JRC Scientific and Technical Reports

On the Underground Production of High Purity Germanium Detectors

Mikael Hult[1], Sergey Belogurov[2], Allan Caldwell[3], Josef Janicsko[3], Vasiliy Kornoukhov[2,4], Stefan Schönert[5]

[1] European Commission-Joint Research Centre-Institute for Reference Materials and Measurements (EC-JRC-IRMM), Geel, Belgium
[2] Institute for Nuclear Research of the Russian Academy of Sciences, Moscow, Russia
[3] Max Planck Institut für Physik, München, Germany
[4] Institute of Theoretical and Experimental Physics, Moscow, Russia
[5] Max Planck Institut für Kernphysik, Heidelberg, Germany

EUR 23237 EN - 2008

JRC EUROPEAN COMMISSION

Institute for Reference Materials and Measurements

GERDA

The mission of the IRMM is to promote a common and reliable European measurement system in support of EU policies.

European Commission
Joint Research Centre
Institute for Reference Materials and Measurements

Contact information
Mikael Hult
Institute for Reference Materials and Measurements
Retieseweg 111
B-2440 Geel, Belgium
E-mail: Mikael.hult@ec.europa.eu
Tel.: +32-(0)14-571 269
Fax: +32-(0)14-571 864

http://irmm.jrc.ec.europa.eu/
http://www.jrc.ec.europa.eu/

JRC 43108

EUR 23237 EN
ISBN 978-92-79-08276-4
ISSN 1018-5593
DOI 10.2787/13569

Luxembourg: Office for Official Publications of the European Communities

Executive Summary

Detectors based on high purity germanium (HPGe) are used in numerous experiments world-wide aiming at detecting rare events like double beta decay and interactions of dark matter (see e.g.: Schönert and the GERDA Collaboration, 2005; Aalseth and the Majorana Collaboration, 2005; Chapellier and the Edelweiss collaboration, 2003). Furthermore, HPGe-detectors for gamma-ray spectrometry that are installed underground have become laboratory workhorses and are frequently employed to check materials that go into constructing large scale detectors for "big science" (the Borexino collaboration, 2002). Such HPGe-detectors have in recent years been used in increasing numbers of other applications ranging from environmental radioactivity measurements to applications in plasma physics and dosimetry as described in some recent reviews: (Hult, 2007; Laubenstein et al., 2003; Hult et al., 2003; Hult et al., 2006).

Work on identifying contributions to the background have been successful (see e.g. Heusser, 1996) and the relative background contribution from cosmogenically produced radionuclides in germanium is increasing every year. In order to reach the goals of the most recent attempts to measure the double beta decay of ^{76}Ge as well as constructing state-of-the-art HPGe-detectors for use in deep underground laboratories, it is necessary to have access to germanium that is free of cosmogenic radionuclides. The two most prominent cosmogenic radionuclides in germanium are ^{68}Ge and ^{60}Co, with half-lives of 0.8 y and 5.2 y, respectively.

Triggered by the Joint Research Action 1 (Low Background Techniques for Deep Underground Science Laboratories, LBT-DUSL) within ILIAS, this report summarises the advantages of having access to germanium produced underground. It also summarises the technical installations necessary for such an arrangement. The report also describes the second best approach to minimising production of ^{68}Ge and ^{60}Co and other cosmogenic radionuclides, which is to store the germanium underground and bring it above ground when it is processed. A more imminent need was discussed within the ILIAS Joint Research Activity 2 (Integrated Double Beta Decay – IDEA) where solutions for the GERDA double beta decay experiment were discussed. By optimising the logistics it was possible to reach a germanium quality that was acceptable for the ongoing GERDA experiment's first phase.

The report finishes with some cost estimates for installing a facility for underground production of germanium and compares the cost with the present solution of optimising the logistics.

Table of contents

Forewords - ILIAS

ILIAS (Integrated Large Infrastructure for Astroparticle Science) is a consortium that was established through a contract between the European Commission (EC) and 21 participating institutes. It was launched in April 2004 for a duration of 5 years, with a total EC financial contribution of € 7.5 Million. The ILIAS website (ILIAS, 2007) presents the ILIAS mission as follows:

"ILIAS has pulled together a major part of Europe's leading infrastructures in Astroparticle Physics, namely Underground laboratories and Gravitational Waves observatories, to produce a focused, coherent and integrated project. Its goals are multiple:
• organise and structure the European Astroparticle Physics community,
• improve the existing infrastructures and their operation,
• prepare the best infrastructures for the future."

ILIAS is divided into Joint Research Actions (JRAs), networks and transnational access. This report presents work carried out in within JRA1 and JRA2 with the aim of investigating possibilities for underground production of germanium for High Purity Germanium detectors (HPGe-detectors). This is in line with the last line of the ILIAS mission to "prepare the best infrastructures for the future".

JRA1 is named Low-Background Techniques for Deep Underground Science Laboratories (LBT-DUSL). It aims at structuring the activity of the Deep Underground Science Laboratories in their common scientific and technical objectives. It undertakes the improvement and implementation of innovative ultra-low background techniques in four of the European underground laboratories. Several teams that are operating experiments in the underground laboratories are contributing to the JRA. This report is a deliverable within JRA1 as it could potentially be possible to install equipment for HPGe-detector production underground.

JRA2 is named Integrated Double Beta Decay (IDEA) and it aims to further develop and integrate the most promising techniques in the field of underground double beta decay (DBD) experiments culminating in the foundation of a European Observatory for DBD (EODBD). Within the EODBD one should be capable of studying this elusive nuclear process with different isotopes and complementary technical approaches, and to cross-check stringent limits. The ultimate goal is the measurement of a finite value of the Majorana neutrino mass. The role of this Observatory will extend well beyond the 5 year time assigned to ILIAS-IDEA: it aims at being the world reference infrastructure for the study of DBD and for the determination of the neutrino mass scale. The GERDA experiment is one of the most promising experiments to detect the double beta decay. GERDA aims at studying the neutrinoless double beta decay in ^{76}Ge by using an array of high purity germanium crystals immersed in liquid argon. For the GERDA experiment it is absolutely vital that the germanium detectors are not exposed to cosmic rays or at least that the exposure is kept to a minimum. For this reason this report is a deliverable also in IDEA. It was clear that it was possible to merge the two deliverables since so many things were common for this study within JRA1 and JRA2. Within JRA1 it was necessary to look in a wider perspective to underground production while the JRA2 approach was to solve the more urgent needs of GERDA. These aims coincided in many respects.

1. Introduction

1.1. Germanium

The website Caryacademy (2007) gives a good brief description of germanium:
" Germanium was discovered by Clemens A. Winkler in 1886 in Germany. It was discovered during the isolation of a mineral called agyrodite. Because it was discovered in Germany, the name comes from the Latin root Germania, meaning Germany. Germanium's existence was first predicted by Mendeleev in 1871, due to the gaps in his newly created Periodic Table. He originally called it ekasilicon due to its predicted similarities in properties to silicon, and due to its predicted spot in the Silicon group of the Periodic Table."
"Germanium is obtained by the melting of zinc "ores". The Germanium is the by-product of the melting. Germanium may also be produced by burning various types of coal, and again it would be found as a by-product. However, almost a quarter of the Germanium that is used comes from its extraction from recycled metals" (Caryacademy, 2007)

Figure 1. Clemens Alexander Winkler, who discovered Ge (1838-1904) (From Seilnacht, 2007)

Natural germanium is composed of 5 isotopes with relative natural abundances as given in Table 1, which also gives the half-life of germanium radionuclides with half-life longer than 1 minute. It is clear from Table 1 that for underground science with germanium detectors, it is mainly ^{68}Ge that may be a cause of concern. The other Ge radionuclides will decay shortly after the detector or sample is taken underground.

In the early days of semiconductor developments in the 1940ies, germanium was actually the material of choice particularly for bipolar devices. The less advantageous properties of the germanium oxide (GeO_2), which provided insufficient passivation, hampered its use in field-effect devices. The introduction of the planar process in the 1960ies (Hoerni, 1960) gave the Si-based technology an edge over the Ge-based technology. Today, germanium is used in semiconductor industry albeit not to the same extent as silicon (Claeys and Simoen, 2007).

Table 1. Natural Isotopic abundances of the 5 "stable" isotopes of germanium as well as half-life of the radioactive ones, without mentioning the few short-lived (< 1 min) metastable states.

Mass number	Natural Isotopic abundance (%)	Half-life	Reference
65		30.9(5) s	WWW ToI, 2007
66		2.26(5) h	WWW ToI, 2007
67		28.9(3) min	WWW ToI, 2007
68		270.95(16) d	DDEP (2007)
69		39.05(10) h	WWW ToI, 2007
70	20.84(87)		NIST, 2007*
71		11.43(3) d	WWW ToI, 2007
72	27.54(34)		NIST, 2007*
73	7.73(5)		NIST, 2007*
74	36.28(73)		NIST, 2007*
75		82.78(4) min	WWW ToI, 2007
76	7.61(38)		NIST, 2007*
77		11.30(1) h	WWW ToI, 2007
78		88.0(10) min	WWW ToI, 2007

* Based on the publication by Rosman and Taylor (1997).

There are, however, prospects to the future use of Ge in ULSI (Ultra Large Scale Integration) devices and in nanoscale MOS (Metal Oxide Semiconductor) dielectrics and junctions. Other special features that makes Ge an interesting material is that it is permeable to IR-radiation (infra-red). The main applications today for bulk single crystalline germanium are:

- Lenses for IR optics
- Substrate for III-V based opto-electronic devices used for.
    - Solar cells
    - LEDs (Light Emitting Diodes)
    - HEMTs (High Electron Mobility Devices)
- Detectors for gamma-ray spectrometry

It is he latter point that is the focus of the study of this report. There are of course other uses of germanium that do not involve bulk single crystals. Examples and amount of such uses are given in reports of the US Geological Surveys (2007).

For further information on general properties of germanium there are now several sources of good quality information available on internet. The list below was accessed on December 5, 2007.

Augenbraun, Eliene. "Germanium Comes of Age". PBS.
http://www.pbs.org/transistor/science/info/germanium.html
Barbalace, Kenneth. "Element Germanium-Ge". EnvironmentalChemistry.com.
http://environmentalchemistry.com/yogi/periodic/Ge.html
Bentor, Yinon. "Germanium". ChemicalElement.com.
http://www.chemicalelements.com/elements/ge.html.
Gagnon, Steve. "Germanium". Jefferson Lab. http://education.jlab.org/itselemental/ele032.html
"Germanium". The Columbia Encyclopedia. http://www.bartleby.com/65/ge/germaniu.html
"Germanium". Wikipedia Encyclopedia. http://en.wikipedia.org/wiki/Germanium
"Germanium-Ge". LennTech. http://www.lenntech.com/Periodic-chart-elements/Ge-en.htm
Klimasauskas, Edward. "Germanium Statistics and Information". USGS.
http://minerals.usgs.gov/minerals/pubs/commodity/germanium/

Winter, Mark. "Germanium". WebElements.
http://www.webelements.com/webelements/elements/text/Ge/key.html

1.2. Development of HPGe-detectors

Ge-detectors have taken over the role as radioactivity laboratory workhorses from the NaI(Tl) detectors. The reason for this is the great developments in the production of perfect high purity single crystals, which is described in a recent review article (Hult, 2007). The first Ge-detectors for measuring ionising radiation was reported by Freck and Wakefield in 1962. Their detector was small with a sensitive volume of about 0.15 cm^3 and had poor resolution (3% at 662 keV). The resolution could be improved already within a year or two but it took many years before detectors of significant volume could be produced. In the 1960ies the lithium drifting was introduced. It enabled cancellation of p-type acceptor impurities in the entire crystal. In the early years of Ge-detector production this was necessary, since the intrinsic purity of Ge-crystals was not sufficiently high. It was not until 1971 that Hall and Soltys could improve the Czochralski pulling technique so that the production of high purity Ge-crystals in which they were able to reduce the concentration of electrically active impurities down to 10^{10} cm^{-3} could be achieved. It took another 10 years before the size of the HPGe-detectors (High Purity Germanium detectors) could be produced at the same size s lithium drifted detectors. Today it is possible to grow crystals of HPGe-detector quality that are 10 cm in diameter. The largest single-crystal HPGe-detectors in operation today weigh nearly 5 kg and are well-detectors 10 cm in diameter and 12 cm in length (Cazala et al., 2003; Gurriaran et al., 2004). The largest coaxial detector was slightly less massive and reported by Sangsingkeow *et al.* in 2003. It is tremendously difficult to produce such big HPGe-detectors and manufacturers may well need to carry out many attempts before succeeding in producing such large crystals with electrical properties that are acceptable. The strive for large detectors for carrying out low-level measurements is of course driven by the fact that one can achieve lower detection limits by use of detectors of high efficiency. Hult and Gasparro (2007) define a Figure of Merit (FoM), which can be used to study the improved performance of Ge-detectors over the past 45 years.

