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Production Rates of Cosmogenic Radionuclides in HPGe Spectrometers Operating Underground*

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Abstract: GEANT4 software package was used for calculation of production rates of cosmogenic radionuclides in HPGe detector operating in Modane underground laboratory. Production rates of several cosmogenic radionuclides produced by interactions of neutrons with Ge crystal (^{3}H, ^{54}Mn, ^{57}Co, ^{65}Zn, ^{68}Ge), with copper (^{46}Sc, ^{54}Mn, ^{56}Co, ^{57}Co, ^{58}Co, ^{59}Fe) and with Al+Si alloy (^{22}Na, ^{26}Al) were calculated. It has been found that cosmogenic radionuclides may contribute significantly to HPGe spectrometers background during first years of their operation underground.

Keywords: Monte-Carlo simulation, GEANT4, HPGe detector, underground laboratory, cosmogenic radionuclides, background.

1. Introduction

Experiments designed to observe rare nuclear processes require detectors with high efficiency, good energy resolution and low background [1–5]. These conditions are met by Ge detectors with large effective volumes as their detection limits mainly depend on the detector efficiency, resolution and background. The typical extremely rare nuclear events included neutrinoless double beta-decay and searchers for dark matter (DM). The neutrinoless double beta-decay energy scale is given by energy of beta-particles (<3 MeV), with expected half-life of this process $>10^{25}$ yr. On the other hand, the observation of DM candidate particles is focused on elastic scattering (nuclear recoil) of DM particles with nuclei in a detector. The energy scale of interest is lower than 100 keV, and less than 1 event per day is expected [4]. The gamma-ray background of HPGe detectors is induced by cosmic rays, by contamination of construction materials (detectors, shields, laboratory walls) with primordial (^{40}K, ^{232}Th, ^{238}U) and anthropogenic (^{60}Co, ^{137}Cs) radionuclides and by radon in the air and its decay products [5]. Cosmic rays at sea level consist of soft component (photons, electrons and positrons), nucleonic component (protons and neutrons) and hard component (muons). The elimination of the background induced by soft and nucleonic components can be arranged by shields made from high Z material (lead, steel, copper). Low Z materials (polyethylene, paraffin) with boron or lithium are used for slowing down and absorption of neutrons. Reduction of muon-induced background is, however, only possible by placing the detectors deep underground, or partially, by applying muon-veto detectors [6–9]. As in deep underground laboratories the muon flux is very low (by about five-six orders of magnitude lower than at the surface) [2, 4, 10], the contribution of radionuclide contamination from construction materials is the dominant background component, higher by about two to three orders of magnitude than the background induced by

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*Dedicated to Professor Peter Prešnajder on the occasion of his 70th birthday cosmic rays [6, 10]. Monte-Carlo simulations of HPGe detectors background were carried out in the past with the aim to determine the background components and to find ways how to decrease contributions from cosmic rays and the radio-contamination of materials [6–10]. The developed Monte Carlo models did not include, however, contributions to the detectors background from cosmogenic radionuclides produced by interactions of cosmic-ray particles with HPGe detector components and its shield. The construction components of HPGe detectors are irradiated by cosmic rays during production and storage of construction materials, during the detector production and its transportation to an underground laboratory. The production of cosmogenic radionuclides is stopped once the detector is installed in a deep underground laboratory. The short-lived cosmogenic radionuclides contribute to the detector background mainly during the first year of its operation in an underground laboratory [10–12]. The aim of this paper has been to quantify production rates of cosmogenic radionuclides and their contributions to the HPGe detector background using Monte Carlo simulations.

2. Monte-Carlo model

All simulations were carried out by GEANT4 toolkit developed at CERN [13]. For radioactive decay of nuclei with emission of , $^+$ or $^-$ particles, the Radioactive Decay Module (G4RadioactiveDecay) was used. The nuclear de-excitation of the excited daughter product was simulated using G4PhotoEvaporation class. The module is empirical, and it uses data from Evaluated Nuclear Structure Data File (ENSDF) [13, 14]. The position of radioactive decays was simulated in various parts of the HPGe detector as shown in Fig. 1.

