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Optimization of the Transport Shield for Neutrinoless Double-beta Decay Enriched Germanium

E Aguayo RT Kouzes
JL Orrell DJ Reid
JE Fast

April 2012



Pacific Northwest
NATIONAL LABORATORY

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Richland, Washington 99352

Summary

This document presents results of an investigation of the material and geometry choice for the transport cosmogenic shield of enriched germanium, the active detector material used in the MAJORANA DEMONSTRATOR neutrinoless double-beta decay search. The objective of this work is to select the optimal material and geometry to minimize cosmogenic production of radioactive isotopes in the germanium material during transport. The design of such a shield is based on the calculation of the cosmogenic production rate of isotopes that are known to cause interfering backgrounds in enriched germanium neutrinoless double-beta decay searches. As part of this study, we will examine the reliability of estimates of cosmogenic activation from this study under different shielding scenarios.

This study utilizes Monte Carlo techniques to simulate the transport and attenuation of the incident cosmic ray particles in the shielding material and the nuclear reactions in the germanium. This method of calculating production rates relies completely on the accuracy of the simulation model and library data. The calculated production rate of an isotope is the product of the energy-dependent cross section of the reaction of interest and the incoming cosmic ray produced flux of particles, integrated over the relevant energy range. The production of ^{68}Ge from the various germanium isotopes have Q values starting at approximately 20 MeV, so the energy range of interest in this study is above 20 MeV. Based on the transport shield design used by GERDA and the MAJORANA DEMONSTRATOR, we present one modification to the shield that will further reduce the production rate of undesired isotopes ^{68}Ge and ^{60}Co and still comply with requirements of official transportation standards. The conclusion, based on Monte Carlo models, is that increasing the amount of iron to the container's maximum allowable weight results in a shield that would be an order of magnitude more efficient in the attenuation of the production of isotopes of interest for neutrinoless double-beta decay searches than the current GERDA transport shield design.

Acronyms and Abbreviations

$0\nu\beta\beta$	Neutrinoless double-beta decay
CMS	Compact Muon Solenoid
Geant4	Geometry and Transport 4
GERDA	European ^{76}Ge experiment
HPGe	High-purity germanium
LHC	Large Hadron Collider
MJD	MAJORANA DEMONSTRATOR
PNNL	Pacific Northwest National Laboratory

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1.0 Introduction

The search for neutrinoless double-beta decay ($0\nu\beta\beta$) is of fundamental importance for physics (Elliott and Vogel 2002) to determine if neutrinos are their own antiparticle. This study is part of collaborative work to answer this question by designing a tonne-scale experiment to search for the process of $0\nu\beta\beta$ in germanium enriched in the ^{76}Ge isotope. One of the keys to this experimental program is to minimize the intrinsic radioactive background to unprecedented levels and then to quantify the remaining background. At these ultra-low background levels, these experiments must account for potential sources of background due to cosmic activation in the detector material.

This experiment will be performed deep underground in order to shield against cosmic ray showers. At the depth where these experiments are performed (several thousand meters of water equivalent), the cosmic ray neutron and proton flux is entirely shielded by the ground's overburden, and only high-energy muons will remain. However, prior to deployment, the detector material will be above ground, exposed to all components of cosmic ray showers. At the Earth's surface, formation of radionuclides is caused mostly by spallation reactions by fast nucleons from cosmic rays. The neutron component is responsible for approximately 85% of the nuclear disintegrations that are induced by cosmic ray showers, with the rest due mostly to protons (Barabanov et al. 2006). This work focuses on improving transport shielding to reduce neutron-induced cosmogenic production in germanium detector material.

The enriched germanium detector material for a $0\nu\beta\beta$ experiment is only produced by a separations plant in Russia. The material then travels overseas to reach the crystal growth facility in the U.S. Further ground transport is required to take the material to the detector manufacturer and finally to the underground laboratory where the experiment will be conducted. Before reaching the underground facility, the detector material will be exposed to all the particle components of cosmic showers at or above sea-level. The neutron and proton components of the cosmic rays are attenuated after a few meters of water equivalent shielding, so once the detector material is in the deep underground site, these components of the cosmic shower will no longer contribute to backgrounds in the detectors.

The European ^{76}Ge $0\nu\beta\beta$ experiment (GERDA) used Monte-Carlo simulations to design a transport shield, using data from the simulation tools ISABEL and SHIELD (Barabanov et al. 2006). Their work resulted in a movable iron shield with cylindrical shape. Their design is the basis for this optimization study, although instead of using two separate codes, this work is based on the physics simulation and transportation tool Geant4 (Agostinelli et al. 2003).

The study of cosmogenic activation at sea-level was previously started by simulation of three different particles present in cosmic rays: neutrons, muons and protons (Aguayo-Navarrete et al. 2010). This report draws heavily from the conclusions reached in this referenced work. The ultimate goal of this study is to accurately determine the best shielding against cosmogenic production in germanium during transport. This study will be completed once the present calculations are benchmarked against experimental measurements of the quantities studied. To a first approximation, cosmogenic activation due to factors other than cosmic neutrons (protons, muons) is <15% of the total production rate (Barabanov et al. 2006); in other words, a conservative assumption would be that cosmic protons and neutrons generated by muons in the shield are responsible for <15% of the total cosmogenic production. Figure 1 shows the differential neutron cross section used by the simulation tool. The main mechanism of energy loss for cosmic neutrons below 20 MeV in the shield is elastic scattering. Above this energy, inelastic scattering

dominates. Cosmic protons behave in the same manner, as shown in Figure 2. These two plots show comparable nuclear reaction cross sections for protons and neutrons with energies >10 MeV which can lead to activation of germanium, since elastic scattering at any energy would not lead to activation. One can assume conservatively that the activation induced by protons is expected to be on the order of magnitude of that for neutrons. Because the incident proton flux is much lower than the incident neutron flux over the energy range of interest the proton component is not expected to be significant compared to the neutron component. The cosmogenic production due to neutrons generated by muons were simulated in (Aguayo-Navarrete et al. 2010). In that study it was shown that the neutron production in shielding materials by muons is three orders of magnitude smaller in the energy region of interest, so it is assumed that in the different shielding scenarios considered in this work, these neutrons will remain a second order contributor to the cosmogenic production. Because of the dominance of the cosmic ray neutrons, the remainder of this paper focuses on this contribution only.