Eq. 1
$$FoM = \frac{\varepsilon(E)}{\sqrt{R(E)B(E)}}$$

In Equation 1, R is the resolution (i.e. FWHM in keV), B the background count rate per keV and ε the relative efficiency (here, for a point source located 25 cm above the centre of the detector), all measured at the energy E. Gasparro and Hult concludes that at 1332 keV the FoM (excluding coincidence techniques and multi-crystal arrangements) has doubled every 3 years since 1963. Note that since the larger the crystal is, the higher the background is, and efforts to reduce the background become more important the larger the crystal is. Great improvements in reducing background of Ge-detectors have been reported from several research groups. The main developments have been achieved by the groups at Max Planck Institut für Kernphysik in Heidelberg and the Pacific Northwest Laboratories in Washington, while performing experiments on the double beta decay of ^{76}Ge. A significant part of the background in these experiments comes from cosmogenic activation of the germanium crystals. The two main radionuclides are ^{60}Co and ^{68}Ge but this will be discussed further in a later section. Such activation will become even more important in further developments like in the two largest low-background Ge-detector systems under development today. They are the GERDA and the MAJORANA experiments. Both of these efforts aim at detecting the neutrinoless double-beta decay of ^{76}Ge by using arrays of large Ge-crystals. In order for these two experiments to be successful and reach their target goals, it is essential that the Ge-crystals be kept above ground as little as possible. In order to accomplish this one must either (i) arrange for very strict and well-planned logistics and a large amount of transports between a deep underground storage and a the Ge-producer as well as the Ge-detector manufacturer or (ii) develop underground production of Ge-crystals and/or Ge-detectors.

1.3. Cosmogenic radionuclides produced in germanium

There are numerous radionuclides produced in germanium from the interaction of cosmic rays or cosmic ray induced radiation. It is ultimately these radionuclides that determine the detection limits in deep underground germanium detection systems were all primordial and anthropogenic radioimpurities have been suppressed to an insignificant level[♣]. In deep underground laboratories, only radionuclides with long half-lives (say longer than one month) are of concern if one assumes cosmogenic activation in the underground laboratory to be negligible. Furthermore if one look at the specific case of the neutrinoless double beta decay in ^{76}Ge ($0\nu\beta\beta$), it is only long-lived radionuclides that may generate a pulse at 2038 keV (the decay energy of the $0\nu\beta\beta$ of ^{76}Ge) that are of concern. Such cosmogenic radionuclides are often called "dangerous" ones in the context of ^{76}Ge-double beta decay experiments. For "normal" radioactivity measurements and dark matter experiments, more or less all long-lived cosmogenic radionuclides are of concern. The most imminent need for the ILIAS-IDEA Joint Research Action was to come up with solutions for the ongoing double beta decay experiments and that is why this discussion will focus on the "dangerous" cosmogenic radionuclides. The most important longlived cosmogenic radionuclides in germanium are Ge-68, Co-60, Mn-54 and Zn-65. The first two are the most investigated ones because they may contribute into background of the experiments aiming at measuring the neutrinoless double beta decay in Ge-76 and are thus labelled "dangerous". Ge-68 has the highest production cross section and relatively short half life (9 months). Co-60 and other isotopes have lower production cross sections. The longest half-life amongst the considered isotopes is that of Co-60 (5.27 y). Thus, the activity of Ge-68 is determining the time before a deep underground Ge-detector can take up operation, while Co-60 gives dominating background contribution in the 10 year scale, or during the expected life time of the experiment. We will focus this section on the last two isotopes. Our consideration of Co-60 and Ge-68 is mainly based on work by Barabanov and co-workers (2006).

At the Earth's 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. Mn-, Zn, and Co-60 isotopes can be produced after a muon capture. However the probability of this channel should be much less than 10^{-4}, see e.g. (Wyttenbach et al. , 1978). The muon capture rate at the sea level is $\approx 10^{-6}$ $g^{-1}s^{-1}$ (Charalambus, 1971). So the isotope production rate should be much less than 10^{-2} kg^{-1} day^{-1}. The contribution of muon induced fast neutrons may be roughly estimated using the results of Cocconi (1951), that only about 2% of nuclear disintegrations by cosmic rays are due to muon induced neutrons. About 98% of cosmogenic activations are produced by nuclear active component (N-component) of cosmic rays and might be in principle attenuated by a reasonable shield. It is necessary to stress, that the muon induced contribution

[♣] When all background components including cosmogenic activation products have been "removed" it is of course the double beta decay of ^{76}Ge that determines the background.

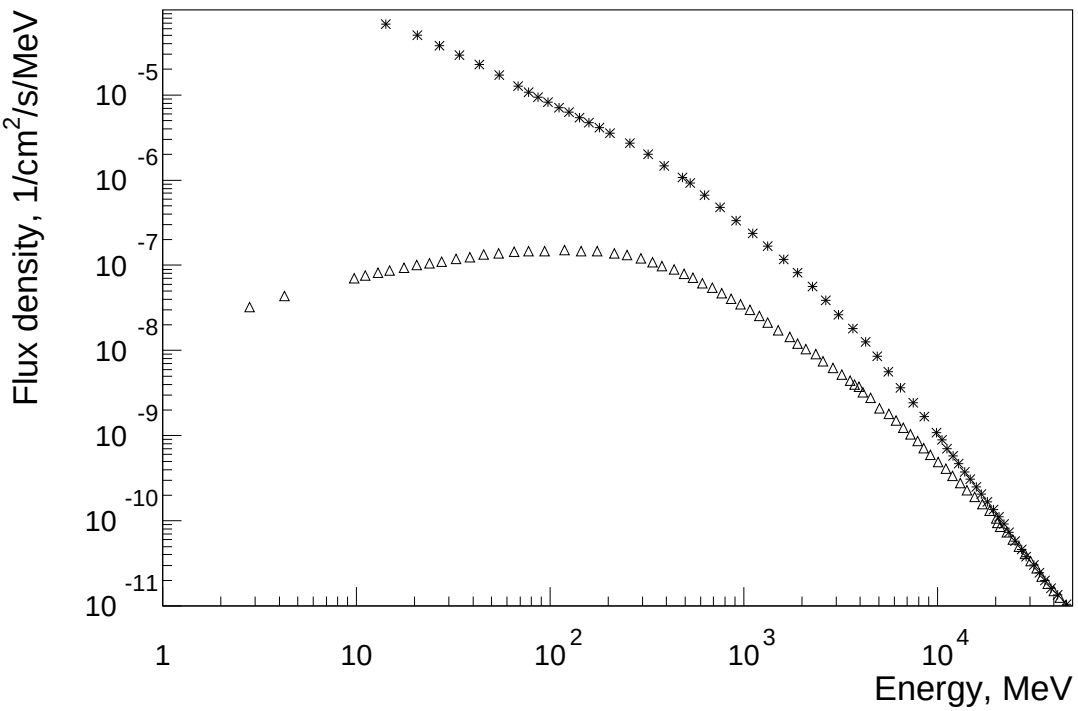
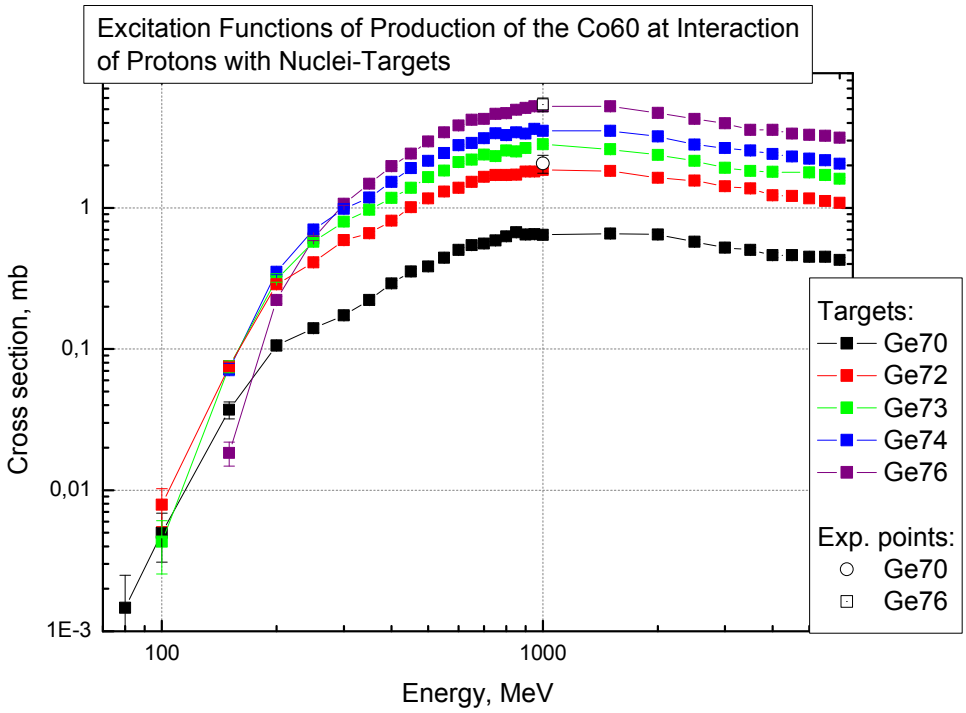


Figure 2. Neutron (asterisks) and proton (triangles) flux density at the sea level. This parameterisation was used by Ziegler and Sellschop (1981).

practically does not decrease in a shield of about 1000 g/cm^2 thickness. The composition of the N-component of cosmic rays at sea level is the following: more than 95% neutrons, about 3% protons and about 2% π-mesons (Hayakawa, 1969; Ziegler, 1996).

In the cited work we used the energy spectra and fluxes of neutrons and protons from (Ziegler and Sellschop, 1981), Figure 2. The angular distribution was supposed to be proportional to $\cos^{3.5}\theta$, where θ is zenith angle. Analyses of uncertainties of the flux and spectral parameterisation may be found in a report by Ziegler (1996).

Another important entry point for simulating the cosmogenic background is the knowledge of partial cross sections for the production of Co-60 and Ge-68 by spallation reactions.



