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Measurements of thorium and uranium daughters in radioenvironmental samples using $\gamma\gamma$-coincidence spectrometry

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HIGHLIGHTS

- We discussed the performance of a $\gamma\gamma$-coincidence spectrometer for measuring thorium and uranium daughters in environmental samples. The coincidence efficiencies for γ-ray cascades from ^{208}Tl and ^{214}Bi decay are small (<1%), but at the same time the coincidence room background is lower by three orders of magnitude compared to the singles count rate. We described the most favorable coincidence gating schemes, detection efficiencies, background count rates, and minimum detectable activities.
- We employed the device to estimate thorium and uranium daughter activities in three different samples: Brazil nuts, potting mix, and magazine paper. Our uranium and thorium activities for Brazil nuts agree with some, but not all, previous measurements. Neither thorium nor uranium activities have previously been reported for commercial potting mix. Our measured values are in the range of activities reported for various soils in the United States. For magazine paper, our measured activities are lower than previously determined results.

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ABSTRACT

We present the performance of a $\gamma\gamma$-coincidence spectrometer for measuring the activities of thorium and uranium daughters in environmental samples. The spectrometer consists of two NaI(Tl) detectors facing each other inside a low-background passive shield. We present coincidence gating schemes for achieving the best signal-to-noise ratios, coincidence detection efficiencies, background levels, and minimum detectable activities. The spectrometer is simulated using Geant4 to correct sample efficiencies for self-absorption effects. The device is used to measure thorium and uranium daughter activities in Brazil nuts, potting mix, and magazine paper. Our results for Brazil nuts agree with some, but not all, previous measurements. Thorium or uranium daughter activities have previously not been reported for commercial potting mix. For magazine paper, our measured activities are lower than most previously determined values.

1. Introduction

Understanding the effect of naturally occuring radionuclides, such as the uranium and thorium series, on humans requires a detailed understanding of their behavior in the environment. These nuclides are continually being redistributed by various natural mechanisms and can thus serve as tracers of the various biological, meteorological, or geophysical processes. Studies of environmental radioactivity require the availability of sensitive techniques capable of measuring concentrations at very low levels in small samples. In addition, the investigation of complex mixtures of naturally occuring radionuclides requires highly selective measurement techniques. Because of their supreme energy resolution, many investigators have used for this purpose high-purity Germanium (HPGe) detectors with complex passive shields, sometimes located in deep underground laboratories. Spectrometers based on NaI (Tl) crystals represent an interesting and affordable[1] alternative, especially if they involve $\gamma\gamma$-coincidence spectrometry (Roedel, 1970; Cooper and Perkins, 1971; Povinec, 1981; Grismore et al., 1998; Zhang et al., 2011, 2014; Burnett and Davies, 2011; Britton et al., 2012). The advantages are significant, since (i) samples of small sizes can be

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[1] The total cost of our spectrometer was about $50,000, including the two detectors, passive shielding, electronics, computer, and the machining of parts. It is thus much less expensive than a HPGe-based setup, which in addition requires the use of liquid nitrogen.

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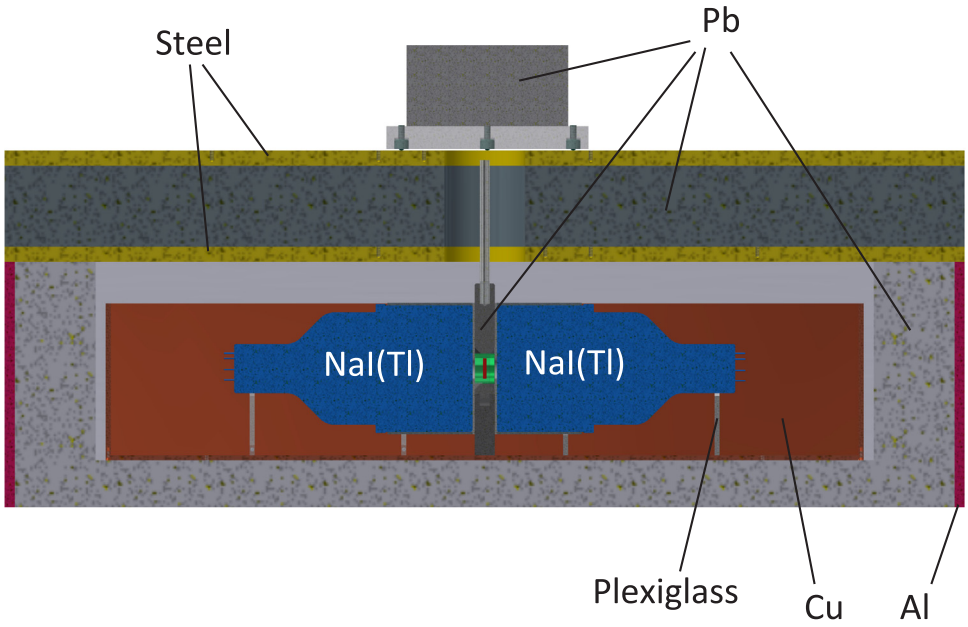