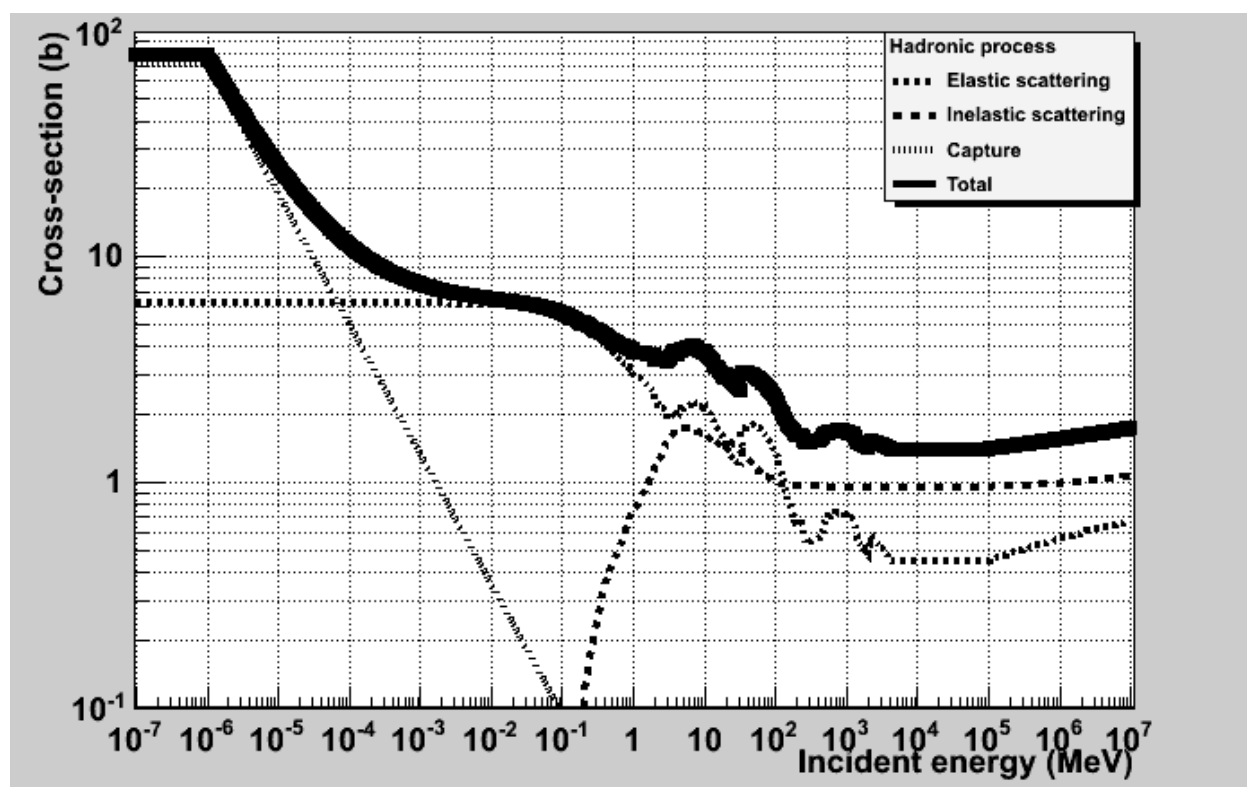


Figure 1: Differential neutron cross section in isotopically pure germanium 72. Parameterization extracted from the simulation tool used in this study.

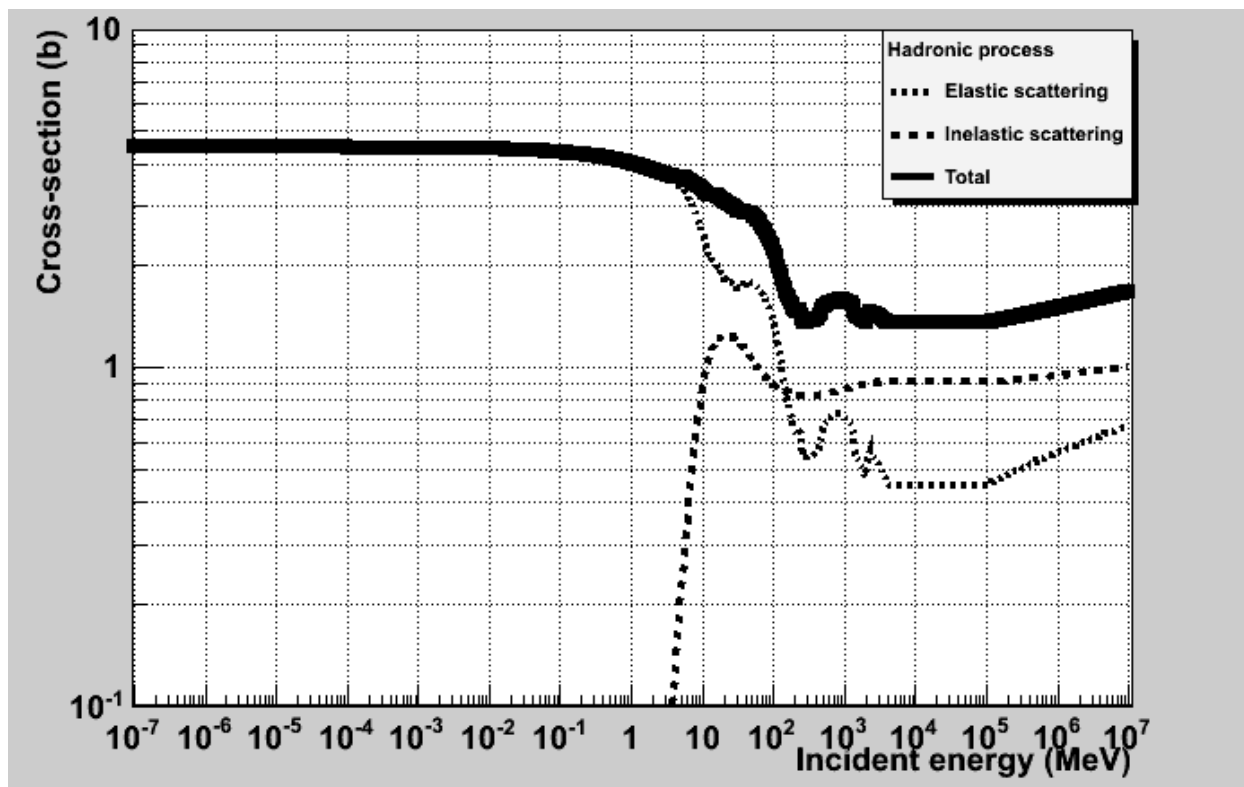


Figure 2: Differential proton cross section in isotopically pure germanium 72. Parameterization extracted from the simulation tool used in this study.

This document is divided into six sections. Section 2 offers a comparison of the simulation results reported in the literature for the GERDA shield with those found using the simulation tool described in this paper. Section 3 describes the tools used in this work to compute the cosmogenic production rates. In Section 4 the movable shield used by GERGA for overseas transport and the proposed shield modification is described. Section 5 presents a comparison of the production rates computed in this work with those reported in the literature.

2.0 Cosmic rays and physical reactions

At the Earth's surface, production of radionuclides is primarily caused by cosmic rays that undergo fast hadron spallation reactions. The neutron component represents more than 95% of the fast hadron shower at the Earth's surface (Ziegler 1998). The cross sections of the nuclear reactions which produce ^{68}Ge and ^{60}Co from sea-level fast neutrons are implemented in the Geant4-based Monte-Carlo simulation tool, described in (Aguayo-Navarrete et al. 2010). Table 1 lists the calculated Q-value energies for neutron-induced reactions for all stable germanium isotopes that are involved in a nuclear reaction that produces ^{68}Ge . Similarly, Table 2 presents the calculated Q-value energies for neutron-induced reactions in all stable germanium isotopes that produce ^{60}Co . These values assume no binding energy between the exiting nucleons. Thus, some production is possible at energies below these unbound Q-values. These results indicate that neutrons must have at least 20 MeV in kinetic energy to create the ^{68}Ge or ^{60}Co from the stable germanium isotopes.