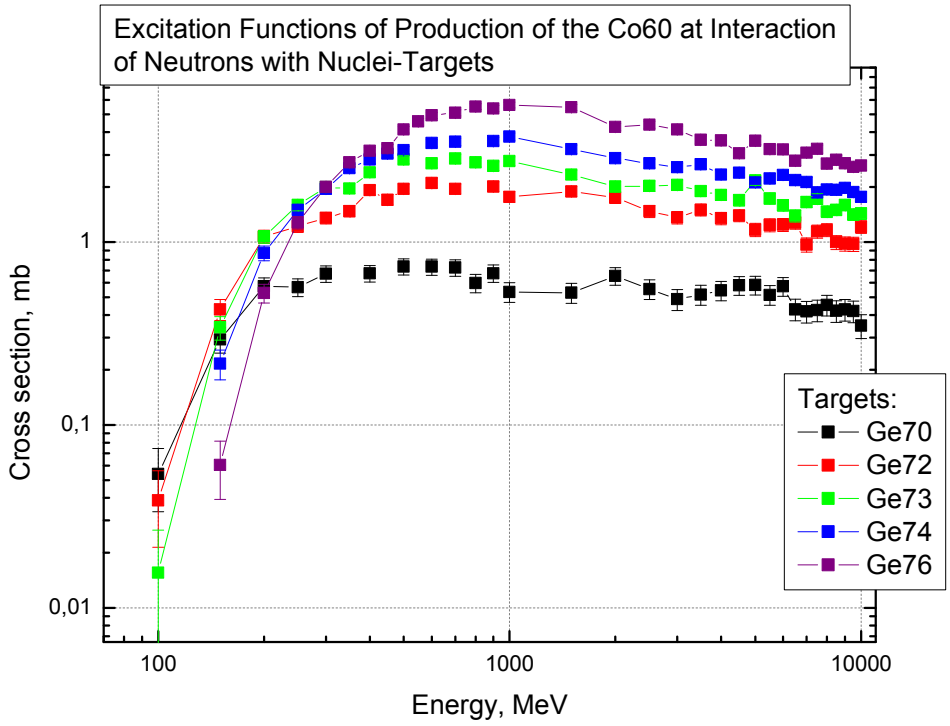
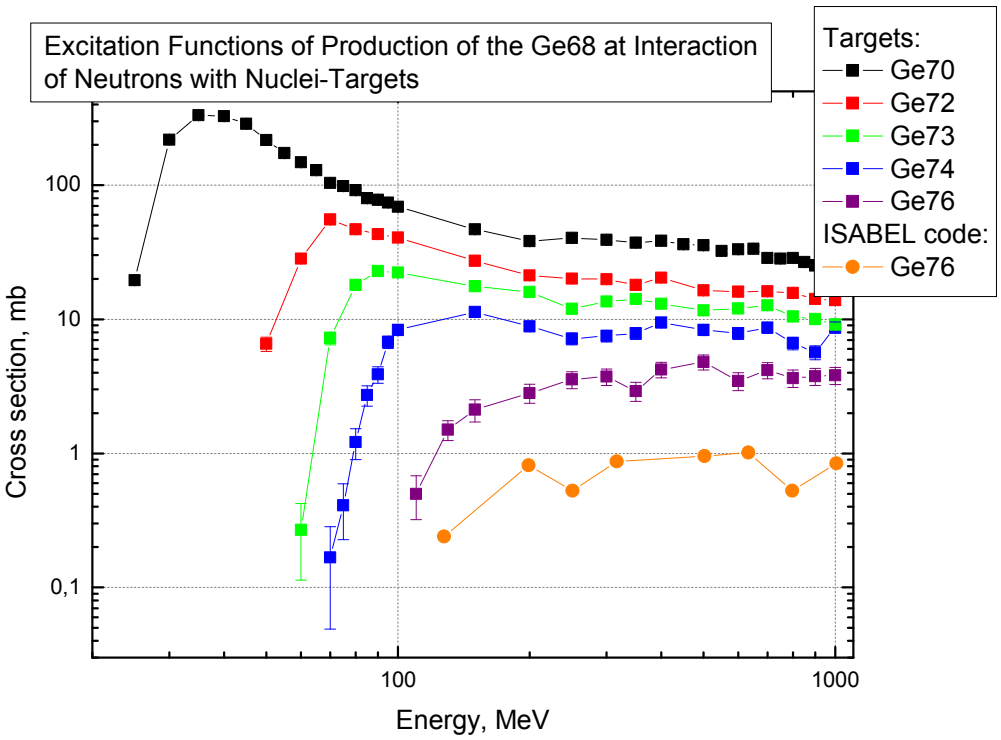


Figure 3a-b. Excitation functions for cosmogenic isotopes production from Barabanov and Belogurov (2006).



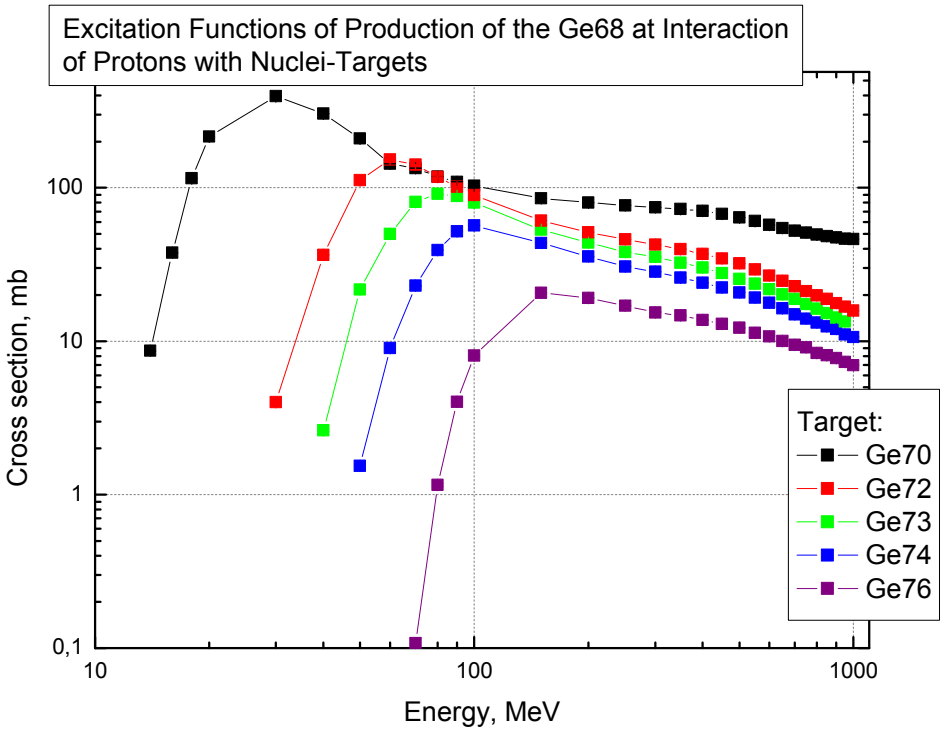


Figure 4a-b. Excitation functions for cosmogenic isotopes production from Barabanov et al. (2006).

There exist several physical models describing the spallation process (Sobolevsky, 2004) and a few experimental results for interesting nuclides. Some of the models, together with compilation of experimental data, are implemented in the nucleon transport codes like LAHET, SHIELD, FLUKA etc. A general analysis of the most diffused codes is done by Sobolevsky (2004), see also Cebrian (2004).

A recent version of Geant4 (9.0) was also tested for isotope production and the result was found to be within the uncertainty of the existing data and other simulations. This will allow us in the future to simulate the activation rate within the framework of our general Monte-Carlo software.

Excitation functions for the production of Co-60 and Ge-68 by neutrons and protons on the stable isotopes of germanium were generated with the SHIELD code. Results for protons coincide within a factor of between 1.5 and 2 with the experimental data from Horuguchi et al. (1983) and Batist et al. (1982). The curves for excitation functions are shown in the Figures 3 and 4.

Besides the excitation functions, results for production rates of cosmogenic isotopes may be compared between various codes and with experiments. In the Table 2 production rates of Ge-68 and Co-60 at sea level on all the stable isotopes of Germanium are shown. Combining these data according to isotopic composition of the natural germanium detector we get our prediction for the production rate of the radioactive nuclides by nuclear active component of cosmic rays at the Earth's surface. It is 81 Ge-68 atom/d/kg and 2.9 Co-60 atom/d/kg for natural germanium. For the material enriched to 87% in the Ge-76 the production rates are 5.6 Ge-68 atom/d/kg and 3.3 Co-60 atom/d/kg

Previous estimates of cosmogenic activation of germanium detectors were done using excitation functions calculated with ISABEL code and supported by special measurement with natural germanium (Avignone, 1992). Those results were between 2 and 6 times lower than ours for Ge-68 and about 2 times higher for Co-60. However such discrepancies between different measurements and different simulation methods are typical for this kind of research, see e.g. Cebrian (2004).

Our results for Ge-68 production rate on natural germanium agree quite well with the latest estimate of Cebrian (2005), while results for Co-60 probably should be doubled for a conservative estimate (see Table 3)

Table 2 Numbers necessary for computation of cosmogenic activity

r/a product (j)	half life, ($T_{1/2j}$, y)	target (i)	α_{ij}, 1/d/kg
Ge-68	0.74	Ge-70	281.4
		Ge-72	55.34
		Ge-73	28.0
		Ge-74	14.53
		Ge-76	4.22
Co-60	5.27	Ge-70	1.73
		Ge-72	2.88
		Ge-73	3.14
		Ge-74	3.35
		Ge-76	3.31

Table 3 Production rates of cosmogenic isotopes at sea level in natural Ge, $kg^{-1}d^{-1}$ following Barabanov et al. (2006) and Cebrian (2005).

	HMS-ALICE +YIELDX	GENIUS	Miley'92	Avignone'92 (MC)	Avignone'92 (exp)	SHIELD
Ge-68	89	58.4	26.5	29.6	30±7	81
Co-60	4.8	6.6	4.8			2.9
Zn-65	77	79	30	34.4	38±6	
Mn-54	7.2	9.1		2.7	3.3±0.8	

A couple of remarks should be made about the backgrounds related to cosmogenic activations: the clock for Co-60 activity is started only after the crystal growth, while for the Ge-68 it is started after enrichment procedure. On the other hand activity of Ge-68 decreases rather fast due to relatively short half life.

Now let us discuss what can be done in order to reduce the cosmogenic activity. Generally two kind of efforts may be performed: (i) decrease of the duration of unshielded exposure of the germanium during detector manufacturing, i.e. optimization of logistic, and (ii) construction of shielding above all (or the most of) the equipment used for the crystal growth and the detector manufacturing and for the transport. One should keep in mind that about 2% contribution to the see level activation are due to muon induced hadron cascades and can not be attenuated with shields of the order of 10 m w.e.. For this reason any feasible shielding above the technological equipment or for transportation will always have the efficiency between factors 10 and 30. Further reduction may be achieved only deeper underground

The following formulae may be used for a determination of the cosmogenic activity of a detector produced from arbitrary isotope mixture and with special precautions against activations. Let $\alpha_{i\,j}$ be production rate of j-th radioactive product (j=Ge-68, Co-60) from i-th Germanium stable isotope (i=70, 72, 73, 74, 76) at sea level. $T_{1/2j}$ is half life of j-th nuclide and P_j is probability to have a count in ROI when nucleus of j-th type decays inside the detector. In this case the background rate in the ROI due to cosmogenic isotopes will be:

Eq. 2
$$R=\sum_{i,j}\eta_i\cdot P_j\cdot\alpha_{i,j}\cdot t_{aj}e^{-\ln 2\cdot t/T_{1/2\,j}}$$

Time t is measured since the beginning of the detector operation. Parameters $t_{a\,j}$ may be adjusted according to activation history of the detector including shielding, saturation of the activity, and storage underground before measurements.

Eq. 3
$$t_{aj}=\frac{T_{1/2\,j}}{\ln 2}\sum_k\frac{1-\exp\left(-\ln 2\cdot t_k/T_{1/2\,j}\right)}{s_k}\exp\left(-\ln 2\cdot t_{dk}/T_{1/2\,j}\right)$$

here s_k is shielding efficiency at k-th stage of a sample history, t_{dk} -- delay between the end of k-th stage and beginning of the detector operation. Note, that in our approach underground storage before data taking is hidden inside effective activation time.

In the work presented in 2006, Barabanov and Belogurov showed that a simple cylindrical movable iron shielding container may reduce isotope production rate by the factor about 15 for Co-60 and about 10 for Ge-68 in the mixture of 87% of Ge-76 and 13% of Ge-74. For other isotope mixtures the reduction factors are slightly different, but not significantly.

A cylindrical iron container was designed and used for transportation of the enriched germanium for the Phase II of GERDA experiment, See Figure 5. The container size is Ø140 cm x 126.5 cm. There is a cavity in the container Ø54 cm x 40 cm. The cavity is situated in such a way that the bottom thickness is only 3 cm. The total mass of the container is 14.5 tons. The fast nucleon spectra inside the iron container were analyzed. The spectra have different shape compare to the sea level ones. Most of the activations are produced by neutrons with energies around 100 MeV. For the first order conservative estimations of a shield efficiency an attenuation length of 240 g/cm^2 should be used.

Figure 5a-d. The transportation container for the transport of enriched GeO_2 powder from Krasnoyarsk to Munich, March 2006.

Using the estimates above, Figure 6. shows how the concentration of Ge-68 and Co-60 evolves during the GERDA Phase-II detector production, estimated with shielding during transportation and intermediate storage underground (at the HADES underground facility). The vertical red line shows the current situation. The future evolution is projection with conservative assumptions about the time needed for each production step like purification, crystal pulling and detector manufacturing, as shown on the figure.

