Cosmogenic activation of Germanium and its reduction for low background experiments

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Abstract

Production of $^{60}$Co and $^{68}$Ge from stable isotopes of Germanium by nuclear active component of cosmic rays is a principal background source for a new generation of $^{76}$Ge double beta decay experiments like GERDA and Majorana. The biggest amount of cosmogenic activity is expected to be produced during transportation of either enriched material or already grown crystal.

In this letter properties and feasibility of a movable iron shield are discussed. Activation reduction factor of about 10 is predicted by simulations with SHIELD code for a simple cylindrical configuration. It is sufficient for GERDA Phase II background requirements. Possibility of further increase of reduction factor and physical limitations are considered. Importance of activation reduction during Germanium purification and detector manufacturing is emphasized.

Key words: Double beta decay; Spallation; Excitation functions

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1 Introduction

Search for neutrinoless double beta decay (DBD) is of fundamental importance for Physics. As follows from oscillation experiments, neutrino is a massive particle. If the neutrino mass has Majorana nature, neutrinoless DBD may be observed [1]. Development of germanium semiconductor detectors, availability of enriched material, and sufficiently high transition energy make \(^{76}\text{Ge}\) one of the best nuclei for neutrinoless DBD search. First indication of \(^{76}\text{Ge}\) neutrinoless DBD is done in [2]. GERDA is a new \(^{76}\text{Ge}\) DBD experiment, recently accepted at LNGS [3]. Another advanced project, Majorana is ready to be started in US [4]. Background analysis of both the experiments has shown, that the principal background source is the internal activity of Germanium crystals due to cosmogenic isotopes of \(^{60}\text{Co}\) and \(^{68}\text{Ge}\) [3,4,5,15]. Without special efforts aimed to reduction of cosmogenic activation, background index in the energy range near \(Q_{\beta\beta}\) is hard to be pushed below \(0.01 - 0.02\, \text{cpy/keV/kg}\) [3] (here cpy stands for counts per year). The biggest amount of cosmogenic activity is expected to be produced during transportation of the enriched material to crystal growth facility, then to detector manufacturer and finally to the laboratory.

In order to achieve the level of total background index of \(10^{-3}\, \text{cpy/kg/keV}\), required for the GERDA experiment we propose to use a movable shielding container for germanium transportation. In this paper we report simulation results for a sample configuration of such a container. Possibility of shielding properties optimization and limitations on feasible reduction factor for cosmogenic activations are discussed.

Relevant physical processes

At the Earth surface, formation of radioactive isotopes is caused mostly by spallation reactions of fast nucleons from cosmic rays. Smaller contributions are due to capture of stopped negative muons and muon induced fast neutrons.

Only one dangerous isotope — \(^{60}\text{Co}\) may be produced after muon capture. However the probability of this channel should be much less than \(10^{-4}\), see e.g. [6]. Muon capture rate at the sea level is \(\approx 10^{-6}\, 1/g/s\) [7]. So \(^{60}\text{Co}\) production rate should be much less than \(10^{-2}\, 1/kg/day\). Contribution of muon induced fast neutrons may be roughly estimated using the result of Cocconi [8], that only about 2% of nuclear disintegrations by cosmic rays are due to muon induced neutrons. Thus about 98% of cosmogenic activations are produced by nuclear active component (N-component) of cosmic rays and might
be attenuated by movable iron shield. It is necessary to stress, that muon
induced contribution practically does not decrease in a shield of about 1000
\( \text{g/cm}^2 \) thickness.

Composition of N-component of cosmic rays at sea level is the following:
more than 95% neutrons about 3% protons and about 2% pi-mesons [9,10].
Though production rates of neutrons and protons in nuclear disintegration
cascades are close to each other, the flux of protons is generally smaller and
harder, because protons are stopped more efficiently than neutrons due to
electromagnetic losses.

For our study we used the energy spectra and fluxes of neutrons and
protons from [11], fig.1. Angular distribution is supposed to be proportional
to \( \cos^{3.5}\theta \), where \( \theta \) is zenith angle. Analysis of uncertainties of the flux and
spectral data may be found in [10].

