}$$

For each year, the relative reduction in its activity amounts to:

$$1 - \frac{Activity(\text{year } 2)}{Activity(\text{year } 1)} = 1 - \frac{\lambda N(\text{year } 2)}{\lambda N(\text{year } 1)} = 1 - e^{-\lambda} = 6.60139 \times 10^{-12}$$

This is a very small number. Practically, over the operational life of the detector, it could even be assumed that there is virtually no change in the self-activity level associated with ^{138}La. However, the actinide (^{227}Ac, $t_{1/2} = 21.77$ years) contamination in the crystal may contribute a small source of self-activity that may change over the lifetime of the crystal.

For crystals free of actinide and similar short-lived nuclide contaminations, once the self-activity spectrum is calculated during the manufacturing process and stored in the internal memory as spectral correction parameters, the end users rarely need to perform the self-activity separation again, although the self-activity spectrum may be re-calculated at the user's end. In a situation where trace quantity, short-lived nuclide contamination (such as ^{227}Ac) exists, periodically repeated calculations of the spectral correction parameters may be desired.

The clean energy range $E_{\text{Clean,LaBr3}}$ is used for computing the average relative efficiency of the detector. The exact selection of the clean energy range $E_{\text{Clean,LaBr3}}$ depends on the crystal type and contents of the impurities, as well as the energy range and energy resolution of the detector.

In the worst case, the whole spectral range may be used as E $_{\text{Clean,LaBr3}}$. In our study, we used E $_{\text{Clean,LaBr3}}$: 1500~2000 keV. This energy range can be further refined, and can even be automatically refined for individual crystals during the detector manufacturing process. Similar discussion is also true for separating self-activity from $CeBr_3$ spectrum. Likewise, the clean range E $_{\text{Clean,LaBr3}}$ for $CeBr_3$ can be different and may fluctuate from crystal to crystal. In our research, we used E $_{\text{Clean,CeBr3}}$: 300~1200 keV.

Self-activity Reduction Module

The self-activity reduction module performs real-time self-activity reduction. Taking the $LaBr_3$ detector as example, suppose that the initial self-activity spectrum $S_{LaBr3,A}(e)$ in counts per minute (CPM) per channel has been estimated by the manufacturer (or the user) and stored in the memory; the raw gamma spectrum $S_{LaBr3,R}(e)$ obtained over a counting interval τ (in minutes) can then be corrected by simple spectral subtraction:

$$\tilde{S}_{LaBr3,R}(e) = S_{LaBr3,R}(e) - \tau \times S_{LaBr3,A}(e)$$

Therefore, the self-activity reduction module can be automated. The output spectrum $\tilde{S}_{LaBr3,R}(e)$ has been shown in our research to mimic the output spectrum obtained by detectors without self-activity and or similar high-energy resolution. The simplified flowchart is shown in Figure 4.

Test Examples

The self-activity removal procedure described in the paper has been tested in our study[5]. Two sets of spectra of low-radiation background and high-radiation Eu-152 were collected using various detectors for the test. The spectra collected by $LaBr_3$ and $CeBr_3$ detectors were processed using the self-activity removal procedures described in the paper. Uncorrected count rate spectra are shown in Figures 5a and 6a; the corrected spectra are shown in Figures 5b and 6b, respectively.

The self-correction procedure described in the paper is capable of removing the self-activity, as shown in Figure 5a and 5b, and is also capable of preserving the characteristic features of high radiation sources, as seen in Figure 6a and 6b.

Summary

This paper presents a technology for a gamma spectrometer capable of performing automated self-activity correction. This technology would enable the designing and manufacturing of gamma spectrometers using crystals, such as $CeBr_3$ and $LaBr_3$, with potential for energy resolutions superior to NaI, but technically hindered by their strong internal radioactivity. This technology substantially reduces the need for high purity crystals (and associated cost) for making gamma spectrometers of similar spectral characteristics. The relevant computational procedures and algorithms for estimating and correcting detector crystal self-activity were described in Guss' work[4]. This paper presents the method of incorporating those computational procedures into an applicable detector technology.

Acknowledgement

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List of Figures

Figure 1. Conventional scintillation detector data processing flow.

Figure 2. Scintillation detectors with automated self-activity reduction data flow.