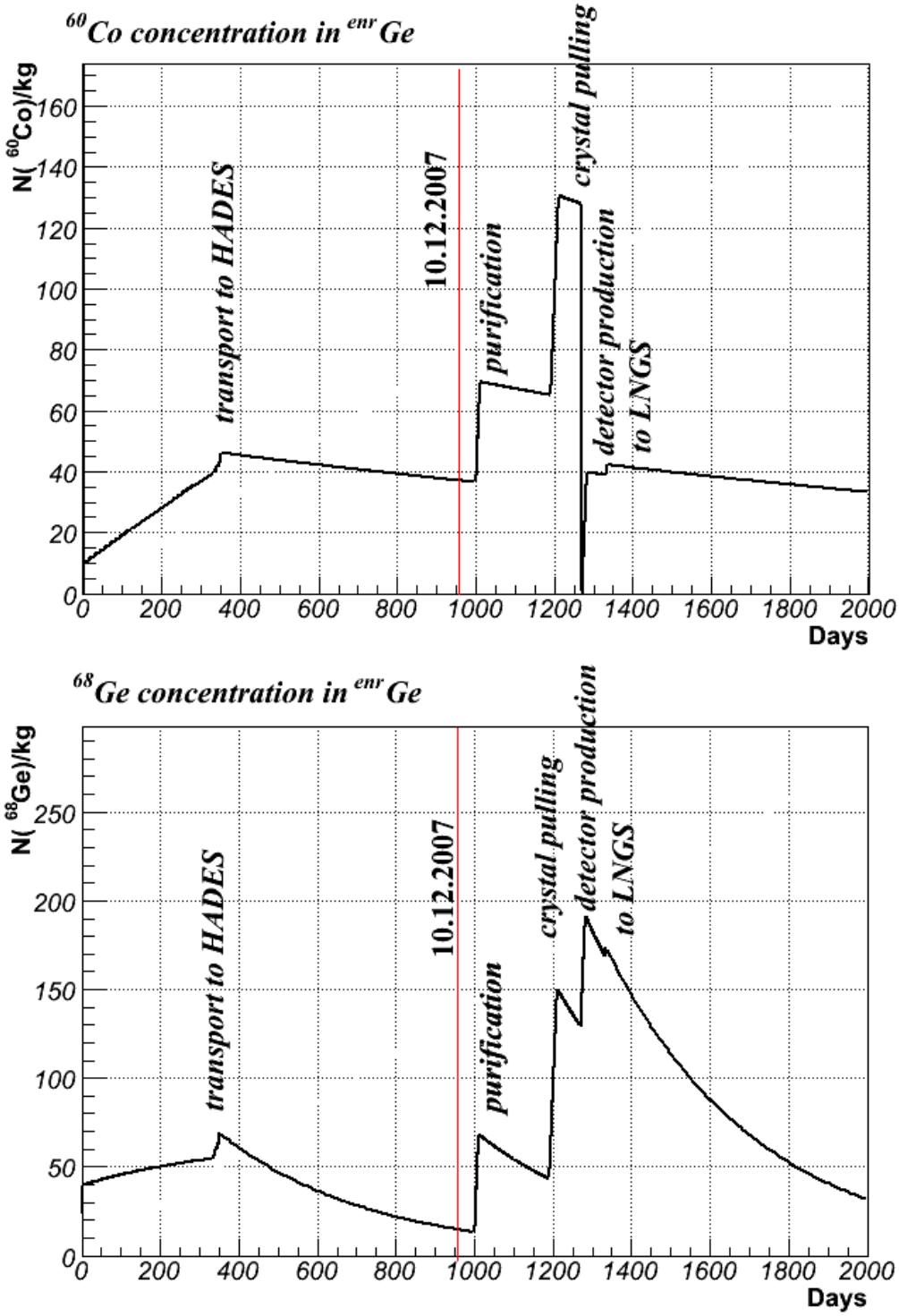


Figure 6: Ge-68 and Co-60 concentration during the process of the detector manufacturing

1.4. European users of HPGe produced underground

The European users of HPGe produced underground are mainly to be found amongst the deep underground laboratories (>1000 m w.e.) In case there were a production facility for this and the cost for underground produced Ge could be kept at bay, it is probable that also semi-deep underground laboratories (100-1000 m w.e.) would be customers. The advantage for shallow depth laboratories (< 100 m w.e.) is

Table 4. An overview of underground sites of laboratories in the European Union were physics experiment have been/are being/will be carried out or.

Depth (m w.e.)	Institute, Country	Name of underground laboratory	Main activity	CELLAR partner	Comment
5000	Italy	Mont Blanc tunnel	Cosmic ray studies		
4800	LSM/CNRS/CEA etc., France	Modane	Measurement of environmental radioactivity	YES (2 groups)	Frejus tunnel
4000	University of Oulu and Jyväskylä, Finland	Pyhäsalmi mine	Muon measurements		
3500	INFN-LNGS, Italy	Gran Sasso	Searching rare events like Dark matter and Neutrino interactions	YES	Teramo motorway tunnel
2800	Sheffield University and others, UK	Boulby Palmer underground laboratory	Astroparticle science		Rock salt and potash mine
2450	University of Zaragoza, Spain	Canfranc	Dark matter search and Neutrino Physics		
2000	PSI & other Inst., Switzerland	Gotthard tunnel	double beta decay		
2000	Polkowice, Poland	Sieroszowice mine	Searching rare events like Dark matter and Neutrino interactions		In planning stage
1500	Rustrel-Pays d'Apt, France	Laboratoire Souterrain Bas Bruit (LSBB)	Searching rare events like Dark matter and WIMPs		
1000	PTB, Germany	UDO	Dosimetry and reference measurements	YES	Salt mine Asse II, Braunschweig
600	University of Neuchatel	La Vue-des-Alpes	Radioactivity measurements		
500	EC-JRC-IRMM, Belgium (EU)	HADES	Reference measurements	YES	Underground laboratory in clay,in Mol, Belgium
500	IFIN-HH, Romania	Unirea	Low level measurements	YES	Salt mine
390	Mining Academy Freiberg, Germany	Underground lab Freiberg	Low level measurements		lead/silver mine
320	CERN, Switzerland	CERN	muon measurements		
300	Universita dell'Insubria, Italy	Baradello Hill	Materials selection for CUORE and environmental radioactivity	YES	Como
110	VKTA, Germany	Felsenkeller	Measurement of environmental radioactivity	YES	Rossendorf, Saxony
70	University of Bern, Switzerland	Underground lab Bern	Low-level counting		
70	Glasgow University, UK		GM-counter measurements of cosmix rays		Coal mine beneath Glasgow University
60	London, UK	Holborn	Radiopurity measurements for SNO		Tube station, lease terminated in 1993
30	IAEA-MEL, Monaco	CAVE	Marine environmental radioactivity	YES	
15	Max Planck Inst. für Kernphysik, Heidelberg	Low-level Laboratory	Measurements in support of underground science	YES	
~10	University of Tübingen		Measurements in support of underground science		Under construction

marginal so those users are not likely to generate any demand for such a product. Table 4 gives an overview of underground laboratories or sites where physics experiments and measurements have taken place or are being planned.

In the year 2000, eight underground laboratories involved in radioactivity measurements formed a network named CELLAR. The mission of CELLAR is "to promote higher quality and sensitivity in ultra

low-level radioactivity measurements for the improvement of crisis management, environment, health and consumer protection standards of Europe". It is probably fair to say that the promotion of (in particular) underground gamma-ray spectrometry has been successful as many new applications have been identified. Figure 7 shows the almost perfect linear increase with time of the analytical power (represented by the number of HPGe-detectors available for gamma-ray spectrometry) in the CELLAR laboratories taken place the past 20 years. This increase in analytical power is a clear indication that underground gamma-ray spectrometry is a technique with many benefits that have found an increasing number of applications in recent years. It is expected that this increase will continue as there are new laboratories starting up (e.g. in Poland and Romania) and existing ones expanding (e.g. Modane, Canfranc). The biggest underground experiment in Europe requiring HPGe-crystals produced underground is the double beta decay experiment GERDA. In its first phase only existing Ge-crystal enriched in Ge-76 are being used. GERDA Phase II will require pulling of some 14 crystals.

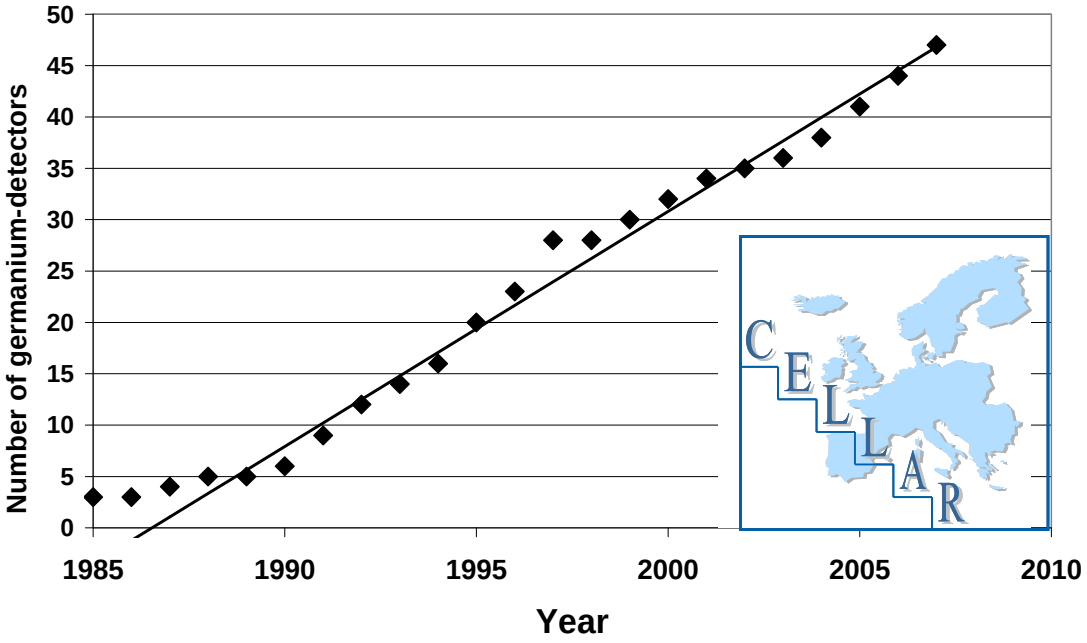


Figure 7. The number of low-background HPGe-detectors in operation underground within the present set of partner laboratories of the network CELLAR, shown as a function of time. The fitted trend line shows there to be a close to linear increase since 1990 with about 2.3 detectors per year.

1.5. World-wide users of HPGe produced underground

Table 5 gives an overview on underground sites outside the EU where physics experiment have been carried out or will be carried out. There are only few laboratories with several HPGE-detectors (e.g. Ogoya with about 12 detectors) so it is likely that there will be a bigger increase than 3 HPGe-detectors per year in these laboratories.

Table 5. An overview of underground sites of laboratories outside the European Union were physics experiment have been/are being/will be carried out.

Depth (m w.e.)	Intitute, Country	Name of underground laboratory	Main activity	Comment
8500	Tata Institute of Fundamental Research , India	Kolar Gold fields	Cosmic ray studies	Closed for experiments in 1992
6000	Snolab, Ontario, Canada	SNO lab - Sudbury Neutrino Observatory	neutrino physics	
4400 - 5500	USA	DUSEL - Deep Underground Science and Engineering Laboratory	Vast program on Cosmic rays and Neutrino Physics and support activities including radioactivity measurements	In planning stage - no decision yet: In the former Homestake gold mine
4400	JINR, Russia	Baksan	Cosmic rays and Neutrino Physics	Baksan Valley, Prielbrusye
3700	University of Bern, Switzerland	Gotthard tunnel	Cosmic rays and Neutrino Physics	
3500	India	INO - India based Neutrino Observatory	Cosmic rays and Neutrino Physics	In planning stage
3300	University of Sydney, Australia	Broken Hill mine	Measurements of mine environment / neutrino physics	silver/lead/zinc mine
2700	University of Tokyo, Japan	Kamioka Observatory	Cosmic rays and Neutrino Physics	rock
2000	University of Minnesota, USA	SUL - Soudan Underground Laboratory	Cosmic rays and Neutrino Physics	
2000	Seoul National University	Y2L	Double beta decay, radioactivity measurements	rock
1500	IMB - Irvine, Michigan, Brookhaven, USA	Morton salt mine	proton decay, supernova detection	Stopped in 1991
1400	Osaka University, Japan	OTO-Cosmo Observatory	Double beta decay	tunnel in rock
900	INR of UNAS, Ukraine	Solotwina	Double beta decay	salt mine
700	Gifu, Japan	Miboro		hydroelectric dam
600	California, USA	Orville Dam		hydroelectric dam
350	Science Institute of Iceland.		Measurement of low activity materials for scintillation detectors	In train tunnel - CELLAR partner
270	Kanazawa University	Ogoya	Radioactivity measurements	
180	Georgia	Saberio		limestone
180	Chiba, Japan	Nokogiriyama,		
150	University of Tbilisi, Georgia	Saberio		
100	Irvine, USA	Hoover dam		hydroelectric dam
100	Tokyo, Japan			University of Tokyo
70	University of Bern		Dark matter search	University of Bern
37	Pacific Northwest Laboratories, Richland, USA	Lower Monumental Dam		hydroelectric dam
17	Stanford, USA	Stanford Underground Facility	Dark matter	

Provided there were an underground facility for production of Ge-crystal, would such a facility be able to distribute crystals to distant locations? It is clear that special transportation is necessary or else the benefits of the underground production are lost. One case is known where 2 HPGe-crystals were transported by submarine from USA to Europe. This must be considered an exception and not an option that can be routinely used. It should, however, be possible to transport crystals near to the bottom of freight ships and covered by cargo that amounts to more than 10 m water equivalent.