Nuclear Reaction	Unbound Q value (MeV)
$^{70}\text{Ge} (n,3n)^{68}\text{Ge}$	-19.8
$^{72}\text{Ge} (n,5n)^{68}\text{Ge}$	-37.8
$^{73}\text{Ge} (n,6n)^{68}\text{Ge}$	-44.7
$^{74}\text{Ge} (n,7n)^{68}\text{Ge}$	-54.9
$^{76}\text{Ge} (n,9n)^{68}\text{Ge}$	-70.8

Table 1: Calculated Q values for the neutronic ^{68}Ge reactions considered in this work.

Nuclear Reaction	Unbound Q Value (MeV)
$^{70}\text{Ge} (n,5p\ 6n)^{60}\text{Co}$	-83.2
$^{72}\text{Ge} (n,5p\ 8n)^{60}\text{Co}$	-101.3
$^{73}\text{Ge} (n,5p\ 9n)^{60}\text{Co}$	-108.1
$^{74}\text{Ge} (n,5p\ 10n)^{60}\text{Co}$	-118.3
$^{76}\text{Ge} (n,5p\ 12n)^{60}\text{Co}$	-124.8

Table 2: Calculated Q value for the neutronic ^{60}Co reactions considered in this work.

2.1 Nuclear Reaction Cross Section

The nuclear reaction cross section of $^{70}\text{Ge}(n,3n)^{68}\text{Ge}$ was investigated in detail in order to compare the simulation tool performance against other reported work (Avignone, 1992) (Elliott, 2010) (Baravanov, 2006) (Cebrian, 2010). The effective cross section of Geant4 was extracted by determining the production rate of ^{68}Ge from monoenergetic neutrons impinging on a 2-cm thick ^{70}Ge sample. The neutron energies ranged from 20 MeV to 1 GeV. Figure 3 shows the effective cross section along with the cross section from other models. The simulation results show that the lowest neutron energy that can produce ^{68}Ge atoms is close to 20 MeV, in agreement with the calculated Q-value for the corresponding isotope reaction.

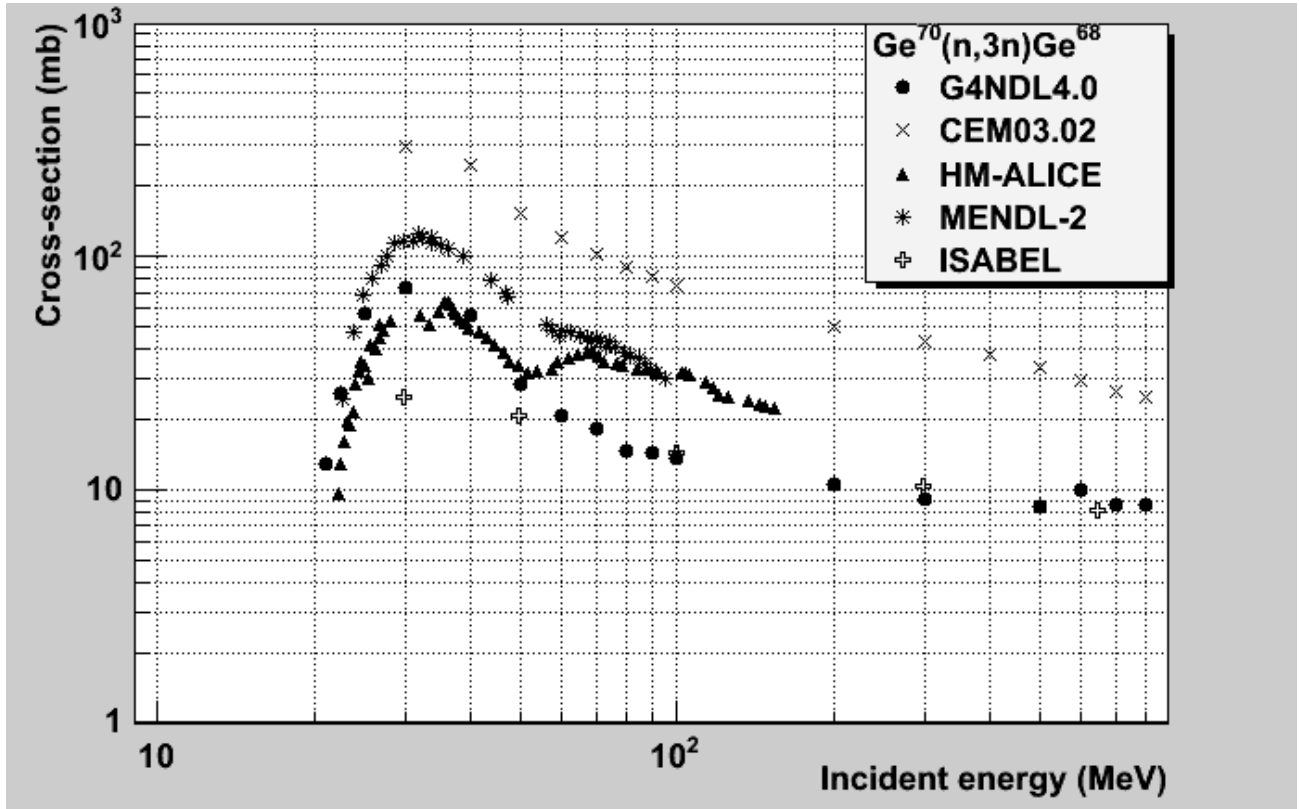


Figure 3: Comparison of cross sections for the reaction $^{70}\text{Ge}(n,3n)^{68}\text{Ge}$. See text for description of the models.

The nuclear cross section for the G4NDL4.0 library presented in Figure 3 was calculated using Equation 1:

$$\sigma(E) = \left(\frac{P_{G4}(E)}{I_n} \right) \left(\frac{A}{l \cdot N_O \cdot \rho} \right), \quad (1)$$

where

P_{G4} is the number of nuclei created in the simulation run,
 I_n is the total number of source particles simulated,

A is the atomic number of the target isotope,
 l is the thickness of the target in cm,
 N_0 is Avogadro's number,
 ρ is the density of the target in g/cm^3 .

This is the most recent nuclear data library released for Geant4 and used in this report. G4NDL data comes largely from the ENDF/B-VII library, which is developed and maintained by the Cross Section Evaluation Working Group (CSEWG, 2012). G4NDL data also comes from the JENDL (JENDL, 2012) library which is developed and maintained by Nuclear Data Evaluation Center of the Japan Atomic Energy Agency. The rest of the entries in Figure 3 are from cross section models used by other authors. The most modern one is CEM03.02 (Mashnik, 2001), a nuclear evaporation code applied to activation studies, which gives the largest cross section. MENDL (Yu, 1995) libraries are based on several versions of HMS-ALICE (Blann, 1996) codes, containing excitation functions, which cover a wide range of targets and product nuclides. ISABEL (Yariv, 1981) is a standard spallation reaction code, which is an extension of a compound nucleus reaction code. The large variation in magnitude of these cross sections produces uncertainty in the resulting production rate.

