

Detection of escape peaks in environmental gamma spectrometry

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Abstract

Pair production (PP) events are observed with gamma spectrometry, when investigated samples emit sufficient numbers of photons with energies greater than 1.022 MeV. Because this condition is not typical of environmental samples, detection of PP is uncommon for environmental radioactivity. Nonetheless, this work experimentally demonstrated the detection of PP for an environmental sample of relatively high amount of natural, radioactive isotope of potassium ⁴⁰K, utilizing low background gamma spectrometry with sufficiently long counting times. Such detection was spectrally seen through the escape events of the annihilation photons that typically follow PP.

Keywords: Double escape; gamma spectrometry; NORM; pair production; single escape.

1. Introduction

Natural sources are the main contributors to environmental radioactivity. Examples of natural radionuclides are ²³⁸U and its decay series, ²³²Th and its decay series, and the primordial radionuclide ⁴⁰K. In addition to being long-lived with half lives in the order of 10⁹ years, these natural occurring radioactive materials (NORM) reside in aquatic, terrestrial, and other environmental components in different quantities and with varying levels of radioactivity. Such ubiquitous and ever-lasting presence makes detection of natural radiation an expected outcome for environmental samples (Alrefae *et al.*, 2012; Hermanspahn, 2009).

Measurement of environmental radioactivity is typically performed with gamma spectrometry (IAEA 1989; Knoll, 2000). This reliable technique is based on detecting photons that are emitted by radionuclides residing in the investigated sample, where the emitted photon energy (E_γ) acts as a signature that uniquely identifies the emitting radionuclide. For example, the specific E_γ values of 46.5 keV, 93.4 keV, and 1460.8 keV are identifiers for ²³⁸U, ²³²Th, and ⁴⁰K respectively (IAEA, 1989; Knoll, 2000). These energy values are spectrally seen, when a sufficient number of emitted photons deposit their full energies inside the detector. As a result, a prominent peak rises in the gamma spectrum known as the full energy peak (Ph), thereby identifying the radionuclide from which the photons were emitted.

Hence, the magnitude of this peak is explicitly used to measure the activity of the emitting radionuclide in the investigated sample (IAEA, 1989; Knoll, 2000).

The performance of the gamma spectrometer is limited by the detection efficiency. The value of this efficiency is a function of the geometry of the measurement experiment, the constituents of the detector, and the constituents of the investigated sample. As any measurement device, a gamma spectrometer requires calibration for its efficiency. The most common method for performing efficiency calibration is the measurement of a standard source of known radioactivity with a constituency and a geometry that are similar to those of the investigated sample (IAEA, 1989). Such standard sources are commercially available to interested users. Alternatively, efficiency calibration can be performed using computational methods. This non-experimental approach is performed numerically using Monte Carlo methods and appropriate physics models to simulate the interaction of photons with matter (Alrefae, 2014). Thus, the detection efficiency is obtained for the given sample geometry and constituency at the desired photon energy.

For emitted photons of ($E_\gamma \geq 1.022$ MeV), a pair production (PP) process is possible, where an electron-positron pair is produced (Knoll, 2000). This process is followed by pair annihilation, where two photons are emitted each of energy 0.511 MeV. Possibly, both annihilation-produced photons may undergo

photoelectric absorptions, thus contributing to the rising Ph. Alternatively, one annihilation-produced photon may undergo a photoelectric absorption, while the other leaves the detector with no interaction. This process is known as a single escape (SE) event, which contributes to a spectral peak of energy ($E_\gamma - 0.511$ MeV). Yet, it is also possible for both annihilation-produced photons to leave the detector with no interaction, in a process known as a double escape (DE) event, thus contributing to a spectral peak of energy ($E_\gamma - 1.022$ MeV).

A search in the literature yields a number of studies that investigated escape peaks (Johnson & Mann, 1973; Chao, 1992; Sardari & Baghini, 2004). These studies involved producing man-made, high energy radionuclides thus enabling definite observance of SE and DE in relatively short counting times (~ 30 minutes). However, for environmental radioactivity, detection of escape peaks is rather uncommon owing to two main reasons. Firstly, most NORM radionuclides of interest emit photons with energies less than the threshold for pair-production (1.022 MeV). Secondly, for NORM radionuclides with $E_\gamma \geq 1.022$ MeV their amount of presence in investigated samples is typically small, thereby causing their already low SE and DE peaks to be completely concealed by the background levels. Nonetheless, it is hypothesized that these two obstacles can be experimentally overcome by selecting environmental samples of relatively large quantities of NORM, in addition to maintaining relatively long counting times during measurements. However, it is noteworthy, that the cross section for pair production is very low at energies not much higher than the threshold energy of 1.022 MeV, which is a typical case for environmental samples, and of ^{40}K in particular. Hence, the aim of this work was to observe escape peaks in environmental radioactivity gamma spectrometry.