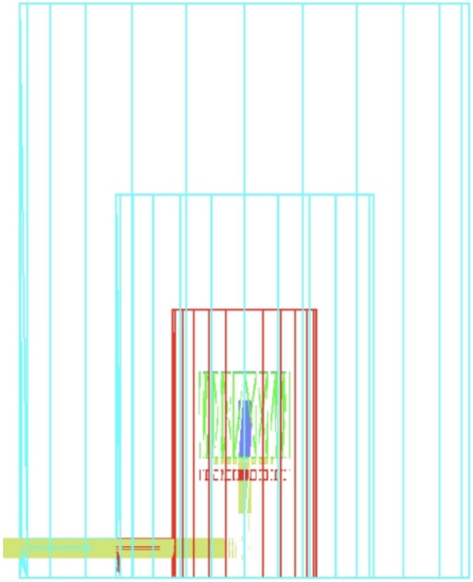

Fig. 1. Model of the HPGe detector used in GEANT4 simulations.

The production rate of the cosmogenic nuclide *j* at the Earth surface *D* is given as

$$P_{j(D,M,\ )} \quad \underset{i}{\ } N_i \quad \underset{k}{\ } \underset{0}{\ } \ _{ijk}(E_k)\ J_k(E_k,D,M,\ )dE_k \tag{1}$$

where N_i is the number of atoms in the target element *i* per kg of material, $_{ijk}(E_k)$ is the cross section for the production of the nuclide j from the target element *i* by particles of type *k* with energy E_k, and $J_k(E_k, D, M,\ )$ is the total flux of particles of type *k* with energy E_k at location *D* inside the atmosphere for the geomagnetic field M and the solar modulation function . The particle fluxes $J_k(E_k,D,M,\ )$ were calculated using the GEANT [21] and MCNP [15] codes. A description of the interface between these two codes is given in [16]. The cross sections $_{ijk}(E_k)$ were those evaluated from many measurements and used in earlier calculations [16]. The solid Earth was considered as a sphere with a radius of 6378 km with a surface density of 2 g cm^{-3}, and an average chemical composition. The Earth's atmosphere was modeled as a spherical shell with a thickness of 100 km. The atmospheric density and temperature were approximated by the U.S. Standard Atmosphere, 1976, model [17]. The primary cosmic ray flux at the Earth's orbit has two components: galactic (GCR) and solar (SCR). The GCR particles are a mixture of ~87% protons, ~12% alpha particles and ~1% of heavier nuclei with atomic numbers from 3 to ~90 [18]. The approximation [19] for primary galactic cosmic ray spectra have been used in the calculations. In the simulations within the Earth's magnetic field, alpha particles have to be treated separately. This is due to the different geomagnetic effects on primary protons and alpha particles. From the fitting of lunar experimental data [20], the effective flux of protons including the contribution of alpha particles with energies above 10 MeV at 1 A.U. was determined to be 4.56 nucleons cm^{-2} s^{-1}. Solar cosmic rays have relatively low energies, therefore their contribution to the production is not considered in these simulations. Technical details of the codes, used cross sections and their application to the cosmogenic nuclide production rate calculations are given in [16, 21], where contributions to uncertainties of these calculations are also discussed. Statistical uncertainties of the simulations were about 5%, while the systematic ones were of the order of 10%.

3. Results and discussion

The Monte Carlo simulations were focused on the Obelix HPGe detector which is operating in the Laboratoire Souterrain de Modane, LSM (France) at the depth of 4800 m w.e. (water equivalent) [22, 10]. It is p-type coaxial detector with diameter of 93.5 mm and length of 89.6 mm. The useful volume is 600 cm^3 and the relative detector efficiency is 162%. The resolution of the detector is 1.12 keV at 122 keV, and 1.98 keV at 1332 keV. The cryostat and the detector holder are made from Al-4%Si alloy; the thickness of entrance window is 1.6 mm. The oxygen free high conductivity copper was used for production of the cold finger. The inner shield (thickness of 12 cm) of the HPGe detector was made from Roman archeological lead. The outer shield (thickness of 20 cm) was made from low activity lead [10, 22]. The production of cosmogenic radionuclides was simulated in the Al-Si cryostat, Al-Si detector holder, Ge crystal and in the Cu cold finger.