2.2 Cosmic neutron simulation

The third element in the calculation of production rate in different shielding scenarios is the attenuation and transport of the source particles in the shield material. Figure 4 shows the simulated transmission neutron flux after passing the neutron component of the cosmic ray showers in the energy range of interest for this shielding application (20 MeV to 10 GeV) (Aguayo-Navarrete et al. 2011) through different materials of varying thicknesses. From this graph, one can observe that iron and lead (high-Z materials) demonstrate the best attenuation of high-energy neutrons. This attenuation occurs largely by converting high-energy neutrons to low-energy neutrons that do not produce the undesirable reactions. Iron out-performs lead due to its lower atomic mass, which reduces secondary particle generation over the energies of interest. From this result it was concluded that additional iron would be the most efficient material to shield against cosmogenic production in the germanium shielding scenario. This conclusion agrees with that of (Baravanov, 2006).

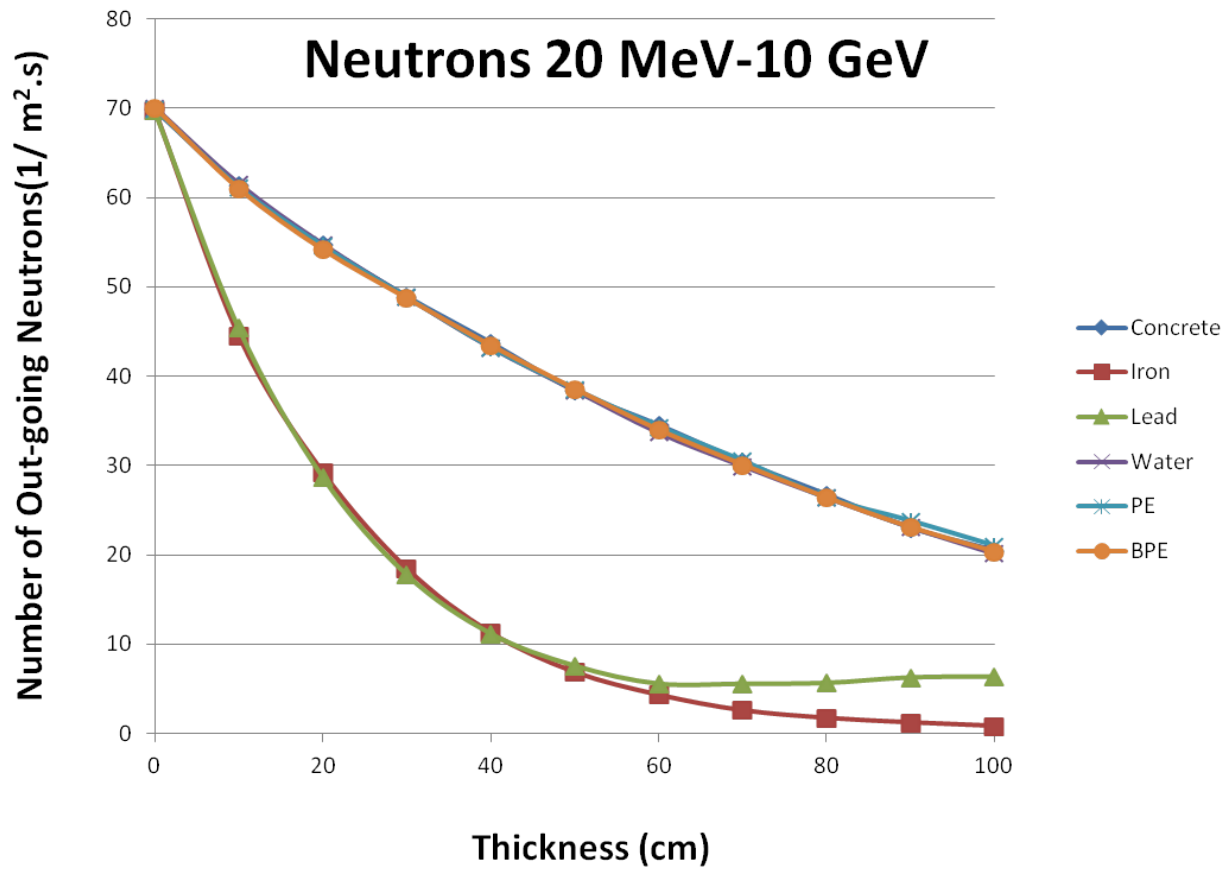


Figure 4: Transmitted flux of CRY sea-level cosmic neutron flux for different thicknesses of material, from (Aguayo-Navarrete et al. 2011)

3.0 Production Rate Calculation

3.1 Monte Carlo Tool Description

A combination of Geant4 and several parameterizations of the cosmic neutron flux at sea-level were used for the hadronic simulation tool to obtain the results presented in this work. This tool was intended to simulate hadronic interaction of cosmic rays impinging on a radiation shield. The tool has three components:

- Geant4 Toolkit
- Neutron flux parameterization (including CRY library)
- ROOT data analysis tool

A complete description of the tool can be found in (Aguayo-Navarrete et al. 2010). Table 3 presents the toolset for a Monte Carlo application based on Geant4 run in a Windows computing environment. The identified drawbacks of using this code are the questionable reliability of hadronic physics, which have not yet been validated with experimental data. In this work a variety of physics list are evaluated, including the QGSP_BERT_HP physics list, recommended by the Geant4 collaboration for high-energy physics calorimetry and shielding applications at all energies (Agostinelli et al. 2003).

	Version	Source
Geant4	9.3/9.4/9.5	Geant4.cern.ch
CRY	1.6	www.llnl.gov
ROOT	5.10	www-glast.slac.stanford.edu /software/root/walkthrough/install.htm
Cygwin	1.7.9-1	www.cygwin.com
Microsoft Visual Studio	2010 Express	http://www.microsoft.com /visualstudio/en-us/products/2010- editions/visual-cpp-express

Table 3: Software versions used in this work, from (Aguayo-Navarrete et al. 2010)

The specific composition of the detector material used in this study is given in **Error! Reference source not found.** (Kouzes et al. 2011).

Isotope	Fractional Composition
^{70}Ge	0.006
^{72}Ge	0.011
^{73}Ge	0.033
^{74}Ge	0.086
^{76}Ge	0.914

Table 4: Isotopic composition of enriched Ge detector material used in the simulations

3.2 Simulation source particle: sea-level cosmic neutrons

The representation of the neutron spectrum in Figure 5 is a standard lethargy plot. The vertical intensity is conceptually simpler to plot, but for neutrons, that typically requires a log-log plot covering many orders of magnitude on both axes, making details difficult to see. The large range of energies and intensity needed for neutrons stems from their characteristics when they slow down in a scattering medium. The lethargy plot shows the details of the number of neutrons in a more easily interpreted way.