2. Materials and methods

A low background, high purity germanium (HPGe) detection system (Canberra, CT, USA) was used for this study. This counting system, which was electro-mechanically cooled, carried a crystal with dimensions of 79.50 mm in diameter and 58.50 mm in length. For data acquisition, the system was connected to a complete arrangement of devices that included a preamplifier, a linear amplifier, a bias supply, an analog-to-digital converter (ADC), and a multi-channel analyzer (MCA). Data analysis was performed with the Gennie2K (version 3.2.1) software, which was installed on a personal computer that was connected to the counting system

as well. The shaping time constant for the detector was chosen to be 6 μs , to produce an energy resolution of 2.1 keV (FWHM) at an energy of 1.33 MeV. The detector was housed inside a 10 cm thick lead shield with 1 mm tin and 1.6 mm copper inner linings. Compared to a standard NaI(Tl) detector of size 3 in. diameter X 3 in. length, the HPGe system presented a relative efficiency of 80% for an energy of 1.33 MeV. Energy calibration for the counting system was performed using a set of known-activity sources (Spectrum Technologies, Oak Ridge, TN, USA) (Eckert and Ziegler, Valencia, CA, USA), that covered an energy range of 166 – 1836 keV. To prevent coincidence summing, these point sources were placed at a distance of 8 cm from the detector's face.

For measurement, samples containing high amounts of potassium (K) were sought. The rationale was based on the fact that high amounts of stable potassium necessarily meant relatively high amounts of the radioactive, natural, isotope of potassium, namely ^{40}K due to natural abundance. Hence, an environmental type of sample that fitted this criterion was selected, namely plant fertilizer that exhibited a ^{40}K activity concentration of 10748 Bq/kg. Prior to gamma spectrometry measurements, these samples underwent proper lab preparation (IAEA, 1989) before being placed in cylindrical containers of radii to lengths ratios of $\frac{3}{4}$ and 3. For sufficient counting time, each sample was counted for four full days (345600 seconds), where the targeted spectral peaks were located at 949 keV and 438 keV for SE and DE respectively. Under the same measurement conditions, empty containers of the same geometries were counted to obtain the background level.

For comparison purposes, an environmental sample with a lower level of ^{40}K was measured. After proper lab preparation, this sample which was an alum powder, underwent the same counting procedure as its counterparts of the plant fertilizer type with the relatively high ^{40}K content. The alum powder sample exhibited a ^{40}K activity concentration of 117 Bq/kg, thus about two orders of magnitude less than its plant fertilizer counterparts.

3. Results and discussion

The gamma spectra of the plant fertilizer in all measured samples revealed clear SE and DE. These detected escape peaks were large enough to rise above the background level of radiation and the Compton continuum, thus confirming the hypothesis of this work. The ratio of SE/DE, which is known to be independent of E (Johnson

& Mann, 1973; Chao, 1992; Sardari & Baghini, 2004), was experimentally measured for this work to be 2.22 ± 0.030 and 2.17 ± 0.043 for the cylindrical containers of radius to length ratios of $\frac{3}{4}$ and 3 respectively. Compared to theoretical values (Chao, 1992), the experimentally obtained SE/DE ratios were some 20% greater for the given detector's geometry and dimensions. However, it should be noted that the literature-published theoretical values (Chao, 1992) were computed for point sources rather than the volume source geometry, which was investigated in this work. Such variation may be partly responsible for the reported deviation. Furthermore, the closeness of the experimentally found SE/DE values in spite of the variation of sample geometry indicated the insensitivity of the SE/DE ratio to sample geometry.

Measurement of the alum powder sample did not reveal any escape peaks. This outcome was anticipated, since this sample contained a ^{40}K activity concentration of about two orders of magnitude less than its plant fertilizer counterparts.

To the best of our knowledge this work is the first to report detection of gamma spectral escape peaks for non-contaminated environmental samples. Such detection was possible by prolonging the measurement counting time, which enabled the escape peaks to overcome the height of the Compton continuum, thus confirming the hypothesis upon which this work was built. In other words, the long measurement times have greatly improved the counting statistics, thereby allowing for the spectral visibility of the escape peaks. Although only one type of samples namely fertilizer was investigated, it is believed that other environmental sample types would exhibit the same observations so long as the investigated samples fit the criterion of containing relatively high quantities of NORM gamma emitters with $E_{\gamma} > 1.022$ MeV.

It is believed that other experimental techniques may be employed to observe the same findings. For example, a very high efficiency spectrometer (relative efficiency $> 100\%$) can be used for better detection of the escape peaks. Alternatively, an ultra low background counting system could be used to significantly reduce background levels that would otherwise conceal escape peaks. Such

techniques are expected to reduce the counting time for the detection of the sought escape peaks. These alternative techniques are left for future work. Also left for future work are gamma-gamma coincidence measurements, which are expected to reveal interesting findings.

4. Conclusion

This work demonstrated the ability to detect pair production events, and hence escape peaks in environmental gamma spectrometry. This detection was achieved for environmental samples that emit high energy gamma rays such as the naturally occurring isotope of potassium ^{40}K . Thus, using a low background gamma spectrometer with high counting time will surely enhance such detection.

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ملخص

تمت مراقبة أحداث إنتاج الأزواج (PP) بواسطة مطياف جاما، عندما تبعث عينات الفحص أعداداً كافية من الفوتونات ذات طاقات أكبر من 1.022 مليون إلكترون فولت. ولأن هذه الحالة ليست نمطية مع العينات البيئية، فإن الكشف عن إنتاج الأزواج غير شائع بالنسبة للنشاط الإشعاعي البيئي. ومع ذلك، أظهر هذا العمل تجريبياً الكشف عن إنتاج الأزواج لعينة بيئية بها كمية عالية نسبياً من النظائر الطبيعية المشعة من البوتاسيوم ⁴⁰K، وذلك باستخدام خلفية منخفضة لمطياف جاما لها فترات عد طويلة بشكل كافٍ. وتمت مشاهدة هذا الكشف طيفياً من خلال أحداث هروب الفوتونات المندثرة والتي عادةً ما تتبع إنتاج الأزواج.