Figure 3. The $LaBr_3$ self-activity isolation module of the self-correction system with preferred clean energy calibration range of $E_{Clean,LaBr3}$: 1500~2000 keV.

Figure 4. The $LaBr_3$ self-activity removal module of the computational self-correction system. It is similar for $CeBr_3$ detectors.

Figure 5. Gamma count rate spectra (a) collected by different detectors in a low radiation environment, and (b) spectra with the self-activity removed. The self-activity has been significantly reduced in the corrected spectra.

Figure 6. Eu-152 gamma count rate spectra (a) collected by different detectors, and (b) the spectra with the self-activity removed. The self-activity removal process preserves the Eu-152 characteristics in the corrected spectra.

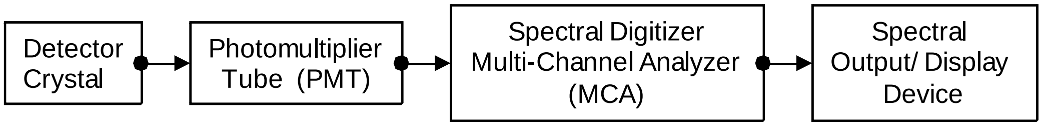

Detector Crystal
Photomultiplier Tube (PMT)
Spectral Digitizer Multi-Channel Analyzer (MCA)
Spectral Output/ Display Device

Figure 1

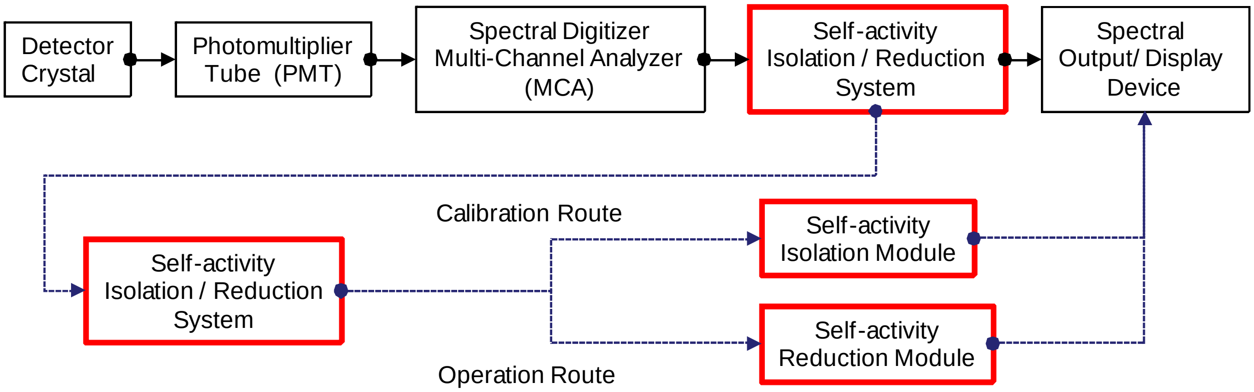

Detector Crystal
Photomultiplier Tube (PMT)
Spectral Digitizer Multi-Channel Analyzer (MCA)
Self-activity Isolation / Reduction System
Spectral Output/ Display Device
Calibration Route
Self-activity Isolation / Reduction System
Self-activity Isolation Module
Self-activity Reduction Module
Operation Route