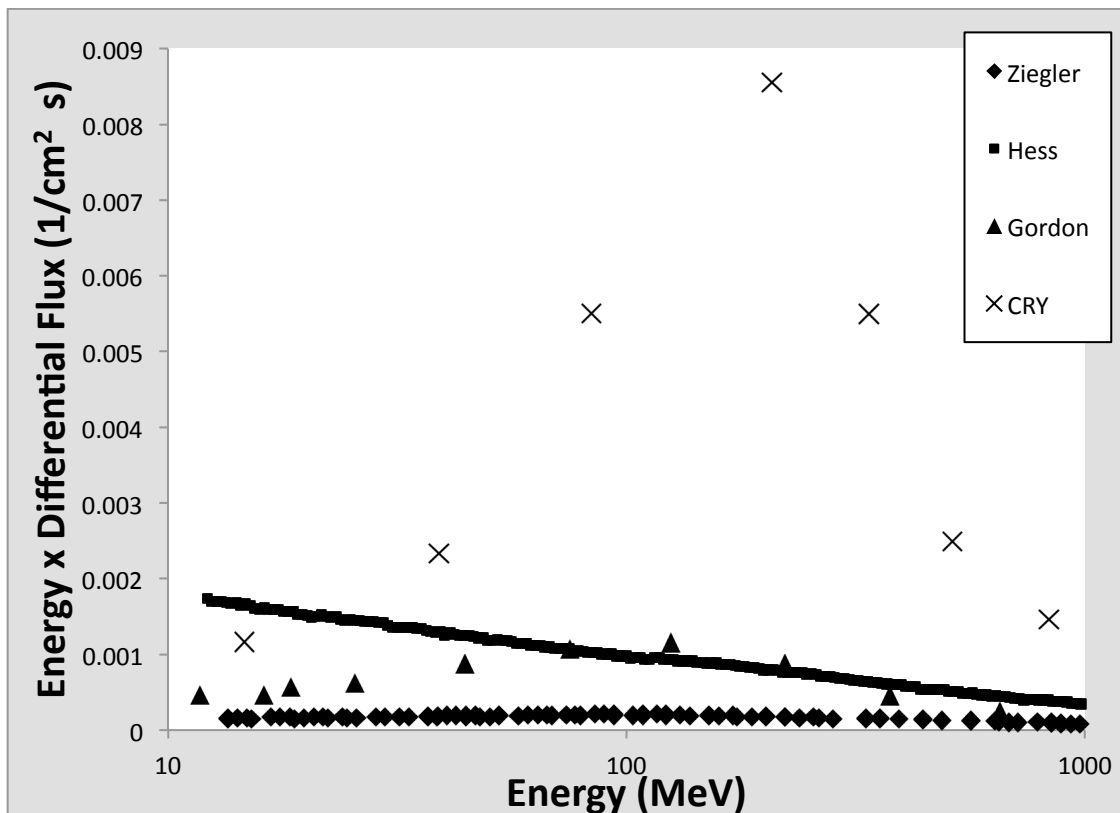


Figure 5: Different parameterization of the cosmic neutron spectrum at sea-level

Differences in data and models for the neutron component of cosmic rays at sea-level from different authors (Hess et al. 1959), (Ziegler 1998), (Kornmayer et al. 1995), (Gordon et al. 2004) are evident in the figure. The experimental methods used to derive each of the parameterizations presented in Figure 5 were very different. The first parameterization of the neutron component from cosmic rays was postulated in (Hess et al. 1959). This parameterization is based on airborne balloon measurements using a single ionization chamber. These first measurements were evaluated over time and eventually needed to be revisited as technology permitted new detectors with more accurate results. The second parameterization (in chronological order) was presented in (Ziegler 1998). The detector technology used was Bonner spheres of different sizes to characterize the differential neutron flux on top of a building in New York. The most modern measurement from (Gordon et al. 2004) was done with the goal of acquiring data over twelve decades of neutron energy, from meV to GeV, although only two decades are plotted in Figure 5. The neutron flux and energy spectrum were measured using an extended-energy Bonner sphere spectrometer. The CRY parameterization is the result of a Monte-Carlo simulation of the primary particles that generate the cosmic ray shower. High energy protons are simulated to interact in the atmosphere (Kornmayer et al. 1995) and the neutron flux parameterization is the result of a Monte-Carlo

simulation of this high energy protons interacting in the atmosphere. The fact that reported cosmic neutron parameterizations are so different must be considered when comparing these results, since the reported cosmogenic production rate is depends strongly on the differential neutron flux.

3.3 Calculation Methods for Cosmogenic Production Rates

Theoretical calculations of production rates of cosmogenics in enriched germanium have been reported in previous works (Avignone et al. 1992; Barabanov et al. 2006; Elliott et al, 2010). Table 5 lists several different past approaches as well as this work.

Paper	Monte Carlo/ Nuclear cross section tool	Cosmic Neutron spectrum
(Elliott et al. 2010)	CEM03.02	4FP60R flux
(Avignone et al. 1992)	ISABEL simulated cross sections	(Hess et al. 1959)
(Barabanov et al. 2006)	SHIELD code	(Ziegler 1998)
This work (User selected n spectrum)	Geant4	(Kornmayer et al. 1995), (Ziegler 1998), (Hess et al. 1959), (Gordon et al. 2004)

Table 5: Cosmogenic production rate calculation of ^{68}Ge in the literature and the one used in this work.

The most modern attempt to experimentally determine cosmogenic activation in enriched germanium is reported in (Elliott et al. 2010). Elliot et al. irradiated a sample of enriched germanium in a neutron beam at the Los Alamos Neutron Science Center (LANSCE) Weapons Neutron Research (WNR) facility on the Target 4 Flight Path 60 Right (4FP60R). This was a dedicated experiment and the sample was gamma-ray counted in an underground lab. The work presented by Avignone et al. (1992) was an experimental study obtained using three natural isotopic abundance germanium detectors, and then the results were extrapolated to the enriched case. Avignone et al. has the caveat that some of the detectors that took the data were flown overseas. This affects the activation greatly. The authors did correct for this fact in their reported results. The third paper mentioned in Table 5 (Barabanov et al. 2006) reports the findings of a simulated model of the enriched germanium transport shield, in a manner similar to this work. The simulation code is different from the one used in this study, and they report an approximate attenuation factor of the production of isotopes inside the shield. This is the only reported work on shielding materials and shield design of interest for neutrinoless double-beta decay searches based in enriched germanium.

4.0 Germanium transport and different shielding scenarios

The enriched germanium detector material produced in Russia for the MAJORANA DEMONSTRATOR is transported via ship to the U.S. The transport container is a standard dry-freight container with the cosmogenic shield inside of it. The dimensions and allowable weight of this container will set the limits on weight and dimensions of this cosmogenic shield. Table 6 displays the dimensions and weight ratings for a standard dry-freight container. The next sub-section describes the GERDA shield. This shield is currently in use by the Majorana collaboration, and in the last section of this paper, the performance reported for the shield is compared to expectations from the simulation tool described in this paper. The last sub-section discusses the improved shield design resulting from this work.

Parameter	Value
Maximum Gross Weight	67,200 lbs. (30250 Kg.)
Tare Weight	4,850 lbs. (2182 Kg.)
Payload	62,350 lbs. (28058 Kg.)
Capacity	1,164 cu. ft. (33 m ³)
Internal Dimensions	
Length	19' 2" (5.85 m)
Width	7' 6" (2.31 m)
Height	7' 8" (2.33 m)
External Dimensions	
Length	19' 10" (6.05 m)
Width	8' (2.44 m)
Height	8' 6" (2.59 m)
Door Opening	
Width	7' 6" (2.29 m)
Height	7' 4" (2.24 m)

Table 6: Weight ratings and dimensions of a standard dry-freight container (Shipping Container Housing Group 2010).

