

Improvement of the detection limit by radon background reduction method

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ABSTRACT

This paper describes a simple method to reduce the radon background component applied in environmental studies using gamma spectrometry. The radon component can be reduced by introducing either nitrogen gas or clean air into the detector chamber in order to create a positive pressure and further minimize

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radon intrusion from outside. This method shows that we can minimize the influence of radon daughter background such as ^{214}Pb , ^{214}Bi and ^{210}Pb (from ^{238}U), ^{212}Pb , ^{212}Bi and ^{208}Tl (from ^{232}Th). By the way, the detection limit of the gamma spectrometry is discussed.

INTRODUCTION

There are many efforts to the issues such as to look for the proper shielding materials for better detector housing and so on. Techniques developed so far for rare event research, dark matter search in particle and nuclear physics research are well known. In this research we would like to investigate the most suitable and sensitive technique for fixing the background origin in the materials.

To improve the detection limits in low level activity measurements, the background has to be reduced as much as possible. The background spectrum of a germanium detector is due to a combination of different components such as environmental gamma radiation, radioactivity in the construction material of the detector, radio impurities in the shield, cosmic rays and radon gas. The first three contributions can be reduced drastically by means of a suitable passive shielding made of old or very low – activity lead and by a careful selection of materials

surrounding the crystal. The cosmic ray component has been reduced by installing the germanium detector in an underground laboratory. However, building and operating an underground laboratory is expensive and inconvenient. Another possibility is to operate a gamma-ray spectrometer with an anticoincidence system i.e. a plastic scintillator surrounding the lead shield in anticoincidence with the germanium detector as active shielding [2].

Radon isotopes, both radon, ^{222}Rn , and thoron, ^{220}Rn , are present in air as active gases emanating from traces of ^{238}U and ^{232}Th in building constructional materials and/or local soils and rock. They can be absorbed on dust particles and surfaces within the detector enclosure and give rise to characteristic peaks of ^{214}Pb , ^{214}Bi and ^{210}Pb , (in the ^{238}U series) and ^{212}Pb , ^{212}Bi and ^{208}Tl (in the ^{232}Th series) in the background. The difficulty with radon is that its concentration around the detector is likely to vary

with time of day and season of the year, and with atmospheric pressure, wind speed, temperature, etc. A reliable and reproducible background from radon daughters is often difficult to achieve [1, 3-6].

In this work, we measure and evaluate reduced radon daughter into background spectra with and without vent gas from the liquid nitrogen Dewar into the shielding and calculate the limit of detection of gamma spectrometry using HPGe detector.

MATERIALS AND METHOD

Detector

The experimental set-up is a low-level gamma spectrometer including an HPGe detector with conventional amplifying and coding systems. This is equipped with an active shielding consisting in plastic scintillators working in anti-coincidence mode. The HPGe detector includes the germanium crystal cylinder with 66 mm outer diameter and 64 mm height. Inside the crystal, and there is a hole with 15 mm diameter, 51 mm depth and the relative efficiency of 51.6 %. Furthermore, there is an outer lithium layer, and an inner boron layer of the crystal. Acquisitions with detector are driven using InterWinner software that is also used for spectra display and processing.

The detector is included in a cylindrical measurement chamber ($\phi = 80$ mm, $H = 400$ mm) made of 4 mm selected copper with a parallel epiped shielding successively composed of a 50 mm-thick very low activity lead ($A < 10$ Bq.kg⁻¹), a 3 mm thick selected cadmium ($A < 50$ Bq.kg⁻¹) and a 100 mm thick low activity lead ($A < 50$ Bq.kg⁻¹). The material composition of the bottom is the same except the thickness of the low activity lead is 150 mm. The measurement chamber is filled with nitrogen gas exiting from the cooling Dewar to remove the radon from the chamber (Fig. 1). Finally, the whole system is installed in an underground

laboratory isolated with 1.50 m concrete walls and 1 m underground. This room is also equipped with specific ventilation and air conditioning system with double dust filtering, thus insuring air regeneration 7 times per day. The active shielding is performed using 5 plastic scintillators with the dimension of 750x750x70 mm³. They are out one at the top, four at the four edges and nothing at the bottom [1].

Standard solutions

The efficiency curve was obtained using the so-called "SG50" volume geometry 50cm³; and the container has the following characteristics: external diameter 40 mm, wall thickness 1.2 mm, bottom thickness 1.4 mm and the liquid is filled to the height of 4.56 mm. The initial efficiency curve was established in 2003, using nuclides such as ²¹⁰Pb, ²⁴¹Am, ¹⁰⁹Cd, ⁵⁷Co, ¹³⁹Ce, ⁵¹Cr, ¹¹³Sn, ⁸⁵Sr, ¹³⁷Cs, ⁶⁵Zn, ²²Na, ⁶⁰Co, ⁴⁰K and ⁸⁸Y. A general efficiency curve (efficiency versus energy) was obtained by fitting a log log polynomial to experimental values obtained by using the liquid standard sources.