The biggest experiment outside the EU requiring underground production of HPGe-detectors is the USA based double beta decay experiment MAJORANA (Aahlseth, et al. 2005). It is of the same order of magnitude as the GERDA experiment. Table 6 gives some numbers and estimates on HPGe-detectors in operation.

Table 6. Data and estimates regarding underground analytical power for gamma-ray spectrometry in 2007. Note that these numbers have an uncertainty that is not stated.

	Number of HPGe-detectors in operation
Gran Sasso (all groups)	12
Modane (all groups)	12
HADES	7
CELLAR in 2007	47
CELLAR in 1987	4
Estimated total in EU	60
Estimated total world-wide	100

2. Production and use of germanium

2.1. Overview of germanium producers

The production of HPGe-crystals with a purity adequate for making state-of-the-art HPGe-detectors is only performed commercially at three places in the world. In USA there are the two companies, Ortec and Tennelec. They were constructed near to the zinc mine in Tennessee. The third producer, Umicore, is located in Olen, Belgium, and also used residue from a nearby zinc factory. Today a large amount of germanium is recycled and advanced scrap smelters are installed with the producers.

Some research institutes like IKZ Berlin (Institut für Kristallzüchtung), the Germanium plant in Krasnoyarsk (Russia) and the Institute of Chemistry of High Purity Substance of Russian Academy of Science in Nizhny Novgorod have facilities for crystal production. For some of them it is also worth mentioning that although they have the theoretical competence, they lack the experience coming from many years of commercial production. Pulling big crystals of detector quality is to a large extent an art in the sense that there are many parameters to vary and it is not entirely clear how they interconnect. It is, however, not inconceivable that institutes or companies other than the three major ones could take up production of germanium crystals of detector quality.

The production of HPGe-detectors do not require as heavy equipment as crystal production and does not need labour performing shift work on the basis of 24 hours per day processes. This means that in principle it is easier to set up a production facility from logistical point of view. In recent years there have, however, been a re-structuring of the production facilities (merger of several companies) so that there are only a limited number of commercial producers in the world for the moment. The main producers are Ortec (USA), Areva (with Canberra and Eurisys in Belgium and France, respectively) Princeton GammaTech (USA) and DSG (Mainz, Germany).

2.2. The technical steps in the production of high purity germanium

2.2.1. Enrichment

To maximize the potential to observe neutrinoless double beta decay of ^{76}Ge the GERDA experiment requires vast quantities of germanium enriched in this isotope, many tens and better hundreds of kg. There are many methods for enriching isotopes. These isotopic separation methods include electromagnetic separation, diffusion, centrifuging, chemical and physical processes, laser ionization evaporation and plasma ion-cyclotron separation methods (see Tables 7 and 8).

Table 7. Main Enrichment Technologies and their short characteristics.

Method	Application	Productivity	Cost
Electro-magnetic isotope separator (calutron)	Universal (all elements)	Very low (tens grams/year)	High
Physical-chemical (rectification, chemical exchange etc.)	Light elements: N, O, C,..	High (tons/year)	Low
Optical (laser) (AVLIS[1], MLIS[2]), Photochemistry	Elements having shift of spectrum lines	High Hundreds of kg/year	Middle
Gas diffusion Thermoacustic diffusion	For all elements formed volatile gas compounds (under RT)	High Thousands of tons/year	Middle Low (?)
Gas ultracentrifuge	For all elements formed volatile gas compounds (under RT)	High Thousands of tons/year	Low
Plasma methods - ICR[3] - MPEP[4]	Universal (all elements)	High Hundreds of kg/year	Middle[3] Low[4] (?)

1* AVLIS – Atomic vapor laser isotope separation
2* MLIS – Molecular laser isotope separation
3* ICR – Ion cyclotron resonance
4* MPEP – mirrored plasma enrichment process.

Electro-magnetic isotope separator (calutron) should be excluded for ^{76}Ge production because of its low productivity and high cost. Chemical and physical processes are only useful for enrichment of isotopes of light elements. Optical (AVLIS) and plasma (ICR) methods in principle could be developed for ^{76}Ge enrichment, however there are no currently operational technologies for ^{76}Ge isotope (except enrichment of ^{235}U). Gas diffusion technology is applied, for certain reasons, only for enrichment of ^{235}U isotope. There are several promising R&D on new isotope separation methods that might enable ^{76}Ge enrichment at a low cost: **a)** new gas diffusion, namely thermoacustic diffusion method; and **b)** new plasma enriched method, the mirrored plasma enrichment process (MPEP). Unfortunately, there are still no big-scaled facilities based on these methods.

Table 8. Technologies used for isotope separation of the elements (in blue – gas centrifuge and diffuse, green – rectification and chemical exchange, yellow – laser (AVLIS and MLIS), lilac – plasma ICR, brown – optical (non laser), only for Hg).

H							He		
Li		B	C	N	O	F	Ne		
Na	Mg	Al	Si	P	S	Cl	Ar		
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni
Cu	Zn	Ga	Ge	As	Se	Br	Kr		
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd
Ag	Cd	In	Sn	Sb	Te	I	Xe		
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt
Au	Hg	Tl	Pb	Bi	Po	At	Rn		
Fr	Ra	Ac	Rf	Db	Sg	Bh	Hs	Mt	110

La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu

Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

As is well known, gas centrifuge technology had been initially developed for uranium isotope separation (E.I Abbakumov et al., 1989). A centrifuge designed for operation with UF_6 cannot be efficiently used for non-uranium isotope separation in different chemical compounds (V.D. Borisevich et al., 2000). Therefore, centrifuges with specific characteristics using either existing or specially synthesized gaseous chemical compounds have to be designed for this purpose. The basic condition for the applicability of the process gas is that the gas vapour pressure should not less than 5-10 mm Hg at RT. Besides, this substance should be corrosion compatible with the structural material of the centrifuge, it has to be sufficiently resistant to temperature dissociation and preferably must possess the maximum possible content of the desired element in the molecule. List of compounds used as working gas for centrifugation procedure is on Table 9.

Only the method of gaseous centrifuge enrichment is well developed for enrichment of ^{76}Ge isotope at industrial scale (tens and hundreds of kg). Moreover only this method is now the most cost-effective one (for example, electricity consumption is in 50 times less than that for gas diffusion method).

Table 9. Volatile compounds for centrifuge separation.

Fluorides, oxyfluorides,	**Oxides**	**Metal organics**	**Noble gases**	**Chlorides, freons**	**π-complex (carbonyls)**
SiF_4, SF_6, **GeF_4**, SeF_6, TeF_6, MoF_6, IrF_6, WF_6, CrO_2F_2, POF_3, $SiCl_3H$, NF_3	OsO_4 CO_2	$Sn(CH_3)_4$, $Pb(CH_3)_4$ $Cd(CH_3)_2$, $Zn(CH_3)_2$ $Ga(CH_3)_3$, $Hg(CH_3)_2$ $Pb(CH_3)_4$, $Zn(C_2H_5)_2$ $In(CH_3)_3$	Xe, Kr	$TiCl_4$, $GeCl_4$ $(CBrF_2)_2$ **BF_3, ClF_3,** $C_7F_{14}CF_4$	$Fe(CO_3)_5$, $Ni(PF_3)$,

There are many centrifuge separation and enrichment facilities for enrichment of uranium around the world: in Russia, China, Brazil, the EU, Japan and USA.
A centrifuge is a facility, which creates centrifugal forces. These forces act on molecules of highly volatile compounds in the vapour and gaseous phases and induce their separation by mass (Fig. 8). A high-speed rotor operates inside a vacuum chamber provided with water cooling. The rotor rests on the lower bearing. There is “magnetic” suspension assembly at the top. This “magnetic” suspension assembly creates the magnetic field whose attractive force mostly compensates the load acting upon the lower bearing.

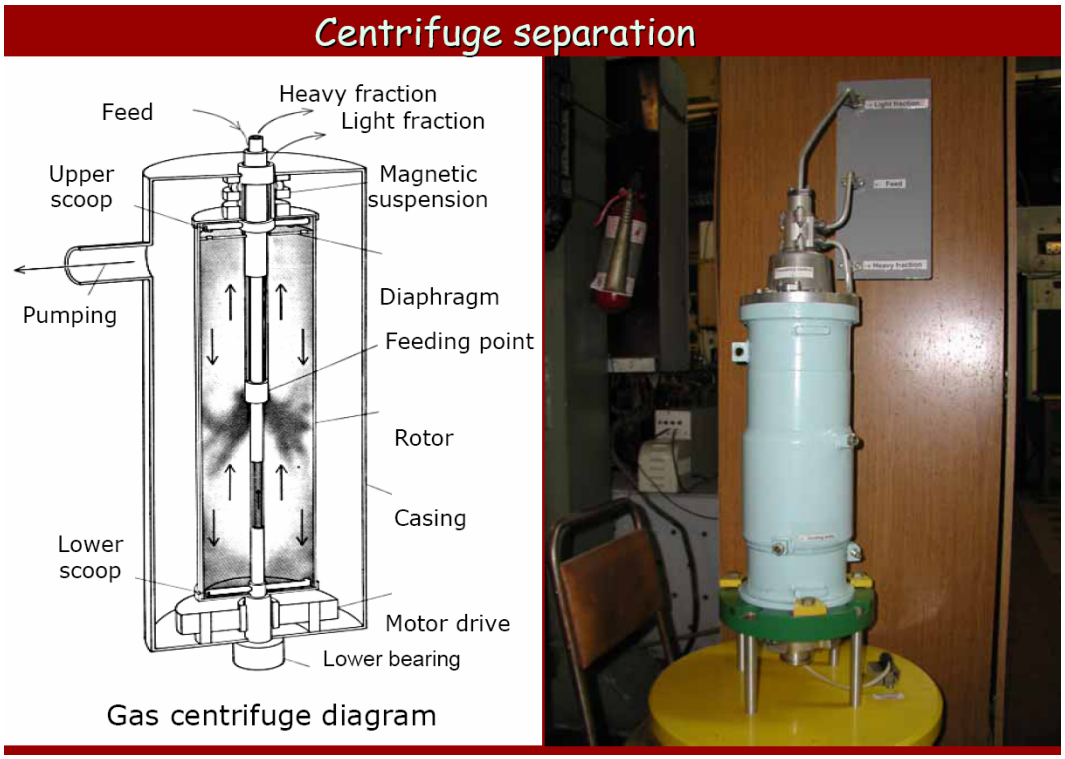


Figure 8. Principle of gas-centrifuge isotope separation (G.Yu. Grigoriev, 2003).

Process gas is introduced into the centrifuge through a feed port in the middle part of the rotor. The scoops (extractors) are located at the extreme ends of the rotor. The top stream of gas enriched in the light isotope and depleted in the heavy one, while in the bottom stream the reverse is true. The separation axial effect is amplified with the help of the circulation rotating flow. Individual centrifuges are connected in a cascade. The length of the cascade and the number of centrifuges in the stage depend on the required separation power.