4.1 GERDA shield

The GERDA cosmogenic shield was designed for use by the GERDA (Schoner et al. 2005) neutrinoless double-beta decay experiment. A model drawing of this shield is presented in Figure 6. The shield is made entirely of steel. Table 7 contains selected parameters that describe the GERDA shield. The shield dimensions and weight presented in this section were taken from (Barabanov, et al. 2006).

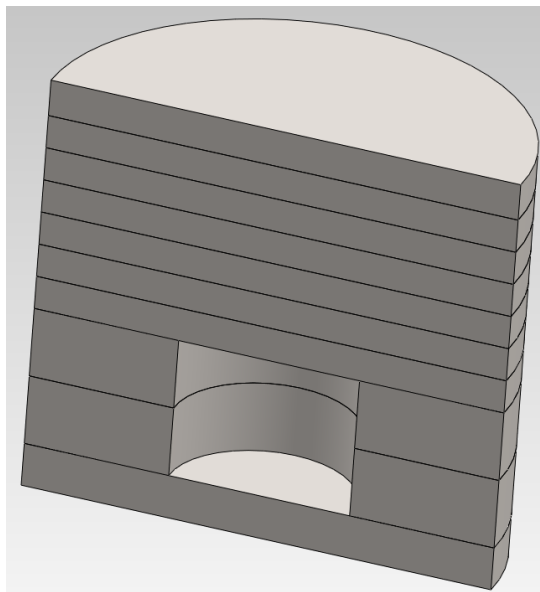


Figure 6: GERDA Shield

Cylindrical iron block with cavity	
Total height	126.5 cm
Weight	14500 Kg.
Diameter	140 cm
Bottom thickness	15 cm
Cavity height	40 cm
Cavity diameter	54 cm
Enriched germanium capacity	
Diameter	42 cm
Height	27 cm

Table 7: Selected parameters describing the GERDA shield

4.2 Improved cosmic neutron shield

There is a difference of 17,250 kg (35,000 lb) between the GERDA shield and the dry-freight container allowable capacity, leaving room for more shielding. The weight of the proposed enhanced shield was calculated to match the maximum capacity of a dry-freight container. The choice of material is based on the shielding properties of different materials at the specific energy range of interest (20 MeV – 10 GeV).

From the list of materials studied in (Aguayo-Navarrete et al. 2011) (concrete, iron, lead, water, polyethylene, borated polyethylene), iron is the best choice of materials to fill the transport container to capacity for the application at hand, as presented in the previous section. Optimizing the shape of the added iron should take into account the angular distribution of the cosmic neutron shower. The angular distribution follows a $(\cos\Theta)^{3.5}$ function (Ziegler 1998), and this angular dependence is implemented as part of the input library of the simulation tool. Figures 8 show the conceptual design of the improved shield. The optimization leads to adding additional shielding only on top of the existing GERDA shield, with a diameter somewhat larger than the existing shield to cover more solid angle from above.

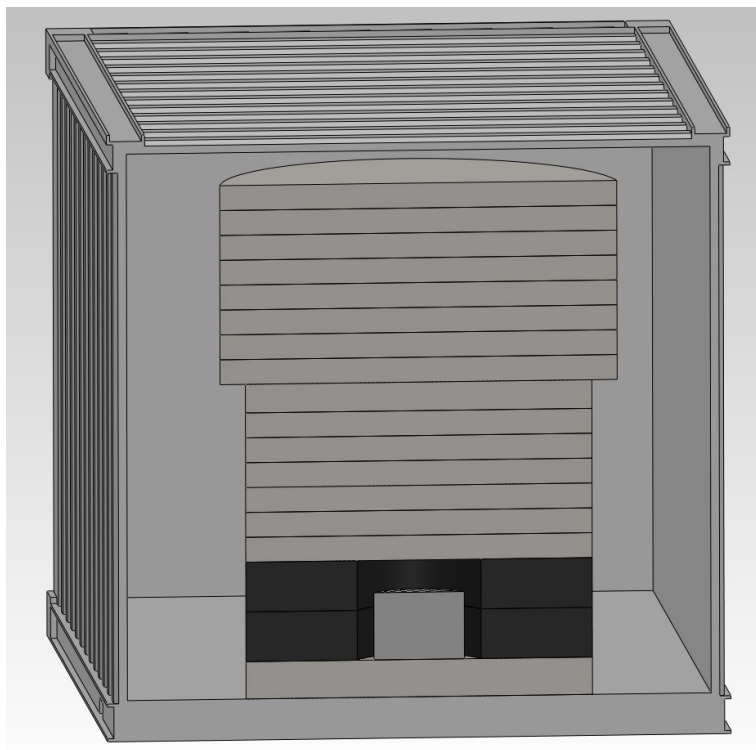


Figure 7: Cross sectional view of the improved transport shield in the dry freight container

5.0 Production rates

This section presents the results obtained using the Monte Carlo application modeling of sea-level neutrons from cosmic rays as source particles, as they would impact a sample of enriched germanium sitting at sea-level unshielded (Section 5.1) and shielded (Section 5.2). The unshielded scenario allows for a comparison with other studies while the shielded scenario provides insight in to the amount of activation that can be anticipated in GERDA and the MAJORANA DEMONSTRATOR.

5.1 Production rates from sea-level cosmic neutrons (unshielded scenario)

Table 8 presents the simulated cosmogenic production rates in enriched germanium and experimental results reported in the literature. The latest version of Geant4, 4.9.5, was reworked to more accurately model backgrounds in underground environments. As shown in the results of this work, the modifications to previous versions have made the toolkit produce results closer to those reported in the literature in the cosmogenic production arena. The isotope production rates presented in Table 8 (R_{ge68} , in atoms per kilo per day) were computed using Equation 2.

$$R_{\text{ge68}} = \left(\frac{In_{\text{ge68}}}{Tot} \right) \cdot \left(\frac{A_{\text{ge}} \cdot \Phi_{\text{ge}}}{M_{\text{ge}}} \right), \quad (2)$$

where

In , number of isotopes of interest produced in the simulation.

Tot , total number of particles simulated.

Φ_n integrated cosmic neutron intensity above 10 MeV at sea-level, in $\text{n}/\text{cm}^2 \text{ s}$.

A_{ge} is the cross sectional area of the top surface of the germanium sample, in cm^2 .

M_{ge} is the total mass of germanium simulated in kg.