The resulted validation efficiency is presented in Fig. 1. This indicates that the established efficiency curve is adequate to calculate the activity concentrations of the sediment marine sample.

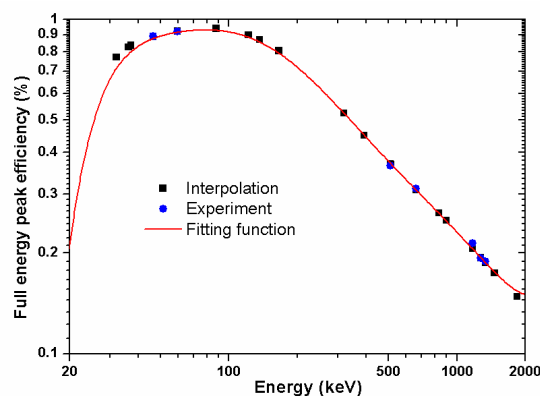


Fig. 1. Validation efficiency curve of SG50 geometry at 10 cm

RESULTS AND DISCUSSION

The effect of the radon suppression is shown in Table 1. Some reduction is observed in the gamma ray peaks of the ²²²Rn progenies, ²¹⁴Pb

and ²¹⁴Bi are by factors of 3.2–5.1 for ²¹⁴Pb and 5.6–6.3 for ²¹⁴Bi. Besides, ²²⁰Rn daughter, the count rate in ²¹²Pb is reduced 1.8 and 1.2 for ²⁰⁸Tl.

Table 1. Comparison count rates minute peak (c.p.m) for with and without nitrogen

Radionuclides	Energy (keV)	With nitrogen (W) (c.p.m)	Without nitrogen (Wo) (c.p.m)	Ratio Wo/W
²¹⁰ Pb	46.5	0.367 (7)	0.382(7)	1.0
²²⁶ Ra	186.2	0.166(4)	0.196(5)	1.2
²¹² Pb	238.6	0.125(4)	0.224(5)	1.8
²¹⁴ Pb	295.2	0.087(3)	0.283(6)	3.2
²¹⁴ Pb	351.9	0.078(3)	0.395(7)	5.1
²¹⁴ Bi	609.3	0.042(2)	0.264(6)	6.3
²¹⁴ Bi	1764.5	0.008(1)	0.045(2)	5.6
²⁰⁸ Tl	2614.5	0.009(1)	0.010(1)	1.2

$$0.367(7) = 0.367 \pm 0.007$$

Fig. 2A and 2B presented the measurement results with acquisition time 504000s with and without nitrogen, respectively.

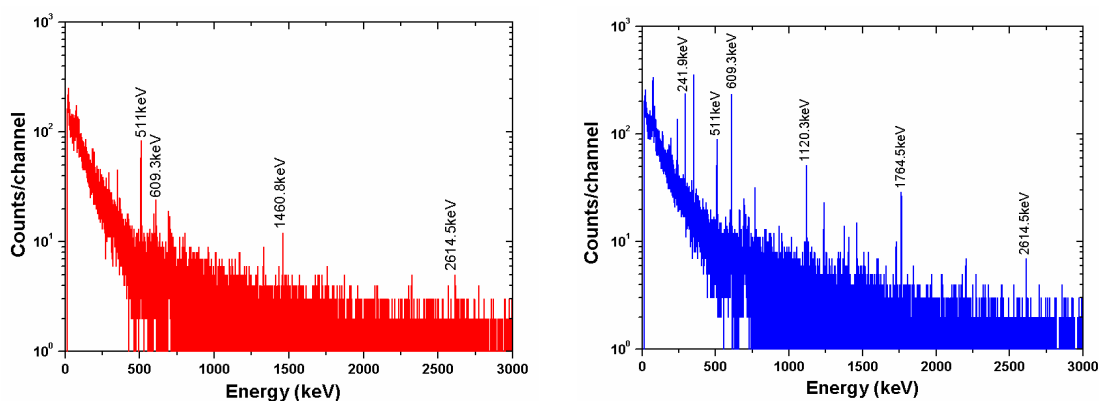


Fig. 2. A. The spectrum with nitrogen; B. The spectrum without nitrogen

However, several applications of environmental radioactivity require not only low background but also lower limit of detection (L_D) for different samples. It has been obtained according to the equation [2]:

$$L_D = 2.71 + 3.29\sqrt{B} \quad (1)$$

where B is the integral background in the region of interest (counts), n is the number of channels in the peak region of interest, m is

the number of background channels on each side of the peak.