3.1. Monte Carlo calculations of production rates of cosmogenic radionuclides in the cryostat and detector holder

The total production rates [atom/g-element/year] for high latitudes and sea level are given for two radionuclides of highest interest as:

$$^{22}\text{Na} = 102[\text{Na}] + 175.1[\text{Mg}] + 62.4[\text{Al}] + 41.7[\text{Si}] + 1.8[\text{Ca}] + 0.187[\text{Fe}]$$

$$^{26}\text{Al} = 0.20[\text{Mg}] + 195.52[\text{Al}] + 73.52[\text{Si}] + 0.26[\text{Fe}].$$

The target-element concentrations are given in weight fractions. The calculated production rates of cosmogenic radionuclides for Al-4%Si alloy are listed in Table 1. The measured ^{26}Al activity in the HPGe detector (0.38±0.19 µBq kg^{-1}) corresponds to about 60 y of exposure to cosmic rays at sea level. The ^{22}Na activity of the detector has been saturated at 1000 µBq kg^{-1}.

Table 1. Production of ^{22}Na and ^{26}Al in the cryostat and the detector holder.

Radionuclide	Half-life (y)	Production rate [atom kg^{-1} day^{-1}]	Measured activity [µBq kg^{-1}]
^{26}Al	7.17 10^5	**530**	0.38±0.19[*]
^{22}Na	2.60	**170**	1000[$]

[*] Measured in [28]

[$] Calculated in this work

3.2. Production of cosmogenic radionuclides in copper

Copper is very often used in low background experiments as a part of the HPGe detector or as a shielding, and several papers have already been dealing with calculation of production rates of cosmogenic radionuclides in copper [23–25]. The activity of a cosmogenic radionuclide in copper or another construction material after its exposure to cosmic rays may be calculated as

$$A \quad \frac{1 \quad e^{\ln(2)\frac{t_{\exp}}{T_{1/2}}}}{3600 \quad 24},$$

where t_{exp} is the exposure time, P is the production rate and $T_{1/2}$ is the half-life of the radionuclide. Table 2 lists the most relevant cosmogenic radionuclides (^{22}Na, ^{46}Sc, ^{54}Mn, ^{59}Fe, ^{56}Co, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn) which are produced in copper. The saturation in activity is reached after about three half-lives after the exposure.

3.3. Production of cosmogenic radionuclides in germanium

Many experiments searching for rare nuclear events have used germanium crystals as the detector, therefore the activation of natural and enriched germanium has been object of many studies [23, 26, 27]. The production rate and calculated activity after exposure is shown in Table 3.

Table 2. Production of cosmogenic radionuclides in copper.

Radionuclide	Half-life	Production rate [24] [atom kg^{-1} day^{-1}]	Measured activity [24] [μBq kg^{-1}]
^{46}Sc	83.787 d	2.67	$27\,^{11}_{9}$
^{54}Mn	312.19 d	15	$154\,^{35}_{34}$
^{59}Fe	44.494 d	4.6	$47\,^{16}_{14}$
^{56}Co	77.236 d	11	$108\,^{14}_{16}$
^{57}Co	271.82 d	51	$519\,^{100}_{95}$
^{58}Co	70.85 d	79	$798\,^{62}_{58}$
^{60}Co	5.2711 y	94	$340\,^{82}_{68}$
^{65}Zn	244.3 d	2.04*	20$
^{22}Na	2.60 y	0.014*	0.14$

*Calculated in [25]

$Calculated in this work

Table 3. Production of cosmogenic radionuclides in germanium.

Radionuclides	Half-life	Production rate [27] [atom kg^{-1} day^{-1}]	Calculated activity [μBq kg^{-1}]
^{54}Mn	312.3 d	5.2	50
^{3}H	12.3 y	48	490
^{65}Zn	244.3 d	63	640
^{57}Co	271.8 d	7.6	80
^{68}Ge + ^{68}Ga	270 d	60	610

3.4. Comparison of calculated and measured gamma-spectra

A comparison of simulated and measured HPGe background gamma- spectra is shown in Fig. 2. The Monte Carlo spectrum in the energy region of 40–1200 keV is in reasonable agreement with the experimental one, except for the energies below 200 keV, which may be associated with underestimation of contributions from bremsstrahlung, formation of delta-electrons and production of Pb X-rays. The calculated peaks of cosmogenic radionuclides (^{57}Co, ^{65}Zn) in the gamma-spectrum are within a factor of two comparable with the measured ones. The simulated high-energy part of the gamma-spectrum (1200–3000 keV, Fig. 2) generally follows the measured spectrum. The simulated cosmogenic ^{22}Na and ^{26}Al peaks agree within a factor of two and four, respectively, with the measured ones.