	Physics list	Neutron reaction cross section model	Cosmic neutron spectrum	^{68}Ge (atoms/day*kg)	^{60}Co (atoms/day*kg)
Geant4.9.3	QGSP_BERT	G4NDL 3.12	CRY	56.9 ± 0.4	1.02 ± 0.09
Geant4.9.3	QGSP_BERT	G4NDL 3.12	Hess	4.9 ± 0.2	0.018 ± 0.008
Geant4.9.3	QGSP_BERT	G4NDL 3.12	Ziegler	24.5 ± 0.9	0.34 ± 0.11
Geant4.9.5	QGSP_BERT	G4NDL 4.0	Ziegler	6.38 ± 0.23	0.44 ± 0.14
Geant4.9.5	QGSP_BERT	G4NDL 4.0	Gordon	3.42 ± 0.21	0.18 ± 0.05
Geant4.9.5	Shielding	G4NDL 4.0	Ziegler	9.49 ± 0.28	0.67 ± 0.07
Avignone et al.	-	ISABEL	Hess	0.94	-
Barabanov et al.	-	ISABEL	Ziegler	4.3 ± 0.2	3.3 ± 0.1
Elliott et al.	-	CEM03	(Exp.)	2.1 ± 0.4	2.5 ± 1.2

Table 8: Summary of production rates with different sea-level cosmic neutron flux. Results from this study are presented in the first six rows.

From the results presented in this section, it is clear that the simulation tool used in this work does not reproduce experimental measurements in terms of production rate for each individual isotope, ^{60}Co to ^{68}Ge . In a recent published work (Mei et al. 2009), the authors noted that much of the large variation in the calculated rates was due to the use of different cosmic ray neutron estimates, in agreement with the findings of this work. The results from the most recent version of Geant4 provide better agreement with the experimental measurement of the ratios of ^{60}Co and ^{68}Ge . In particular, the combination of QGSP_BERT-HP physics list and the Ziegler neutron parameterization looks like the most “correct” as of the latest version of Geant4.

5.2 Shielded scenario

The shielded scenario adds another ingredient to the computation of the cosmogenic production rate, since the attenuation of the primaries and the generation of secondaries in the shield material plays a role in the final production rate. In this study, neutrons are the initial particle. The modeling of the neutron interaction with matter is still in development in the toolkit used in the simulations, and it is expected the future releases of the simulation toolkit will address this issue. Table 9 shows the results from the simulations of the attenuation factors for different for the GERDA shield and the enhanced shield compared to the unshielded geometry. The first row in Table 9 is the attenuation factor reported by the GERDA collaboration. The rest of the rows are calculations derived from simulations performed for this report. The change in the nuclear data file (from G4NDL 3.12 to G4NDL 4.0) had a dramatic effect in these results. With the older nuclear data library, the simulation code over estimated the attenuation factor compared with the GERDA result. With the new nuclear data library the attenuation factors are within an order of magnitude from the GERDA result. The large variations in the simulated results suggest the challenges of determining attenuation factors for cosmic neutrons using Monte Carlo methods. This issue can be addressed by an energy-weighted experimental measurement of the interaction of neutrons in different shielding materials. The cosmogenic production for the most recent version of the tool (Geant4.9.5) is 25 in the GERDA shield scenario and 770 in the enhanced shield scenario. As a result, the enhanced shielding provides a factor of 30 more attenuation. The results presented in rows 2 and 3 for the enhanced shield are limits, since the simulation runs did not yield enough statistics to report a attenuation ratio. The result presented in row 4 is highlighted, as this result is consider it to be most accurate.

Simulation tool	Physics list	Neutron reaction cross section model	Number of ^{68}Ge Unshielded / GERDA Shielded (Overburden of 86 cm of Fe)	Number of ^{68}Ge Unshielded / Enhanced shield (Overburden of 166 cm of Fe)
Barabanov et al.	-	SHIELD	10	-
Geant4.9.3 + CRY	QGSP_BERT	G4NDL 3.12	190	2.5e4
Geant4.9.3 + Hess	QGSP_BERT	G4NDL 3.12	280	>4.2e4
Geant4.9.5 + Gordon	QGSP_BERT	G4NDL 4.0	310	>4.2e4
Geant4.9.5 + Ziegler	QGSP_BERT	G4NDL 4.0	25	770
Geant4.9.5 + Ziegler	Shielding	G4NDL 4.0	80	1100

Table 9: Summary of the relative production rates of ^{68}Ge using the GERDA shield geometry and the enhanced shield geometry presented in this report compared to the unshielded geometry.

6.0 Conclusions

A transport shield design is proposed to reduce cosmogenic activation of enriched germanium for the MAJORANA DEMONSTRATOR. Based on a previous simulation study we find that for the limited weight and volume of the dry-freight overseas transport. The Q-values of cosmogenic production reactions for the isotopes of interest to neutrinoless double-beta decay searches in enriched germanium were calculated and cross-checked with the simulation tool, yielding a satisfactory result. The GERDA transport shield and an enhanced shield adding material within the size and weight limits of a dry-freight container have been evaluated in terms of reduction in cosmogenic production rates in isotopically (^{76}Ge) enriched germanium. The most accurate model used in this work estimates a reduction factor for the GERDA shield that is different from previous estimates by a factor of 2.5. The enhanced shield proposed in this work achieves an additional reduction factor of 30 in the attenuation of the cosmogenic production rate of ^{68}Ge in enriched germanium over the GERDA shield. These results are preliminary as the Geant4 application has not been benchmarked against an experimental result. The significant variation in the results depending on the specifics of the model point strongly suggest that an experimental weighted in energy to evaluate neutron flux attenuation in different shielding materials will yield more accurate estimates of the production of dangerous isotopes for neutrinoless double-beta decay searches in enriched germanium.