The minimum detectable activity (MDA) was calculated:

$$MDA = \frac{L_D}{\epsilon \cdot I_\gamma \cdot t \cdot V} \quad (2)$$

where ϵ is the detection efficiency of the peak, I_γ is the gamma ray emission probability and t is the acquisition time(s) and

V is the sample volume or mass. In Eq. (2), the confidence level is 95 %.

On the other hand, the MDA is calculated for HPGe detector for a SG50 geometry as given in the formula 2. The MDA is clearly improved (see Fig. 3) using radon suppression system for the ^{222}Rn progenies and the ^{220}Rn daughter.

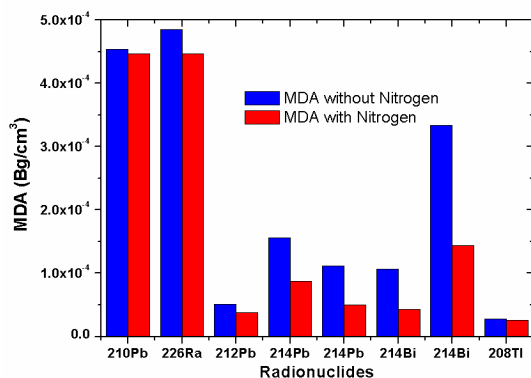


Fig. 3. Comparison of MDA values for SG50 geometry with and without nitrogen in shielding

Because, ‘environmental’ origin soils, waters and such like are measured either to determine background levels of radiation or to assess the

level of contamination as a consequence of human activity. The nuclides usually measured by gamma spectrometry are the cosmogenic nuclides: ^{40}K , ^{235}U , ^{238}U and ^{232}Th . In many cases, it will be necessary to make a peaked background correction in addition to the normal peak background continuum subtraction. All of those difficulties are then compounded by the fact that there are a large number of mutual spectral interferences between many nuclides in the decay series of uranium and thorium.

CONCLUSION

This study shows that with and without nitrogen gas, is necessary to reduce the radon background. This method can use the gamma spectrometry to calculate the radioactivity of the environmental sample.

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Cải tiến giới hạn phát hiện bằng phương pháp giảm phong radon

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TÓM TẮT

Bài báo trình bày một phương pháp đơn giản để giảm thành phần phong radon ứng dụng cho hệ phổ kế gamma trong nghiên cứu môi trường. Thành phần khí radon được giảm bằng cách sử dụng khí nitrogen từ bình làm lạnh hoặc khí sạch từ bên ngoài để tạo ra một áp suất dương bên

Từ khóa: Radon, phổ kế gamma, giới hạn phát hiện, phong

trong buồng đo và thêm vào đó là giảm lượng khí radon từ bên ngoài. Phương pháp này chỉ ra rằng có thể giảm thiểu ảnh hưởng phong từ con cháu radon như ^{214}Pb , ^{214}Bi và ^{210}Pb (từ ^{238}U), ^{212}Pb , ^{212}Bi và ^{208}Tl (từ ^{232}Th). Giới hạn phát hiện của phổ kế gamma được thảo luận.

REFERENCES

- [1]. L. Ferreux, G. Moutard, T.T. Branger, Measurement of natural radionuclides in phosphogypsum using an anti-cosmic gamma-ray spectrometer, *Appl. Radiat. Isot* 67, 957–960 (2009).
- [2]. G. Gilmore, Practical gamma-ray spectrometry 2nd edition, John Wiley & Son, Ltd (2008).
- [3]. M. Hult, Low-level gamma-ray spectrometry using Ge-detectors, *Metrologia*, 44, S87–S94 (2007).
- [4]. S. Hurtado, M. Garcia-Leon, R. Garcia-Tenorio, Optimized background reduction in low-level gamma-ray spectrometry at a surface laborator, *Appl. Radiat. Isot.*, 64, 1006–1012 (2006).
- [5]. T.T. Thanh, L. Ferreux, M.C. Lépy, C.V. Tao, Determination activity of radionuclides in marine sediment by gamma spectrometer with anti cosmic shielding, *J. Environ. Radioactiv.*, 101, 9, 780–783 (2010).
- [6]. T.T. Thanh, T.T.H. Loan, M.V. Nhon, C.V. Tao, Improvement passive shielding to background reduction: application to determinate radioactivity at low-energy gamma rays, *Kerntechnik*, 79, 3, 247–252 (2014).