Understanding of N-component propagation and attenuation in matter
is crucial for an effective shield design. There are a lot of papers devoted to
attenuation of N-component of cosmic rays in atmosphere, water and some
other substances [12,8,?] . Special efforts were applied to measurements and
simulation of artificial fast neutrons fluxes attenuation in matter [13].

Typical attenuation lengths for N-component in some of materials are
shown in the table. 1. [12,10]. Note however, that attenuation length might be
correctly defined only for equilibrium energy spectrum.

One can see that attenuation length increases with A. This can be easily
understood because interaction cross sections for reactions in a hadronic
cascade are roughly proportional to \( A^{0.66...0.8} \) [14]. It worth mentioning, that
for selection of a material for movable shielding not only attenuation length
of N-component, but also the density are important. Increase of density leads
to decrease of the required mass of a container.

From semi-empirical consideration we have chosen iron as an optimal
material for shielding container.

Another important entry point for simulation of cosmogenic background
reduction is knowledge of partial cross sections for production of \(^{60}\text{Co}\) and
\(^{68}\text{Ge}\) in spallation reactions.

There exist a few physical models describing the spallation process. [17].
Some of the models together with compilation of experimental data are im-
plemented in the nucleon transport codes like LAHET, SHIELD, FLUKA etc.
General analysis of the most diffused codes is done in [17], see also [15].
The tool: SHIELD code

An appropriate nucleon transport code may be used for simulation of both hadron propagation and radioactive isotope production. Our choice is the SHIELD transport code.

The SHIELD transport code have been elaborated as an universal tool for study of the interaction of high energy particles with a matter. The SHIELD code includes the Russian models of nuclear reactions, developed at JINR (Dubna) and INR RAS (Moscow), providing simulation of all stages of inelastic nuclear interactions in the exclusive approach. The SHIELD code have been benchmarked extensively and showed good agreement with experimental results for multiple phenomena.

The modern version of the SHIELD code [18,19] allows to simulate interaction of nucleons, pions, kaons, antinucleons, muons and arbitrary (A,Z) nuclei with complex extended targets in energy range up to 1 TeV/nucleon. The ionization losses, straggling and multiple Coulomb scattering are taken into account. Transport of neutrons below 14.5 MeV is simulated by means of the original neutron transport code LOENT based on a 28 group neutron data system ABBN [16].

Capabilities of a hadron transport code depend substantially on the generator of inelastic nuclear interactions used. In the SHIELD code the MSDM generator (Multy Stage Dynamical Model) [20] is used. Details about used physical models may be found in refs.[21,22,23,24,25,27,26]

2 Results and discussion

Simulation details

An iron container shown in the fig.2 was designed for the transportation of enriched germanium for the Phase II of GERDA experiment. Container size is $\phi 140$ cm x $126.5$ cm There is a cavity in the container $\phi 54$ cm x $40$ cm. The cavity is situated in such a way that the bottom thickness is $15$ cm.

Total mass of the container is $14.5$ tons. In the central part of the cavity a germanium cylinder $\phi 42$ cm x $27$ cm is placed as a target. In the simulation model a 3 m thick slab of ground was included. The container is placed 1.2 m above ground, as it were in a truck.
Entire configuration consisting of container and germanium target has been simulated. For understanding of reduction factors activation of Germanium target without iron container was also simulated.

In order to make possible a comparison of our simulation with literature, and for giving a key to the shield optimization we discuss in addition two issues:

1. Excitation functions for $^{60}$Co and $^{68}$Ge production by neutrons and protons and
2. Shielding properties of the container including energy spectra of nucleons inside the cavity

Isotope production rates

In the tables 2,3 $^{68}$Ge and $^{60}$Co production rates are reported for all the stable isotopes of Ge with and without the shielding container. Using this table one can predict activation rate and reduction factor for any isotope composition, e.g. for enriched $^{76}$Ge (87% $^{76}$Ge +13% $^{74}$Ge) reduction factors are expected to be 17 for $^{60}$Co and 10 for $^{68}$Ge. Taking into account limitation due to muon interactions total reduction factor will be about 13 for $^{60}$Co and about 8.5 for $^{68}$Ge. i.e. sufficient for GERDA phase II experiment.