7.0 References

- Agostinelli, S, J Allison, K Amako, J Apostolakis, H Araujo, P Arce, M Asai, D Axen, S Banerjee, G Barrand, F Behner, L Bellagamba, J Boudreau, L Broglia, A Brunengo, H Burkhardt, S Chauvie, J Chuma, R Chytrcek, G Cooperman, G Cosmo, P Degtyarenko, A Dell'Acqua, G Depaola, D Dietrich, R Enami, A Feliciello, C Ferguson, H Fesefeldt, G Folger, F Foppiano, A Forti, S Garelli, S Giani, R Giannitrapani, D Gibin, JJG Cadenas, I Gonzalez, GG Abril, G Greeniaus, W Greiner, V Grichine, A Grossheim, S Guatelli, P Gumplinger, R Hamatsu, K Hashimoto, H Hasui, A Heikkinen, A Howard, V Ivanchenko, A Johnson, FW Jones, J Kallenbach, N Kanaya, M Kawabata, Y Kawabata, M Kawaguti, S Kelner, P Kent, A Kimura, T Kodama, R Kokoulin, M Kossov, H Kurashige, E Lamanna, T Lampen, V Lara, V Lefebure, F Lei, M Liendl, W Lockman, F Longo, S Magni, M Maire, E Medernach, K Minamimoto, PM de Freitas, Y Morita, K Murakami, M Nagamatu, R Nartallo, P Nieminen, T Nishimura, K Ohtsubo, M Okamura, S O'Neale, Y Oohata, K Paech, J Perl, A Pfeiffer, MG Pia, F Ranjard, A Rybin, S Sadilov, E Di Salvo, G Santin, T Sasaki, N Savvas, Y Sawada, S Scherer, S Seil, V Sirotenko, D Smith, N Starkov, H Stoecker, J Sulkimo, M Takahata, S Tanaka, E Tcherniaev, ES Tehrani, M Tropeano, P Truscott, H Uno, L Urban, P Urban, M Verderi, A Walkden, W Wander, H Weber, JP Wellisch, T Wenaus, DC Williams, D Wright, T Yamada, H Yoshida, and D Zschesche. 2003. "Geant4-a Simulation Toolkit." *Nuclear Instruments & Methods in Physics Research Section a-Accelerators Spectrometers Detectors and Associated Equipment* 506(3):250-303. 10.1016/s0168-9002(03)01368-8.
- Aguayo-Navarrete, E, AS Ankney, TJ Berguson, RT Kouzes, JL Orrell, and MD Troy. 2011. *Cosmic Ray Interactions in Shielding Materials*. Report No. PNNL-20693, Pacific Northwest National Laboratory, Richland, Washington.
- Aguayo-Navarrete, E, RT Kouzes, and JL Orrell. 2010. *Monte Carlo Simulations of Cosmic Ray Hadronic Interactions*. Report No. PNNL-20401, Pacific Northwest National Laboratory, Richland, Washington.
- Avignone, FT, RL Brodzinski, JI Collar, HS Miley, E Garcia, A Morales, J Morales, R Nunezlagos, JH Reeves, C Saenz, and JA Villar. 1992. "Theoretical and Experimental Investigation of Cosmogenic Radioisotope Production in Germanium." *Nuclear Physics B*:280-85.
- Barabanov, I, S Belogurov, L Bezrukov, A Denisov, V Kornoukhov, and N Sobolevsky. 2006. "Cosmogenic Activation of Germanium and Its Reduction for Low Background Experiments." *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 251(1):115-20.
- Blann M, *Phys. Rev.*, **C54**, 1341 (1996).
- Cebrian S, Gomez H, Luzon G, Morales J, Tomas A and Villar JA. 2010. "Cosmogenic activation in germanium and copper for rare events searches" *Astroparticle physics* 33, 316-329
- CSEWG Cross Section Evaluation Working Group. Accessed on April 12, 2012 at <http://www.nndc.bnl.gov/csewg/> (last updated date unknown).

- Elliott, SR, VE Guiseppe, BH LaRoque, RA Johnson, and SG Mashnik. 2010. "Fast-Neutron Activation of Long-Lived Isotopes in Enriched Ge." *Physical Review C* 82(5). 10.1103/PhysRevC.82.054610.
- Elliott, SR, and P Vogel. 2002. "Double Beta Decay." *Annual Review of Nuclear and Particle Science* 52:115-51. 10.1146/annurev.nucl.52.050102.090641.
- Gordon MS, Goldhagen P, Rodbell KP, Zabel TH, Tang HHK, Clem JM and Bailey P. 2004. "Measurement of the Flux and Energy Spectrum of Cosmic-Ray Induced Neutrons on the Ground." *IEEE Transactions on Nuclear Science*, 51, 6.
- Hagmann, C, D Lange, and D Wright. 2008. "*Cosmic-Ray Shower Library (CRY)*." Technical Manual, 5 pages. Lawrence Livermore National Laboratory, Livermore, CA. <http://nuclear.llnl.gov/simulation/cry.pdf>.
- Hess, WN, HW Patterson, R Wallace, and EL Chupp. 1959. "Cosmic-Ray Neutron Energy Spectrum." *Physical Review* 116(2):445-57. 10.1103/PhysRev.116.445.
- JENDL Nuclear Data Center at Japan Atomic Energy Agency. Accessed on April 12, 2012 at <http://www.ndc.jaea.go.jp/> (last updated date unknown).
- Kornmayer, H, HH Mielke, J Engler, and J Knapp. 1995. "High-Energy Cosmic-Ray Neutrons at Sea-Level." *Journal of Physics G-Nuclear and Particle Physics* 21(3):439-49. 10.1088/0954-3899/21/3/018.
- Kouzes, RT, M Engelhard, and Z Zhu. 2011. *Germanium-76 Sample Analysis Rev 2*. Report No. PNNL-20368, Pacific Northwest National Laboratory, Richland, Washington.
- SG Mashnik et al., Proc. Int. Conf. on Nuclear Data for Science and Technology (ND2001) LA-UR-01-5391.
- Mei, DM, ZB Yin, and SR Elliott. 2009. "Cosmogenic Production as a Background in Searching for Rare Physics Processes." *Astroparticle Physics* 31(6):417-20. 10.1016/j.astropartphys.2009.04.004.
- S. Schonert, I. Abt, M. Altmann, A.M. Bakalyarov, I. Barabanov, C. Bauer, M. Bauer, E. Bellotti, S. Belogurov, S.T. Belyaev, A. Bettini, I. Bezrukov, V. Brudanin, C. Buttner, V.P. Bolotsky, A. Caldwell, C. Cattadori, M.V. Chirchenko, O. Chkvorets, H. Clement, E. Demidova, A. Di Vacri, J. Eberth, V. Egorov, E. Farnea, A. Gangapshev, G.Y. Grigoriev, V. Gurentsov, K. Gusev, W. Hampel, G. Heusser, W. Hofmann, L.V. Inzhechik, J. Jochum, M. Junker, S. Katulina, J. Kiko, I.V. Kirpichnikov, A. Klimenko, K.T. Knopfle, O. Kochetov, V.N. Kornoukhov, R. Kotthaus, V. Kusminov, M. Laubenstein, V.I. Lebedev, X. Liu, H.G. Moser, I. Nemchenok, L. Pandola, P. Peiffer, R.H. Richter, K. Rottler, C.R. Alvarez, V. Sandukovsky, S. Schonert, S. Scholl, J. Schreiner, B. Schwingenheuer, H. Simgen, A. Smolnikov, A. Tikhomirov, C. Tomei, C.A. Ur, A.A. Vasenko, S. Vasiliev, D. Weisshaar, M. Wojcik, E. Yanovich, J. Yurkowski, S.V. Zhukov, and G. Zuzel,
The GERmanium DETECTOR ARRAY (GERDA) for the search of neutrinoless $\beta\beta$ decays of

Ge-76 at LNGS,
In *Nucl. Phys. B*, volume 145, page 242, 2005.
(DOI: <http://dx.doi.org/10.1016/j.nuclphysbps.2005.04.014>)

Shipping Container Housing Group. 2010. *Shipping Container Standard Dimensions*. Accessed on July 22, 2011 at <http://www.shipping-container-housing.com/shipping-container-standard-dimensions.html> (last updated date unknown).

The Geant4 Collaboration. *Physics Lists - Use Cases*. Accessed on July 22, 2011 at <http://geant4.org/geant4/support/physicsLists/referencePL/useCases.shtml> (last updated July 9, 2011).

Y. Yariv and Z. Fraenkel. *Phys. Rev. C* 24 (1981) 488.

Yu NS, Lunev VP, Konobeyev AY, Ditjuk AI "Cross section data library MENDL-2 to study activation as transmutation of materials irradiated by nucleons of intermediate energies", report INDC(CCP)-385 (International Atomic Energy Agency, May 1995)

Ziegler, JF. 1998. "Terrestrial Cosmic Ray Intensities." *IBM Journal of Research and Development* 42(1):117.



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