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Environmental radionuclides as contaminants of HPGe gamma-ray spectrometers: Monte Carlo simulations for Modane underground laboratory

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ABSTRACT

The main limitation in the high-sensitive HPGe gamma-ray spectrometry has been the detector background, even for detectors placed deep underground. Environmental radionuclides such as ^{40}K and decay products in the ^{238}U and ^{232}Th chains have been identified as the most important radioactive contaminants of construction parts of HPGe gamma-ray spectrometers. Monte Carlo simulations have shown that the massive inner and outer lead shields have been the main contributors to the HPGe-detector background, followed by aluminum cryostat, copper cold finger, detector holder and the lead ring with FET. The Monte Carlo simulated cosmic-ray background gamma-ray spectrum has been by about three orders of magnitude lower than the experimental spectrum measured in the Modane underground laboratory (4800 m w.e.), underlying the importance of using radiopure materials for the construction of ultra-low-level HPGe gamma-ray spectrometers.

1. Introduction

Low-level radioactivity measuring techniques have been widely applied in nuclear and environmental physics, and specifically ultra-high sensitive HPGe gamma-ray spectrometry has been a challenge for new investigations which could not be carried out before either because of lack of the sensitivity or because available sample size was too limited. Typical examples included high resolution ^{137}Cs profiles in the water column of the south Pacific, Indian and Atlantic Oceans (Aoyama et al., 2011; Povinec et al., 2011; Sanchez-Cabeza et al., 2011; Levy et al., 2011), which could not be carried out without underground HPGe gamma-ray spectrometry. Similarly, nondestructive determinations of ^{134}Cs and ^{137}Cs in 1-L water samples collected offshore of Japan after the Fukushima accident could not be carried out without an underground laboratory (Aoyama et al., 2013; Povinec et al., 2013a, 2013b).

Except environmental physics investigations, ultra-low-level gamma-ray spectrometers have been an important pre-requisite for large scale underground physics experiments, searching e.g. for neutrinoless double beta-decay, neutrino mass, and for dark matter particles (Arnold et al., 2010; Tomasello et al., 2010; Bellini et al., 2012; Agostini et al., 2014; Armengaud et al., 2016; Angloher et al., 2017; Povinec, 2017).

Although analyses of long-lived radionuclides (e.g. ^{14}C, ^{129}I, ^{236}U, Pu isotopes) in the environment have been recently mostly carried out by accelerator mass spectrometry (AMS) (Kutschera, 2005, 2010; Jull et al., 2008; Fifield, 2008; Povinec et al., 2008, 2009; Lujaniene et al., 2017) or by inductively coupled plasma mass spectrometry (ICPMS) (Povinec, 2003; Roos, 2008; Nisi et al., 2009), medium and short-lived radionuclides emitting gamma-rays (e.g. ^{134}Cs, ^{137}Cs) have been mainly analyzed by HPGe gamma-ray spectrometry (e.g. Povinec, 2003, 2004, 2005, 2008, 2010, 2012; Komura and Hamajima, 2004; Levy et al., 2011; Povinec et al., 2013a, b, 2018).

Large volume HPGe detectors represent a great break-through in the low-level gamma-ray spectrometry mainly because of high efficiency (up to about 200% relative to 7.62 cm in diameter and 7.62 cm long NaI (Tl) crystal), a high resolution even for large volume HPGe detectors (below 2 keV for 1.33 MeV gamma-rays), and a low background (as HPGe crystals are free of radioactive contamination) (Povinec et al., 2008, 2012).

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Recently important developments included operations of HPGe gamma-ray spectrometers underground, either in shallow laboratories (e.g. Heusser, 1995; Neumaier et al., 2000; Komura and Hamajima, 2004; Povinec et al., 2004, 2005, 2008, 2012, 2018; Heusser et al., 2015), or in deep underground laboratories (e.g. Neder et al., 2000; Laubenstein et al., 2004; Heusser et al., 2006; Hult et al., 2006; Niese, 2008; Simgen et al., 2014; Loaiza et al., 2011, 2015; Brudanin et al., 2017a, b; Laubenstein, 2017; Povinec, 2018).

A typical background of HPGe detectors is consisting of contributions from cosmic rays, from construction parts of the detector and its shields contaminated with radionuclides, as well as from natural radionuclides present in surroundings, e.g. in the laboratory walls. The secondary cosmic rays at sea level are consisting mainly of neutrinos (having no detectable effects in HPGe detectors), a soft component (photons, electrons, and positrons), of a nucleonic component (protons and neutrons) and of a hard component (muons). For elimination of cosmic-ray background induced by soft and nucleonic components it is possible to use a shielding from high Z-material (lead, steel, copper) for absorption of gamma-rays. Low Z-materials (e.g. polyethylene, paraffin) have been used for slowing down of neutrons, and their capture in added boron or lithium.

In surface and shallow underground laboratories, a typical background of HPGe detectors is consisting mainly of the contribution from cosmic-ray muons, therefore an active anticosmic shielding is required to discriminate the muon component (Zvara et al., 1994; Heusser, 1995; Povinec et al., 2004, 2005, 2018). In deep (several thousand meters of water equivalent, w.e.) underground laboratories, where the hard component of cosmic rays has been eliminated by surrounding rocks, the radioactive contamination of construction parts of the detector (cryostat) and its shield as well as of the laboratory walls represents the main background contribution (Laubenstein et al., 2004; Niese, 2008; Heusser et al., 2015; Brudanin et al., 2017a, b; Laubenstein, 2017; Povinec, 2018). The radioactive contamination is represented mainly by the ^{220}Rn and ^{222}Rn decay products (gamma-emitters) in the ^{232}Th and ^{238}U decay series, respectively, as well as by ^{40}K (Povinec et al., 2008, 2012). Selecting radiopure materials therefore helps to partially eliminate their contributions to the HPGe-detector background.