Figure 2

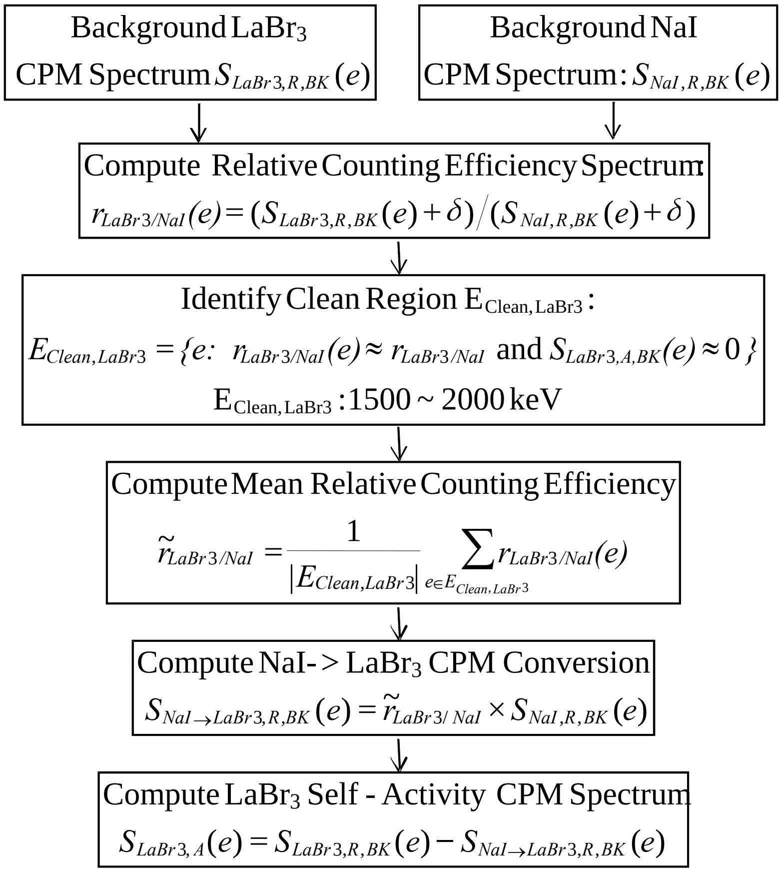

Background LaBr$_3$ CPM Spectrum $S_{LaBr3,R,BK}(e)$
Background NaI CPM Spectrum: $S_{NaI,R,BK}(e)$
Compute Relative Counting Efficiency Spectrum
$r_{LaBr3/NaI}(e) = (S_{LaBr3,R,BK}(e)+\delta)/(S_{NaI,R,BK}(e)+\delta)$
Identify Clean Region $\mathrm{E}_{\mathrm{Clean,LaBr3}}$:
$E_{Clean,LaBr3} = \{e: r_{LaBr3/NaI}(e) \approx r_{LaBr3/NaI}$ and $S_{LaBr3,A,BK}(e) \approx 0\}$
$\mathrm{E}_{\mathrm{Clean,LaBr3}}$: 1500 ~ 2000 keV
Compute Mean Relative Counting Efficiency
$\tilde{r}_{LaBr3/NaI} = \frac{1}{|E_{Clean,LaBr3}|} \sum_{e \in E_{Clean,LaBr3}} r_{LaBr3/NaI}(e)$
Compute NaI-> LaBr$_3$ CPM Conversion
$S_{NaI \to LaBr3,R,BK}(e) = \tilde{r}_{LaBr3/NaI} \times S_{NaI,R,BK}(e)$
Compute LaBr$_3$ Self - Activity CPM Spectrum
$S_{LaBr3,A}(e) = S_{LaBr3,R,BK}(e) - S_{NaI \to LaBr3,R,BK}(e)$