Separation rate of individual centrifuge unit is depended on diameter and length of a rotor of a centrifuge and its speed of rotation. Efficiency of centrifuge unit is proportional to length of its rotor. At the same time a working frequency of a centrifuge is increased with increasing of its length and for certain lengths this working frequency is equal to proper frequency of centrifuge. At this moment mechanical load on rotor's bearing is maximum and rotor could be destroyed. There are two types of a centrifuge-units: sub-critical (working frequency is less than proper frequency of centrifuge) and above-critical (working frequency is above/beyond than proper frequency of centrifuge) ones (G.Yu. Grigoriev, 2003). Soviet/Russian centrifuges are sub-critical ones and they are very reliable in service. Centrifuge units in USA are above-critical ones, with diameter above 50 cm and lengths about 6 m (such a set-up accumulates huge kinetics energy). After several accidents with these centrifuges, the US program has been shut down. Centrifuge units in the EU (URENCO company) are also above-critical ones, but with lengths about 2 m.
Russian centrifugation facilities belong to the Federal State Unitary Enterprises of the Ministry of the Russian Federation for Atomic Energy: The Ural Electrochemical plant at Novouralsk (former Sverdlovsk-44) Sverdlovsk region, the Electro-Chemical Plant at Zelenogorsk (former Krasnoyarsk-45) Krasnoyarsk region, the Siberian Chemical Complex at Seversk, (former Tomsk-7) Tomsk region, and the Electrolysis Chemical Complex at Angarsk, Irkutsk region. Total productivity of these enterprises are about 20 10^6 of SWU's (separation working units) or about 77% of world "centrifugal capacity" (see review in (V.N. Kornoukhov, 2003)). All of them are devoted to separation of uranium, except a handful of facilities, which are specialized on enrichment of stable isotopes for science and technical application. Biggest of such facility, Svetlana Department of the Electrochemical plant (Fig. 9) is placed in Zelenogorsk (Krasnoyarsk region).

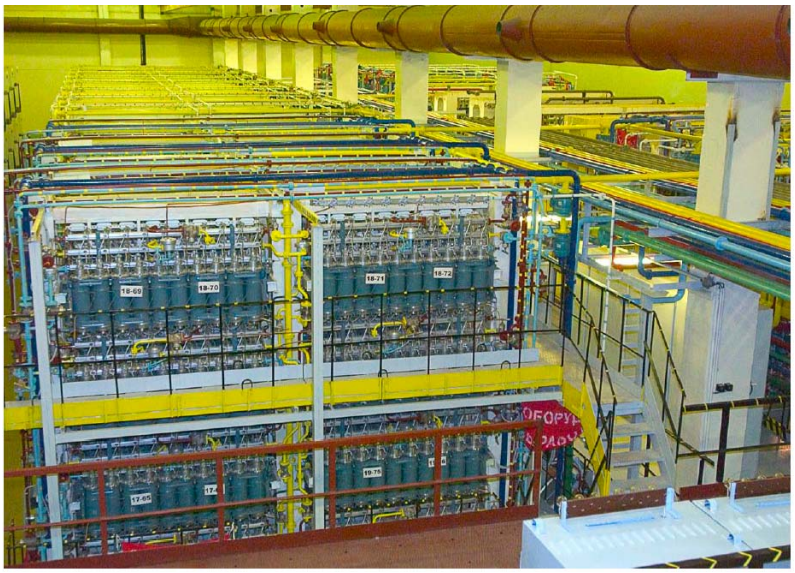


Fig. 9. Cascades of the centrifuges for ^{76}Ge production at the Svetlana Department**.**

The Svetlana stable isotope production capabilities are practically unlimited: from milligrams to hundreds of kilograms (^{57}Fe, ^{76}Ge, ^{82}Se, ^{100}Mo, ^{116}Cd, ^{130}Te, ^{129}Xe, ^{131}Xe, ^{136}Xe). The Plant was commissioned in 1962 and since 1987 it has been producing only low enriched uranium for nuclear power plants using centrifugal technology. In 1972 the iron enriched in Fe-57 was produced in the cascade of gas centrifuges at the Plant. That initiated the development of stable isotope separation based on centrifugal technology. In the 1980s the decision was taken to move the ECP isotopic production to a specially intended workshop. The facility was built at the expense of ECP and given the romantic name SVETLANA (E.Nikitina, 2007).
The Svetlana Department delivers the enriched germanium in form of dioxide $^{enr}GeO_2$ of technical grade quality, 99.8%. Productivity of this facility can be estimated as about 80 - 100 kg/year in standard mode.

Fig. 10 depicts the gas centrifuge enrichment process. The process starts with conversion of pure Ge metal into a gaseous form ($^{nat}GeF_4$). The gas passes through cascades of centrifuges to achieve the desired enrichment of not less than 86% and then it is converted into germanium dioxide ($^{76}GeO_2$) via a hydrolysis procedure. Then germanium dioxide is treated under high temperature (drying and calcinations procedures) to remove a residue of water and other impurities.

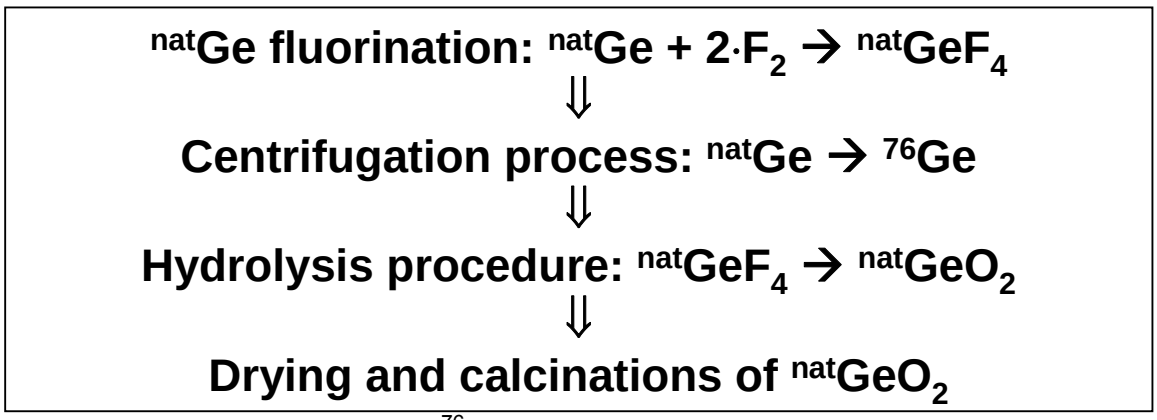


Fig. 10. General scheme of ^{76}Ge production at Svetlana Department of the ECP.

GERDA issues

For the case of the GERDA detector and other underground HPGe-detector systems it is so that when the external background is reduced, an internal background from long-lived radioactive isotopes in the germanium crystal itself will play more important part. For germanium diodes the main dangerous isotopes are ^{68}Ge and ^{60}Co. They are created mostly by spallation reactions of cosmic radiation at sea level (mainly nuclear component, fast neutrons and protons) when the enriched isotope is above ground: during isotope production, purification and transportation, and also during fabrication of detectors (G.V. Gorshkov et. al., 1966; A.A. Vasenko et. al., 1989; F. Avignone et. al**.,** 1992). Cobalt is removed while germanium remains during chemical purification and zone refinement and while a germanium monocrystal is growing. This isotope will be continuously produced while the germanium monocrystal is above ground, i.e. during detector fabrication and its transportation to the underground laboratory. The isotope ^{68}Ge, once produced, cannot be removed from germanium during chemical purification and zone refinement. The only solution is minimize the duration of the stay of enriched germanium above ground or to substantially decrease the cosmic radiation exposure of the germanium.

Below we describe the joint work of the SVETLANA Department of the ECP, INR RAS, SSC ITEP and Max Planck Institute of Physics (Germany) on substantially decreasing of the activation of enriched germanium during production, storage, and transportation from Zelenogorsk to Munich (Kornoukhov, 2005, Shubin et al., 2006).

Calculation of ^{68}Ge and ^{60}Co production have been made by the Sobolevsky' group (INR RAS) according to the SHIELD program (Sobolevsky, 2004; Dementyev and Sobolevsky, 1999; Barabanov et al., 2006), for details see Section 1.3. The results are listed in Table 2 (Section 1.3). During centrifugation process these isotopes, together with the light fraction of germanium (others light germanium isotopes) are separated from the heavy fraction. Thus the process of producing the heavy fraction of ^{76}Ge removes ^{68}Ge and ^{60}Co isotopes. The production of new ^{68}Ge and ^{60}Co isotopes starts right away as portions of gaseous $^{76}GeF_4$ come from the last stages of the cascade to receiving balloons.

General rule to minimize activation is minimize the duration of the stay of enriched Ge above ground or to substantially decrease the cosmic radiation exposure of the Ge.

a) Production. Isotopic composition of enriched germanium. According to standard technology, the collection of a batch of $^{76}GeF_4$ into balloons, the chemical conversion of this compound to germanium dioxide and the drying and calcination process takes an average of 40 days. After modernization of technology accordingly to GERDA' demands, the average time of production of regular portion of enriched germanium is 74 hours (3.1 days).

From Table 9 one can see that the ^{68}Ge production rate is strongly depends on atomic number of germanium isotope and this number for lightest isotope, ^{70}Ge is 60 times as much as for ^{76}Ge isotope. This is why the content of ^{70}Ge isotope should be depleted by 2 orders relative to the ^{76}Ge isotope.

The isotopic composition of enriched germanium for GERDA Collaboration after proper tuning of the cascades is listed in Table 10. As a result, the production of ^{68}Ge in enriched germanium is decreased 14 times compared to activation of natural germanium.

Table 10. Isotopic composition of enriched germanium for the GERDA experiment (V.N. Kornoukhov, 2005).

Isotope	Natural germanium, %	Enriched germanium, GERDA, phase II, %
Ge-70	20.54	0.01 - 0.02
Ge-72	27.54	0.05 - 0.1
Ge-73	7.74	0.14 - 0.17
Ge-74	36.43	11.93 - 12.78
Ge-76	7.75	86.94 - 87.87

b) Storage. The produced portions of enriched germanium (in form of germanium dioxide $^{76}GeO_2$) are poured into hermetic plastic cans and put into underground storage. The storage is in an underground room with multiple layers of concrete and earth with a total thickness of about 900 g/cm^2. For further reduction of the cosmic rays flux a container made of steel was installed into this storage. Its dimensions are 100 x 100 x 82.5 (H) см^3 having inner cavity of 50 x 50 x 40 cm^3 to container storage of the enriched isotope **(**Fig. 11). According to our estimation, the reduction of the production rate of ^{68}Ge and ^{60}Co isotopes on enriched germanium by cosmic rays (nuclear component and muons) is a factor of more than 30.

c) Transportation. To suppress activation of enriched germanium during its transportation from Zelenogorsk to Munich, a special compact transport container made of steel has been designed and fabricated (see Fig. 5 in Section 1.3). Its dimensions are ∅140 cm x 126.5 cm (H) and its weight is 15 tons. Enriched germanium is placed inside the container into a special cavity. Dimensions of this cavity are ∅54 cm x 40 cm (H). Results of calculation of activation rate when enriched isotope is in the container and outside of it are listed in Table 11. To demonstrate the possibility of transportation, and to investigate possible delay of delivery including custom formalities (when truck crosses the borders), we accomplished conveyance of this container from Zelenogorsk to Munich. A portion of 15 kg natural germanium was placed into this container. Block-to-block time took 20 days.

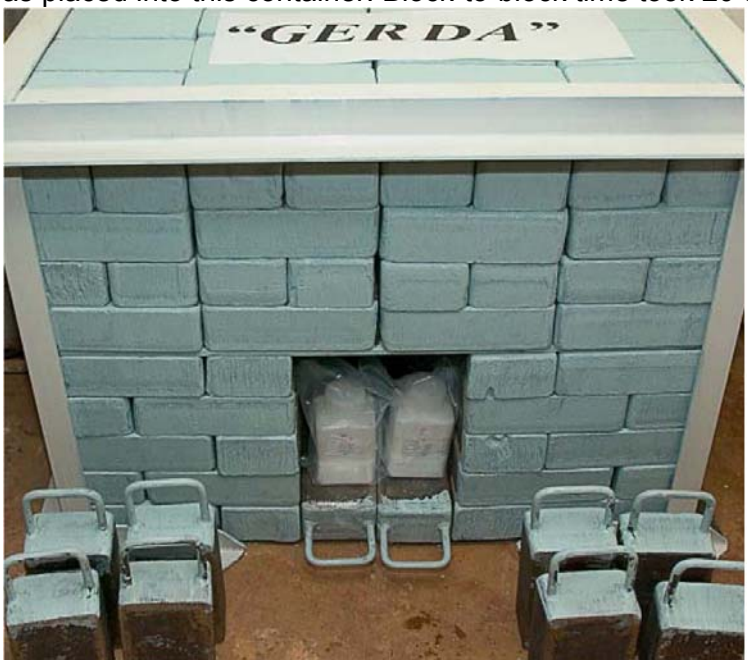


Figure 11. Container storage at the shelter in the Svetlana Department.