In very deep underground laboratories the muon flux is effectively decreased by several orders of magnitude to negligible levels, e.g.

(i) in the Gran Sasso National Laboratory, GSNL, Italy (depth 3800 m w.e.) the muon flux is $\Phi_\mu = (3.41 \pm 0.01)\ 10^{-4}\ m^{-2}\ s^{-1}$ (Bellini et al., 2012);
(ii) in the Laboratoire Souterrain de Modane, LSM, France (depth 4800 m w.e.) the muon flux is $\Phi_\mu = (6.25 \pm 0.01)\ 10^{-5}\ m^{-2}\ s^{-1}$ (Schmidt et al., 2013);
(iii) in the Sudbury Neutrino Observatory, SNOLAB, Canada (depth 6010 m w.e.) the muon flux is $\Phi_\mu = 3.12\ 10^{-5}\ m^{-2}\ s^{-1}$ (SNOLAB User's Handbook, 2013).

Except the above mentioned radioactive contamination by primordial radionuclides (^{238}U, ^{232}Th, ^{40}K) and their decay products, anthropogenic radionuclides either from global fallout, from the Chernobyl and Fukushima accidents (e.g. ^{137}Cs), or from radioactive tracers used during steel production (e.g. ^{60}Co) could also contribute to the detector background.

A specific contribution to the background of HPGe gamma-ray spectrometers operating underground is represented by cosmogenic radionuclides produced by interactions of cosmic-ray particles in the Ge crystal and in surrounding materials, e.g. in the detector holder, in cold finger, and in the cryostat. Therefore, precautions should be in place in the case of special detectors designed for ultra-low-level radioactivity measurements which should be transported in shielded containers and stored in underground places before their installation in deep underground laboratories (Povinec, 2003; Laubenstein, 2017).

For better understanding of background sources of HPGe gamma-ray spectrometers and their possible elimination, Monte Carlo simulations are required for background estimations before the spectrometers have been constructed and placed into operation. The aim of the present work has been to develop a Monte Carlo code that would allow the evaluation of background components of ultra-low background HPGe gamma-ray spectrometers operating deep underground, which originate both from cosmic rays and radioactive contamination of construction materials. This represents the main benefit of Monte-Carlo simulations as radioactive decay events from different radionuclide contaminants can be investigated for different parts of the HPGe gamma-ray spectrometer separately. The Monte Carlo simulations were carried out for the large volume HPGe gamma-ray spectrometer operating in the LSM Modane (France) at the depth of 4800 m w.e. Such multicomponent investigations of the HPGe-detector background have not been published before.

2. Materials and methods

2.1. HPGe detector and its shielding

The HPGe detector under investigation (Obelix) is a coaxial p-type detector of external diameter of 93.5 mm and length of 89.6 mm, with useful surface of 68 cm^2 (Loiaza et al., 2015; Brudanin et al., 2017a; b). The dead layer of the crystal is $<$ 0.5 mm/Ge. Its useful volume is 600 cm^3, the germanium mass is 3.2 kg and the relative efficiency is 162%. The resolution at 122 keV and 1332 keV is 1.13 and 1.98 keV, respectively. The HPGe crystal is mounted in a low background U-type aluminum cryostat. The FET of the preamplifier is placed inside the cryostat and it is shielded by a ring made of Roman lead. The end cap of the cryostat is 115 mm in diameter and 196 mm long, made of Al-Si alloy of 1.6 mm thick. Its distance from the cap is 4 mm. The HPGe crystal is mounted in a holder made of the same Al-Si alloy. The cold finger is made of oxygen-free high conductivity copper (OFHC). The inner lead shield is made of Roman lead with maximum thickness of 12 cm. The outer lead shielding is made from 20 cm of low-activity lead. Experimental set-up of the Obelix gamma-ray spectrometer is shown in Fig. 1 (Brudanin et al., 2017b).

2.2. Radioactive contamination of construction materials

For simulation of background gamma-ray spectra and their comparison with experimental spectrum it is necessary to know radionuclide contamination levels in all construction materials. The low-level Obelix gamma-ray spectrometer was made from materials with different levels of radioactive contamination, however, only limited information has been available on measured activities. The HPGe crystal is of high purity as this is required for its performance as a radiation detector. The FET placed below the crystal holder is contaminated with ^{226}Ra (1.6 ± 0.2 mBq kg^{-1}) and ^{228}Ra (2.9 ± 0.6 mBq kg^{-1}). As this radioactive contamination is not negligible, a ring of 10 mm thickness made of Roman lead is placed on the bottom of the cryostat to screen radiation coming from the bottom (Figs. 1 and 2). The Al-Si alloy used for the end cap is contaminated with ^{226}Ra (0.3 ± 0.2 mBq kg^{-1}), ^{228}Ra (< 0.1 mBq kg^{-1}), ^{228}Th (1.4 ± 0.2 mBq kg^{-1}) and ^{40}K (1.1 ± 0.2 mBq kg^{-1}). The Roman lead, which is used for the inner shielding, has ^{226}Ra and ^{228}Ra contents < 0.3 mBq kg^{-1}, and the ^{210}Pb content is < 60 mBq kg^{-1}. The outer lead has the ^{210}Pb content of 15 Bq kg^{-1} (Loaiza et al., 2015; Brudanin et al., 2017b).

Radon emanating from the construction parts of the gamma-ray spectrometer, of which decay products are important contributors to the detector background, is pushed out from the sample chamber (and of the lead shielding) with radon-depleted air (^{222}Rn concentration is ~15 mBq m^{-3}) from a radon trapping facility installed in LSM (Brudanin et al., 2017a).