3.5. Decay of cosmogenic radionuclides during operation of HPGe detector in underground laboratory

The decay rates of cosmogenic radionuclides with time after operation of HPGe detector in underground laboratory have been observed. The total number of counts decreased from 150 kg^{-1} day^{-1} for the energy interval of 40–3000 keV measured after 10 months of operation underground [10], to 85 kg^{-1} day^{-1} measured after 40 months [28] of the installation of the detector in the underground laboratory.

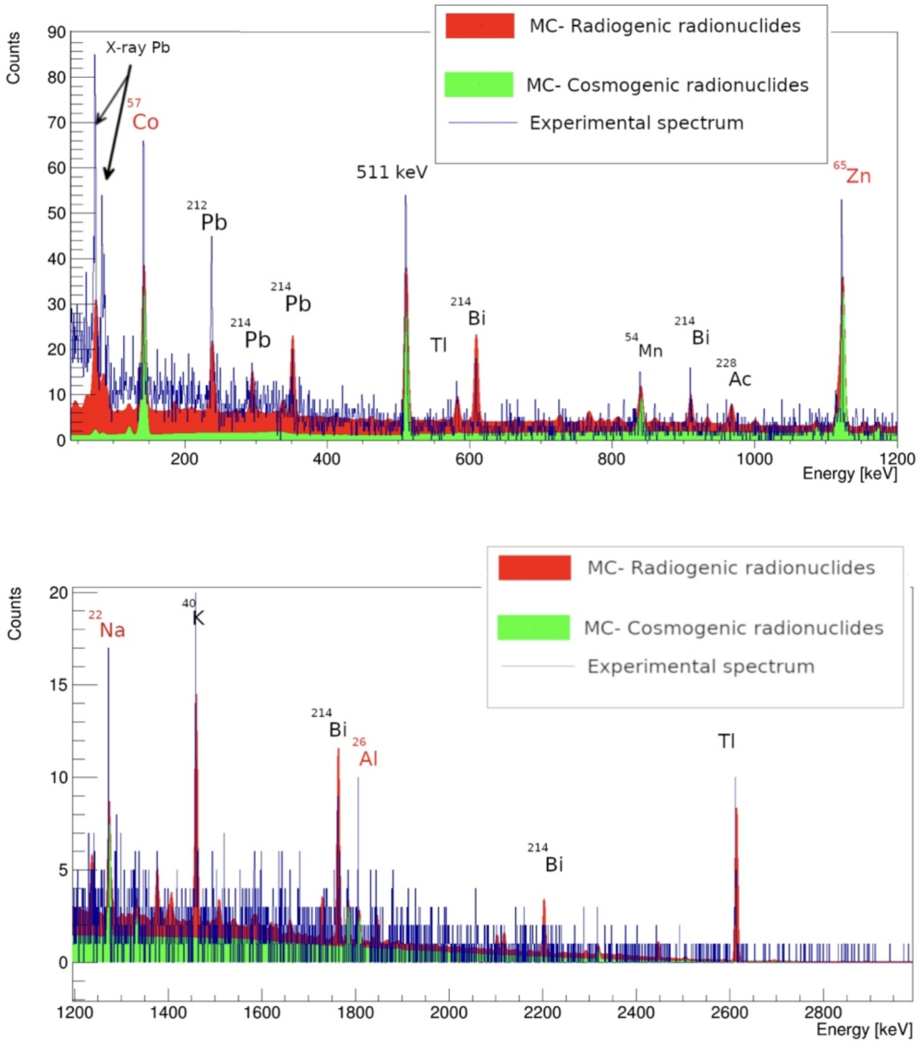

Fig. 2. Comparison of Monte Carlo simulated and measured background gamma-spectra ten months after installation of the HPGe detector in the Modane underground laboratory (counting time was 34 days).

4. Conclusions

A background simulation model based on GEANT4 software package was used for calculation of production rates of cosmogenic radionuclides in HPGe detector operating in Modane underground laboratory (France). Production rates of several cosmogenic radionuclides produced by interactions of neutrons with Ge crystal (^{3}H, ^{54}Mn, ^{57}Co, ^{65}Zn, ^{68}Ge), with copper (^{46}Sc, ^{54}Mn, ^{56}Co, ^{57}Co, ^{58}Co, ^{59}Fe) and with Al+Si alloy (^{22}Na, ^{26}Al) were calculated. The short-lived cosmogenic radionuclides contribute to the detector background mainly during the first year of its operation in an underground laboratory.

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