Table 11. Production rate of Ge-68 and Co-60 on germanium isotopes by nuclear active component of cosmic rays (neutrons and protons) placed into transport container, atoms/day/kg. Reduction factor for production of Ge-68 and Co-60 compare with production rate at sea level (out of shielding) (V.N. Kornoukhov, 2005).

Isotope-target	**Neutrons**		**Protons**		**Sum (neutron s+ protons)/ Reduction factor**			
	Ge-68	Co-60	Ge-68	Co-60	Ge-68	K_{red}	Co-60	K_{red}
Ge-70	28.06	0.09	4.9	0.028	32.96	8.5	0.12	14.7
Ge-72	5.24	0.21	0.96	0.046	6.2	8.9	0.26	11.3
Ge-73	2.49	0.23	0.45	0.035	2.94	9.5	0.27	11.9
Ge-74	1.22	0.18	0.24	0.05	1.46	10	0.23	14.6
Ge-76	0.34	0.12	0.06	0.036	0.4	10.6	0.156	21.2

Based on data from Table 2 (Section 1.3) and Table 10-12, one can conclude that suppression of enriched germanium activation by the nuclear component of cosmic rays is about of factor of 10 (for ^{68}Ge isotope) and factor of 17 (for ^{60}Co isotope). Taking into account capture of cosmic ray muons and direct interaction of muons with germanium nuclei, suppression factors for ^{68}Ge and ^{60}Co are 8.5 and 13 respectively. It means that effective irradiation time during transportation of enriched germanium from Siberia to Germany is about 3 days instead of 20-calendar days of actual duration of the conveyance.

Conclusion on job made by GERDA Collaboration and the ECP at the site. Schedule of ^{60}Co and ^{68}Ge production at different steps of the ^{68}Ge production at Svetlana (in effective days) is presented in Table 12. One concludes, that biggest contribution to activation of enriched germanium for GERDA Phase II was come when the material was at the underground storage at the Svetlana. It explain period of time while the material was in the storage, 1.5 year.

To have the same effect of activation as during production and transportation, the enriched material should be kept at the site about 0.5 year (180 days) or the Svetlana Department should have a storage at depth of several tens meters of water equivalent.

To decrease activation of enriched germanium during its production and storage at the ECP and during its transportation to Germany, much care has been taken. Production of ^{68}Ge and ^{60}Co has been decreased to a level, which satisfies the requirements of the GERDA experiment

Table 12. Schedule of ^{60}Co and ^{68}Ge production at different steps of the ^{68}Ge production at Svetlana (in effective days).

Procedure	^{68}Ge		^{60}Co	
	before	after	before	after
$^{76}GeF_4$ collection into balloons after last cascade	20 days	in total: 3,1 days	20 days	in total: 3,1 days
Hydrolysis procedure $^{76}GeF_4 \rightarrow {}^{76}GeO_2$	20 days		20 days	
Storage at the shelter 14 m.w.e.	~ 548 days	18,3 days	~ 548 days	18,3 days
^{76}Ge transportation from Zelenogorsk to Munich	21 days	2,5 days	21	1,6 days
Total	609 days	~ 24 days	609 days	23 days

2.2.2. Reduction of the raw materials

The metallurgical processing starts with GeO_2, which needs to be reduced to obtain metallic germanium. Reduction, as its name suggests, takes place in a reducing atmosphere, usually hydrogen mixed with some inert gas like N_2. The temperature has to be carefully controlled and to achieve the desired purity a clean environment and a special graphite container is needed. GeO_2 contains the stoichiometric ratio (0.694) of germanium and in powder form has a relatively low density, so the reduction will have a relatively small yield of metal.

The quality of the reduced metal should not be worse than the starting oxide's, purity that can be achieved by chemical methods. This usually means 4N or better purity. Resistivity measurement can be used to verify the purity, at this stage of processing the resistivity should be around 1 Ω·cm.

2.2.3. Zone refinement

Zone-refinement (ZR) in practice means that a molten zone pulled over the metal ingot, hence the name "zone-refinement". Because of the different segregation constant of the different chemical elements, impurities will be concentrated in the head and the tail of the ingot. To melt the metal only in a thin zone, an inductive heating coil is used. The whole process takes place in reducing atmosphere,

to prevent oxidation and neutralize other impurities. The container is a special graphite "boat" and the heating coil moves over the ingot very slowly, with a speed of a few cm per hour.

Commercially available zone-refined germanium has a purity of 6N or slightly better. Usually during the production, the only measure of the purity is the resistivity of the resulting material. We know that the intrinsic resistivity of germanium is around 50 □*cm (cite Knoll) and empirically was found that at room temperature the resistivity saturates at around 50 □·cm when 6N purity is achieved. By consequence, with a relatively simple resistivity measurement we can say if we achieved the 6N purity or not.

An example of resistivity measurement along the zone-refined ingot is shown on Fig.12. We can clearly see that the resistivity drops sharply towards the tail of the ingot, where the impurities are supposed to be concentrated. The measurement is done by the so-called two-probe method, as described in ASTM F *43-88.*

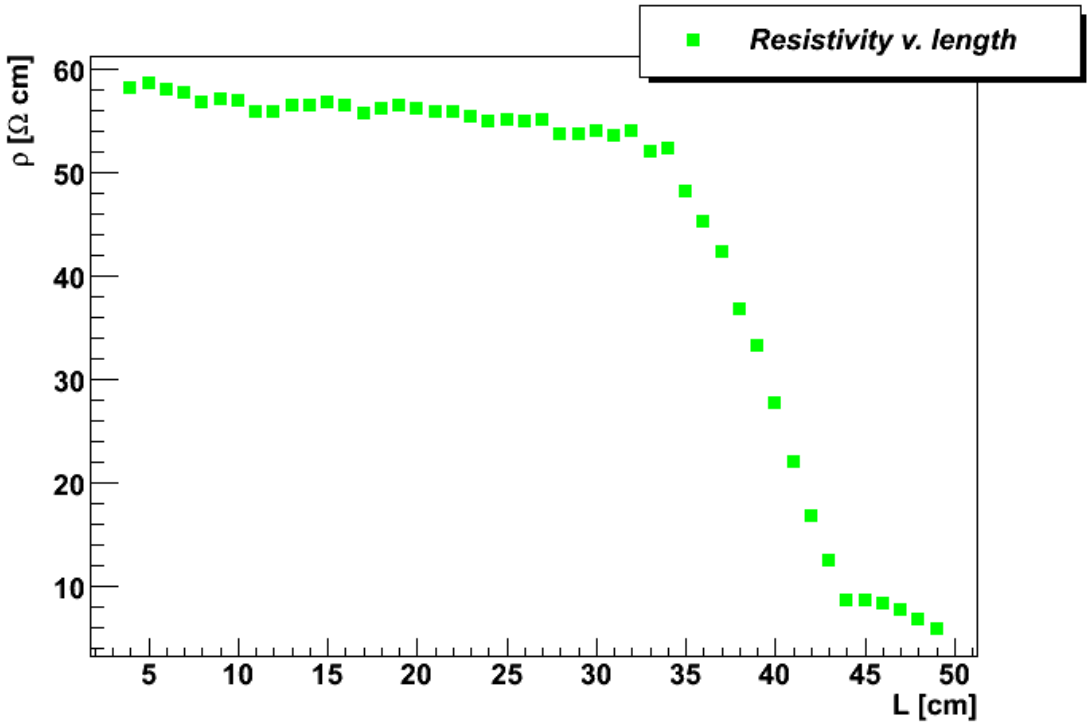


Figure 12. Resistivity in function of the distance from the head of the ingot. Measurement done at PPM Pure Metals

Typically, the yield of 6N material after zone-refinement is around 60%, but depends strongly on the procedure and materials used.

Detector grade crystals can be made of high purity material only. With 6N purity we already reached the purity that cannot be measured any more with mass spectrometry methods. In order to measure the net carrier concentration and the chemical composition of the impurities, more sensitive methods are needed, like Hall-effect measurement and Photo Thermal Ionization Spectroscopy (IPTS) (cite Haller). Unfortunately, these measurements can be done on crystalline material only. This will require several iterations of crystal pulling, analysis and zone-refinement.

2.2.4. Recycling of material

In order to achieve yield above 60% (yield of one ZR pass), we investigating the possibility to zone-refine the low resistivity material over and over again, until we reach a saturation level, i.e. where ZR cannot produce 6N material any more. The low resistivity tails after the first ZR test were cut off and zone-refined again. Results from the first test at PPM Pure Metals show, that after a second pass of ZR, the yield of high resistivity material is still around 60%.
During the forthcoming test we will try to melt together all the low resistivity material and zone-refine until there is enough material to fill a boat. Our expectation is that yield above 80% should be possible.

2.2.5. Czochralski Growth (crystal pulling)

Detector grade crystals are grown with the Czochralski (Cz.) method. Schematic view of a Czochralski puller shown in Fig. 13.

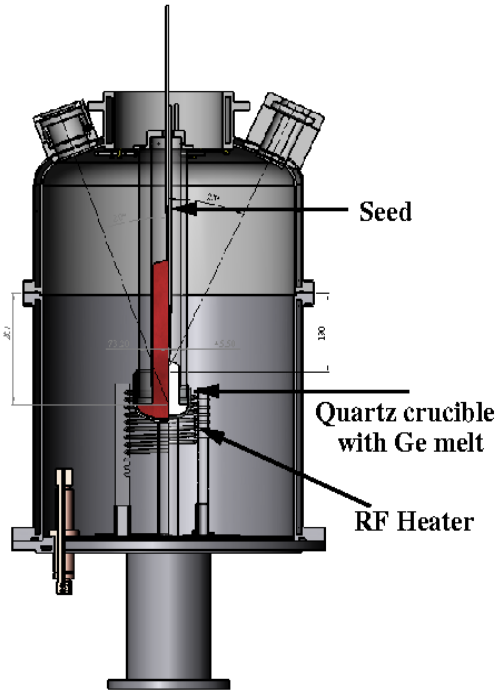


Figure 13. Schematic view of a Czochralski puller, picture courtesy of IKZ Berlin

Crystals are pulled from a melt of germanium, contained in a quartz crucible of ultra-high purity. Resistive and inductive heating can be equally used to melt the material.
Crystal pulling starts with a seed, a small crystal, which has the desired orientation of the crystal axis. During the pulling, the crystal and the crucible are rotating in opposite directions to produce a crystal with circular shape. A reducing atmosphere is used inside the puller. The gas inside the puller can also contain trace elements used for doping, to produce crystals of P or N type.

Pulling germanium crystals should be less difficult, than silicon for example, because of the lower melting point and resulting lower thermal gradients. Unfortunately, germanium like water expands when it freezes. This may cause the quartz crucible to brake at the end of crystal pulling, significantly increasing the cost of manufacturing.

2.2.6. Manufacturing of HPGe-detectors

To produce HPGe-detectors from good quality Ge-crystals does not require as big instrumentation as the production of Ge-crystals. It is, however, in the same way as crystal production a mixture of art and science in the sense that the quality of the final product is depending on small variations in many

different parameters. It is thus necessary for a producer to build up experience from producing a large number of items and iteratively improve the process by varying different parameters. The main steps in producing a HPGe-detectors from a good quality crystal are

1. Mechanical machining of the crystal
2. Forming of contact structures using Li-diffusion, implantation of different ions and creation of passivation layers
3. Testing of the crystal and in case of failure re-forming the contact structures
4. Placing the crystal inside the cryostat and making the electrical connections.

The most sensitive step is of course the formation of the contact structures (point 2 above). Generally speaking, the problems of obtaining a good charge collection grows with the size of the crystal. Different manufacturer have slightly different approaches to the final design of the crystal and how to create the best contact structures.