Figure 3

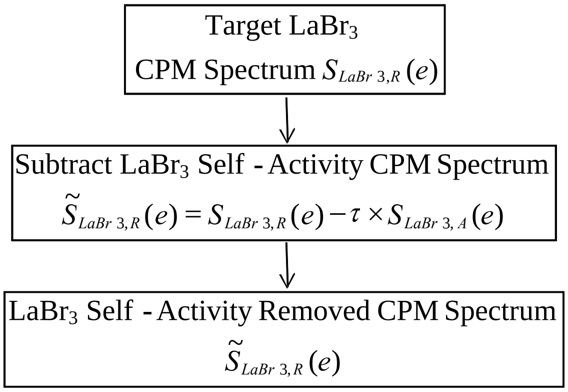
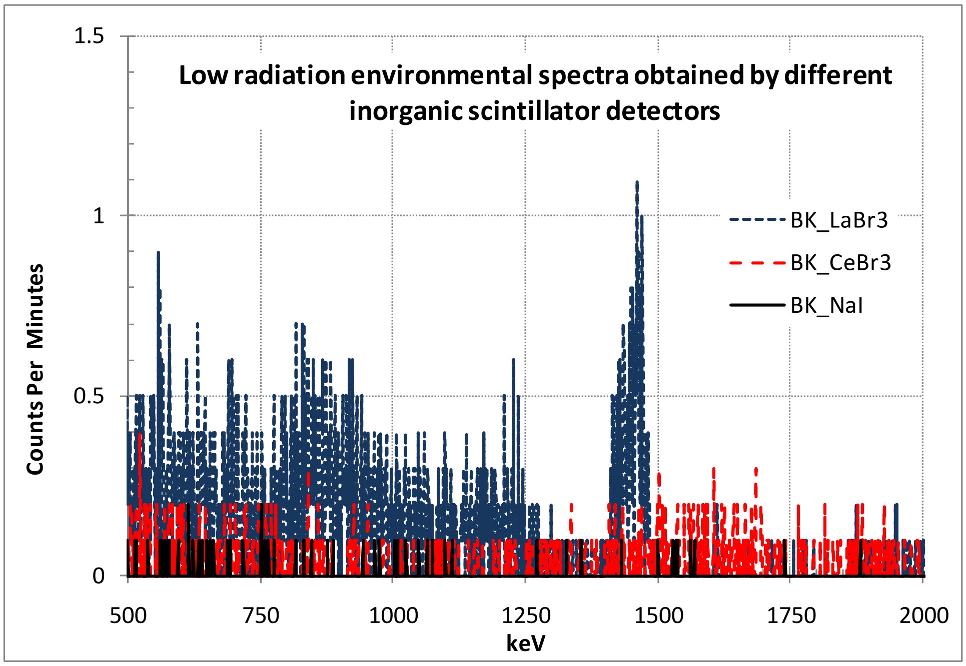
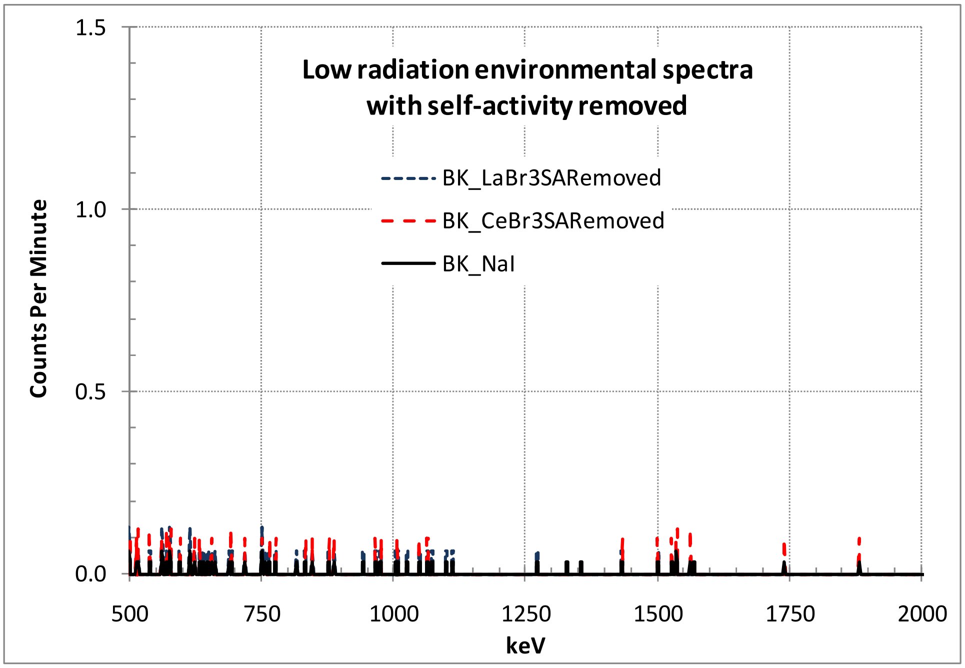

Target LaBr$_3$
CPM Spectrum $S_{LaBr\,3,R}(e)$
Subtract LaBr$_3$ Self - Activity CPM Spectrum
$\tilde{S}_{LaBr\,3,R}(e) = S_{LaBr\,3,R}(e) - \tau \times S_{LaBr\,3,A}(e)$
LaBr$_3$ Self - Activity Removed CPM Spectrum
$\tilde{S}_{LaBr\,3,R}(e)$