Attention should be paid to the fact, that although proton flux at the sea level is only about 3% of neutron one, its contribution to isotope production is about 10% without shielding and up to 20% with shielding. Contribution of the sea level protons is not negligible due to hardness of their spectrum compare to neutron one.

Excitation functions

Excitation functions for production of $^{60}$Co and $^{68}$Ge by neutrons and protons on stable isotopes of germanium were generated with SHIELD code. Results for protons coincide within factor 1.5-2 with experimental data from [28,29]. The most important for our application curves are shown in the fig.3.

Previous estimations of cosmogenic activation of germanium detectors were done using excitation functions calculated with ISABEL code [5], those results were 2-6 times lower than ours, however such discrepancy between
different methods is typical for this kind of simulation. It is important, that accuracy of activation reduction factor of a shield does not suffer from these discrepancies.

Spectra and fluxes of nucleons

The most important characteristics of a radiation field inside the cavity is differential flux density of nucleons ($J_N(\varepsilon)$). In the fig.4 such curves are shown. The first approximation calculations may take into account only neutron spectrum at the sea level and only neutrons inside the cavity. However, one can see, that contribution of the sea level protons to generation of nucleons inside the cavity is more than 15% even though their flux is only 3% of total sea level flux. This statement is consistent with results of the tables 2,3.

Production rate $R_i$ of ($i$) radioactive isotope may be found with sufficient accuracy (in our case discrepancy is less than 10%) using the following formula:

$$R_i = \sum_N \sum_j N_j \int J_N(\varepsilon)\sigma_{ijN}(\varepsilon)d\varepsilon,$$

where $N_j$ is a number of ($j$) target nuclei, $\sigma_{ijN}(\varepsilon)$- excitation function for ($i$) product at ($j$) target by $N$ ($N= n,p$) projectile.

It is interesting to compare attenuation length for N-component obtained in our simulation with other data. In the fig.5 energy differential fluxes of nucleons crossing the upper plane of the cavity are shown for two configurations – with and without iron container. One can see that unique attenuation length can not be introduced for both neutron and proton component and for the whole energy range. It means that spectrum is not an equilibrium one, three main reasons could be pointed out for this fact: systematic errors of determination of spectra in [11], systematic errors of simulations (including high energy cut), and different equilibrium spectra of hadronic shower in air and iron.

One can see that the maximum attenuation length for neutrons is about $240 \text{ g/cm}^2$ at 80 MeV - 200 MeV energy range. Normally attenuation length is measured for neutrons with energy below 50 MeV. Thus, taking into account energy dependence of attenuation length, agreement of our result with table 1 is quite good.

Most of the activations are produced by neutrons with energy around 100 MeV, hence for conservative estimation of shielding properties of an iron container attenuation length of $240 \text{ g/cm}^2$ should be used.
Information about spatial distribution of nucleons in the cavity is useful for a container shape optimization. In the fig.6 energy differential fluxes of nucleons from top, lateral, and bottom surfaces of the cavity are shown.

The most intensive (per unit area) and hardest spectrum comes from the top surface. Spectrum from bottom is the least intensive and soft. From these curves the first shape optimization is rather obvious – iron disk from the bottom part can be removed and thickness of the top part may be increased, keeping the container mass constant. Simulation of such a modified container was done. In the fig.6 total fluxes into the cavity are shown for the two configurations. One can observe $\approx 20\%$ decrease of nucleon fluxes. Taking into account muon induced contribution, activation reduction for a container with thicker top part are 10 and 15 for $^{68}$Ge and $^{60}$Co respectively.

The last configuration will be used for transportation of $^{76}$Ge for GERDA experiment.

Prospects

Further development of $^{76}$Ge DBD experiments will request for few hundred kilograms of target isotope and background index better than $10^{-3}$ cpy/keV/kg. The last objective may be reached by combination of sophisticated background rejection techniques and more efficient shielding against cosmogenic activations.

We have shown that significant reduction of activation is possible during transportation. Now the biggest contribution should arrive during crystal growth and detector manufacturing. Construction of stationary shielding above technological equipment should be considered as a next step in reduction of cosmogenic background.