Fig. 1. Cross-sectional views of the $\gamma\gamma$-coincidence spectrometer, showing the geometry and different layers of materials. Part of the lead shield, located between the two detectors, was removable and contained the sample holder (shown in green). Each detector contained a crystal with a diameter of 152.4 mm and length of 101.6 mm. See also Fig. 2 in Ref. Tillett et al. (2017) (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.).

assayed, (ii) the identification of the radionuclides is virtually unambiguous, (iii) the counting times are reasonable, and (iv) minimum sample preparation is involved, thereby greatly reducing the risk of accidental contamination.

Recently, such an instrument consisting of two NaI(Tl) detectors has been commissioned at The University of North Carolina at Chapel Hill. The performance of the instrument for detecting positron-emitting radionuclides, including the measurement of the cosmogenic ^{26}Al activity in a small (18 g) meteorite fragment, has been described in Ref. Tillett et al. (2017). Here, we discuss the performance of the spectrometer for detecting thorium and uranium daughters in samples of Brazil nuts, potting mix, and magazine paper.

The $\gamma\gamma$-coincidence spectrometer is described in Section 2. Procedures and detection schemes are presented in Section 3. Measurements and results are discussed in Section 4. Conclusions are given in Section 5.

2. Equipment

The spectrometer is shown in Fig. 1. It consisted of two NaI(Tl) detectors, which faced each other at a distance of 38.1 mm along their symmetry axis. Each detector contained a crystal with a diameter of 152.4 mm and length of 101.6 mm, inside an aluminum housing with a wall thickness of 0.81 mm. The crystals were coupled to photomultiplier tubes of 127.0-mm diameter via low-background optical quartz windows. The only spectrometer part selected for low-background radiation was the optical window.

The detectors were surrounded by several layers of different metals. The innermost layer consisted of 0.64-cm thick copper, surrounded by 10.2-cm thick lead. The outermost layer was made of 1.0-cm thick aluminum and provided support for the shielding above the detectors. The top shield consisted of a low-background steel plate of 2.0-cm thickness, which supported a 10.2-cm-thick layer of lead bricks and another 2.0-cm-thick steel plate. A 10.2-cm-diameter hole at the center of the top shield provided convenient access to the sample (shown in green in Fig. 1) located between the two detectors.

A lead plate of 2.54-cm thickness was located between the detector front faces to reduce background coincidence events caused by γ-rays that Compton scatter between the detectors. Background radiation was reduced by factors of 2–4 by this shield, depending on the coincidence gate used. This lead plate had a central hole of 3.8-cm diameter, with tapered edges to reduce the absorption of γ-rays originating from the sample.

The output signal from each detector was split into two branches without pre-amplification. The first signal was processed by a spectroscopy amplifier and was subsequently fed into a 4096-channel VME amplitude-to-digital converter (ADC). The second signal was processed by a fast timing filter amplifier (TFA), and was then fed into a constant-fraction discriminator (CFD). The logic output signal provided the gate for the ADC and the start and stop signals for the time-to-amplitude converters (TAC). The output of the TAC was also fed into the ADC. The data were stored in list mode for subsequent offline analysis, where each event consists of the energy and timing information for both detectors. All timing and energy coincidence gating was performed using the data acquisition system JAM (Swartz et al., 2001).

Several cyclindrical sample holders were fabricated from aluminum. The outer diameter and width were 3.70 cm and 2.70 cm, respectively. The average wall thickness was ≈ 1 mm, resulting in an inner cavity volume of 24.05 cm^3. The sample inside the holder was hermetically sealed using a Viton O-ring.

3. Procedures

3.1. Reference materials and environmental samples

For the measurement of uranium and thorium daughters in environmental samples via γ-ray spectrometry, standards are produced from natural materials for which secular equilibrium can be assumed. For thorium, we used the CRM 107-A reference material, which is certified by the New Brunswick Laboratory.[2] This material was prepared by diluting monazite (rare-earth and thorium phosphate) sand with silica (99.9% SiO_2, essentially free of radioactivity). New Brunswick Laboratory used spectrophotometry to determine a resulting

[2] https://science.energy.gov/nbl.