Figure 4

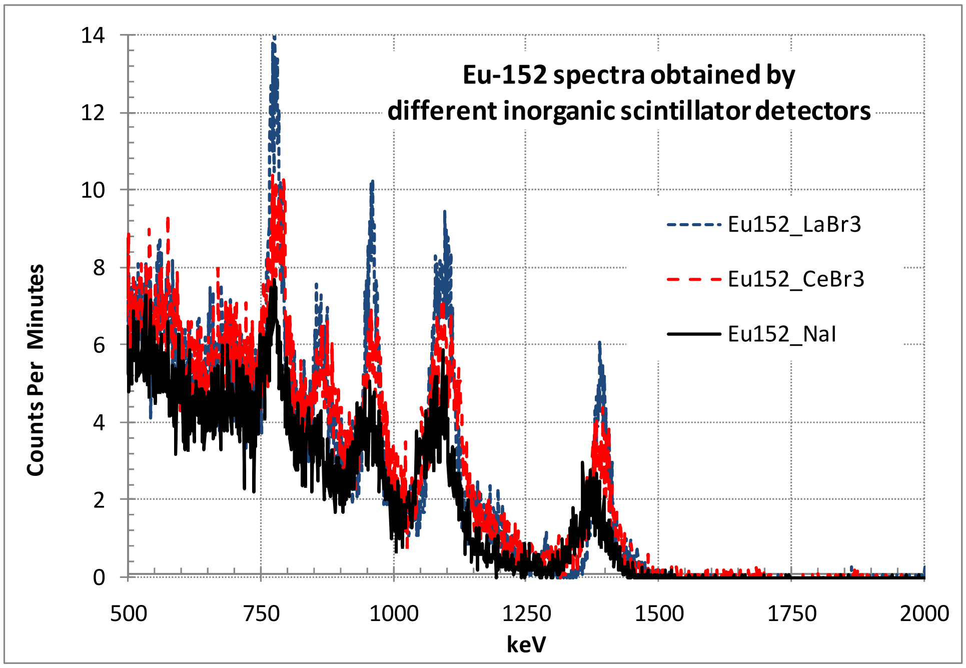


a. Raw count rate spectra of a low radiation environment collected by NaI, $LaBr_3$ and $CeBr_3$ detectors

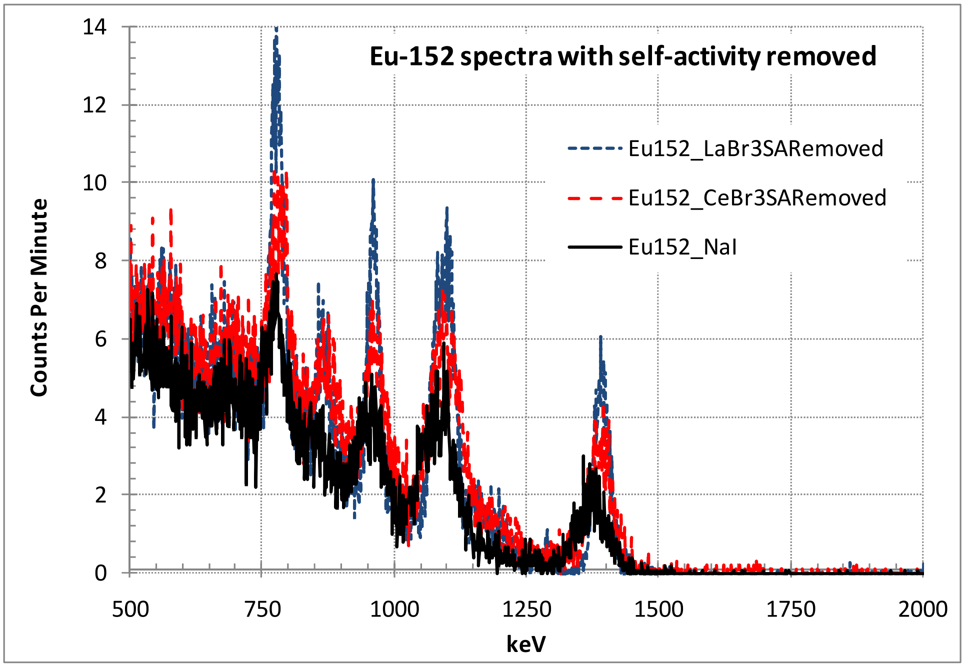


b. Environmental count rate spectra with self-activity removed

Figure 5



a. Eu-152 count rate spectra collected by NaI, LaBr$_3$ and CeBr$_3$ detectors



b. Eu-152 count rate spectra with self-activity removed

Figure 6