Besides, shape of the container may be optimized within fixed mass. It is also necessary to understand better the limitation due to muon-nuclear interactions. In particular, dependence of secondary fast neutron yield on the material should be investigated.
3 Conclusions

Movable iron shielding container is proposed for reduction of cosmogenic activations of $^{76}\text{Ge}$ for DBD experiment. Relevant physical processes are considered. Semi-empirical statements useful for a container design optimization are formulated. Simulation of a simple cylindrical configuration is performed. Estimation of limitations due to interactions of energetic muons is done. Expected reduction factors are 10 and 15 for $^{68}\text{Ge}$ and $^{60}\text{Co}$ production respectively. The proposed container is built and is being used for germanium transportation.

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Table 1
Cosmic Ray neutrons attenuation lengths

| Material   | $\lambda$, g/cm$^2$ |
|------------|---------------------|
| Air        | 140-160             |
| concrete   | $\approx$170        |
| Iron       | $\approx$200        |
| Lead       | $\approx$300        |
68Ge production rates (per day, per 1 kg), statistical standard deviations are shown in parentheses.

| target | total  | by sea level protons |
|--------|--------|----------------------|
|        | no shield | shield | no shield | shield |
| 70Ge   | 281.4 (0.5%) | 33.0 (2%) | 17.17 (1.1%) | 4.90 (1.5%) |
| 72Ge   | 55.34 (1.4%) | 6.20 (4%) | 4.78 (2%) | 0.96 (3%) |
| 73Ge   | 28.0 (1.3%) | 2.94 (7%) | 2.54 (3%) | 0.45 (6%) |
| 74Ge   | 14.53 (2%) | 1.46 (8%) | 1.48 (4%) | 0.24 (6%) |
| 76Ge   | 4.22 (4%) | 0.4 (8%) | 0.54 (6%) | 0.06 (12%) |
Table 3

$^{60}$Co production rates (per day, per 1 kg), statistical standard deviations are shown in parentheses.

| target | total | by sea level protons |
|--------|-------|----------------------|
|        | no shield | shield | no shield | shield |
| $^{70}$Ge | 1.73 (7%) | 0.118 (33%) | 0.170 (11%) | 0.028 (19%) |
| $^{72}$Ge | 2.88 (6%) | 0.256 (19%) | 0.285 (9%) | 0.046 (14%) |
| $^{73}$Ge | 3.14 (4.0%) | 0.265 (24%) | 0.335 (8%) | 0.035 (21%) |
| $^{74}$Ge | 3.35 (4%) | 0.23 (21%) | 0.380 (8%) | 0.050 (14%) |
| $^{76}$Ge | 3.31 (4%) | 0.156 (13%) | 0.455 (7.0%) | 0.036 (15%) |
FIGURE CAPTIONS

Fig. 1. Nucleon flux density spectra at the sea level [11]. Asterisks – neutrons, open triangles – protons.

Fig. 2. Outline of the iron shielding container

Fig. 3. Excitation functions for $^{60}$Co and $^{68}$Ge production by neutrons on stable isotopes of Ge.

Fig. 4. Nucleon flux density spectra inside the cavity. Open triangles – neutrons, asterisks – neutrons from sea level neutrons only, open circles – protons, black triangles – protons from sea level neutrons only.

Fig. 5. Flux of nucleons through the top surface of the cavity in two configurations – with and without iron container. Black squares and circles – sea level neutrons and protons respectively, asterisks and black triangles – neutrons and protons inside the cavity.

Fig. 6. Fluxes of nucleons in the cavity: total and from different sides of the cavity.
Fig. 1.
Ground, depth = 4 m

Air gap, thickness = 120 cm

Cavity, R=27 cm, H=40 cm

Ge shipment R=21 cm, H=27 cm

Bottom thickness = 15 cm

Iron container R=70 cm H=126.5 cm

Normalization sphere II, R= 20 cm

Normalization sphere I, R= 150 cm

Fig. 2.
Fig. 3.
Fig. 5.
Fig. 6.

Neutron flux in cavity, 1/MeV/s

- total
- lateral
- top
- bottom

Line - total optimized