3. Optimisation of logistics for above ground production

3.1. GERDA Phase I detectors

The detectors used in Phase I of the GERDA experiment were "old" enriched crystals previously used in the Heidelberg-Moscow (HdM) double beta decay experiment (Klapdor Kleigrothaus et al., 2001) and the IGEX experiment (Aalseth et al., 2002). The 5 HdM crystals have been kept deep underground in Gran Sasso for about 15 years and the 3 IGEX crystals were kept deep underground in Canfranc. In 2006 they were all transported to the semi-deep underground laboratory HADES, which is located at the premises of the Belgian nuclear research centre, SCK•CEN (StudieCentrum voor Kernenergie • Centre d'Etudes Nucleaire) in Mol, Belgium. HADES is operated by the Belgian organisation EURIDICE, but it was the Joint Research Centre, IRMM (Institute for Reference Materials and Measurements) that was responsible for the storage and a the transports. HADES is located in a clay layer at a depth o f 225 m, which corresponds to 500 m w.e., see Figure 14.

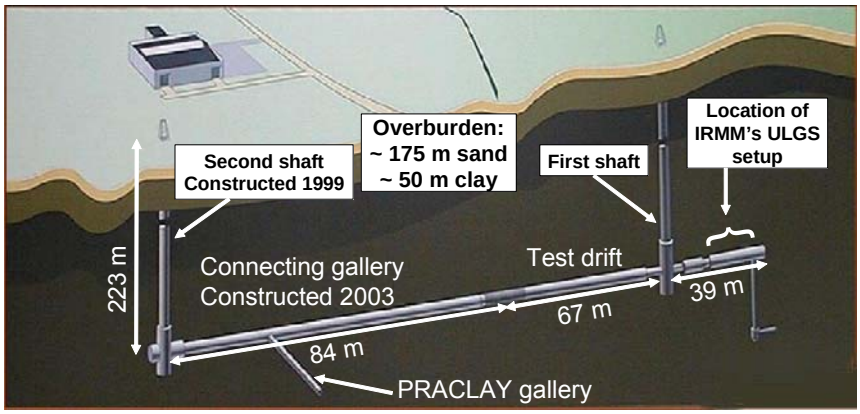


Figure 14. Schematic drawing of the underground laboratory HADES.

HADES is a laboratory were SCK•CEN is studying the geological properties of the clay layer and were IRMM is performing ultra low-background radioactivity measurements. The muon fluence rate in HADES is about 0.1 $s^{-1}m^{-2}$, which is almost a factor of 10,000 lower than above ground. The average radon concentration in HADES is low, about 7 Bq/m^3, due to not very high levels of U in the surrounding materials as well as constant flushing of the gallery with air via an air-conditioning system. The HADES underground laboratory has proven to be a good place for storage of materials that require at least semi-deep underground storage.

The HPGe-detector manufacturer Canberra Semiconductor n.v. is located in Olen only 17 km away from HADES. The strategic position of IRMM and its involvement in underground gamma spectrometry work enabled IRMM to take a leading role in optimisation of logistics for the re-working of the crystals for GERDA phase I. It was necessary that all the old crystals were re-worked in order for them to have the same contact structure so that the same type of electronics could be used. When this report is finalised (December 2007) the contact structures of the front, sides and bore have been created for all the GERDA Phase I detectors, which in total has included 23 transports.

The GERDA detectors will be operated immersed in liquid argon. In normal HPGe-detectors the crystals are inside a cryostat with vacuum. The immersion in liquid argon meant that the contact structures and passivation layers need to be slightly modified from the standard design. This work has been carried out on some non-enriched crystals. As described in Chapter 3.1 this served also as a benchmark test and trial before producing the GERDA Phase I crystals. Up until today 17 transports of test crystals have been performed. Certain processes like the Li-diffusion require many hours of access to the crystal, which meant the crystals needed to be picked up before 8:00 and returned after 16:00 in HADES. The safety regulations of HADES limits work from after 8:00 up until 16:00 on workdays. Through ILIAS it was possible to arrange a separate contract with EURIDICE for extra access from 7:30 until 16:45 which enabled Canberra to perform all the necessary work in time for the crystals to return underground at the end of the day. In most cases the above ground stay was shorter than the 8.5 hours that was planned.

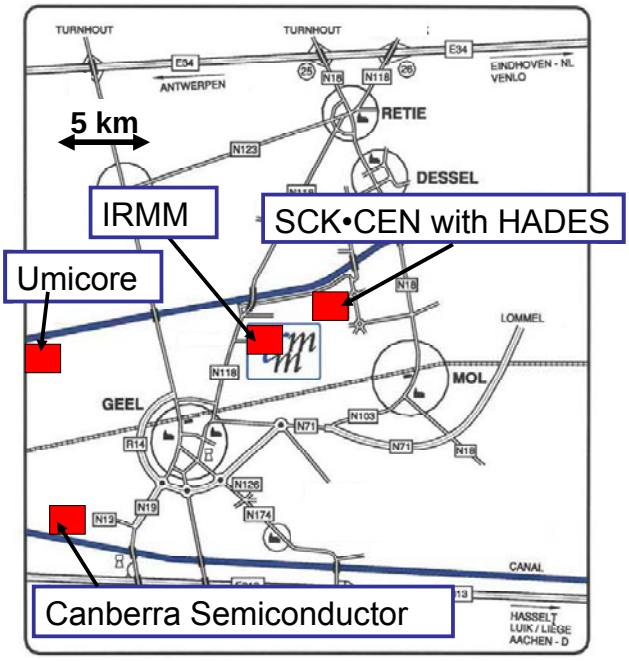


Figure 15. Overview of the location of sites important for the production of the Ge-crystals for the GERDA experiment.

In addition to the GERDA Phase I and GERDA test detectors, ILIAS has enabled IRMM to coordinate logistics for germanium crystals to be used in deep underground laboratories for other purposes than double beta decay measurements, 12 such transports have taken place.

For GERDA Phase II the natural step was to introduce discussions with the Ge-crystal producer Umicore in Olen. Umicore is also located 17 km (~ 25 minutes drive from "door to door"). Therefore all (37.5 kg) the enriched GeO_2 powder that was produced in Krasnoyarsk was transported for storage in HADES. The vicinity of HADES of Umicore is a guarantee that the cosmogenic production during Ge-crystal production above ground will be minimised. Meanwhile there were also discussions on possibilities of placing one or two production steps in HADES (See Chapter 4), but this was found to be unrealistic at this stage.

Table 13. Data on distances and transport times from "door to door", relevant for the logistics of production of GERDA Phase I detectors.

Distance (km) / Time (min)	**IRMM**	**SCK (HADES)**	**Canberra Semicon-ductor**	**Umicore**
IRMM	0	4.5 / 10	12 / 20	12 / 20
SCK (HADES)	4.5 / 10	0	17 / 25	17 / 25
Canberra Semicon-ductor	12 / 20	17 / 25	0	5 / 10
Umicore	12 / 20	17 / 25	5 / 10	0

4. Production of HPGe underground

4.1. Selection of suitable sites

The criteria for the underground production site could be prioritised in the following way:

1. SAFETY: Possibility of adequately solving safety issues mainly relating to the use of hydrogen gas
2. SPACE: Having enough space available so that no other experiments interfere or suffer from the activities
3. INTERFERENCE: No other activity should significantly disturb this activity nor should this activity disturb other activities such as experiments or measurements
4. OVERBURDEN: The depth of the laboratory
5. LOCATION: The location, in order to minimise transport times to other laboratories.

The three first point do somewhat coincide since having ample space and not interfering with other activities creates better safety. Note that for the sole production of HPGe-detectors the safety issues are much less complex than for the production of Ge-crystals. One can furthermore imagine that the extra safety procedures for the safe handling of hydrogen gas (and other gases) would hamper production rate. This would not be a problem for cosmogenic production (like above ground) but it would increase the cost of operation.

The overburden is not as critical for the production as it is for the future use of the detectors. As discussed above already 10 m w.e. is sufficient to create a great improvement compared to above ground production. It would however somewhat wasteful to construct an expensive production facility at such a shallow depth and for future needs (e.g. GERDA Phase III) it would indeed be beneficial with a deeper location. As a guideline value one could say that at least 100 m w.e. would be wishful for such an installation.

Figure 16 shows an overview of European underground laboratories (some in planning stage), that would be potential users of germanium produced underground. It is clear that all transports have to be by road or train transports. Transports by truck has the advantage that transport containers similar to the one in Figure 5 can be used. In principle that would be possible for train as well but the logistical problems are somewhat greater. Train transport is a good option if fast trains are used and heavy transport containers are deemed not to be necessary. Today it is possible to reach the Alps by fast trains from Antwerp although with change in Paris so a possible production centre in Belgium is a realistic possibility. With a solely European perspective it is clear from Figure 16 from an underground production site located in Central Europe, all laboratories indicated in Figure 16 could be reached by road within 24 hours or in some cases 48 hours. But as pointed out above the transport problem would be of less concern than other aspect so in principle none of the sites indicated in Figure 16 would be excluded solely based on their location.

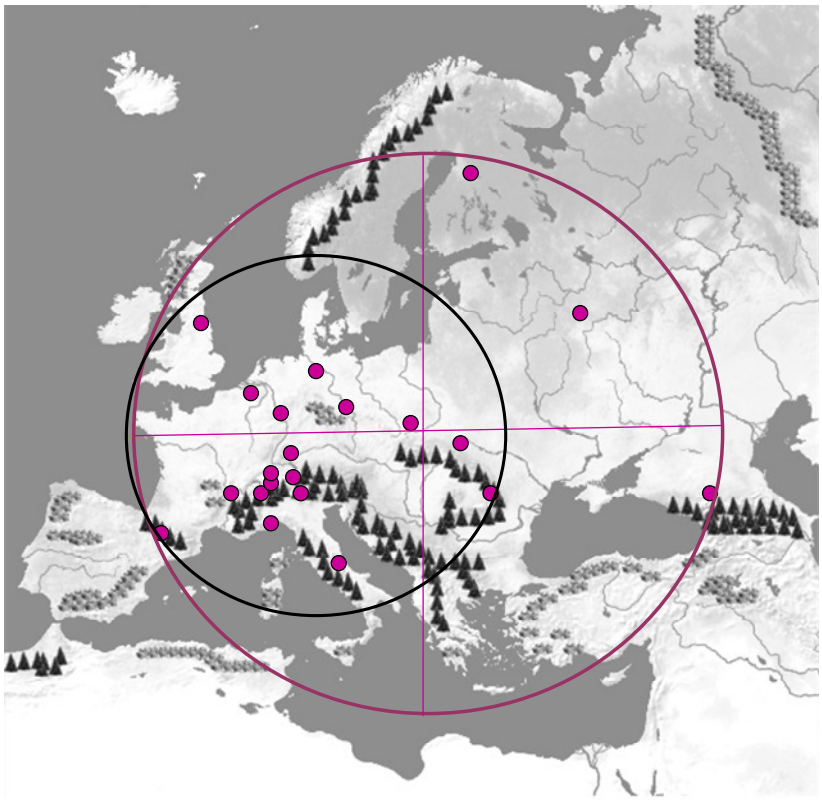


Figure 16. Overview of the location of European underground laboratories (some under construction or planning). The purple and black circles have radii of 1650 km and 1100 km, respectively.

4.2. Selection of suitable production steps

As has been discussed in the previous sections, the production of HPGe detectors requires several steps. In the case of detectors based on enriched Ge, the first step is the enrichment process. Enrichment requires a very large facility and extremely large investments. The activation during this

phase is also not critical. It is therefore not foreseen that enrichment will be performed underground. It is assumed that the starting material will be in the form of GeO_2. Two possible paths are then possible:

Path 1

(a) Chemical purification, via $GeCl_4$ chemistry. This is the standard procedure in industry. It is highly efficient if performed with equipment scaled for the application. The material is returned to GeO_2 form after the purification.

(b) Reduction to Ge in Hydrogen furnace. The yields in this step are typically well above 99%.

(c) Zone refinement of the Ge. The yield per step is approximately 60%. The low-purity tails can be further zone refined, increasing the yield. However, to reach yields above 90%, which is desirable when working with enriched materials (because of the cost of enrichment), then it is likely that tails will have to go back to the chemical purification step.

(d) Crystal pulling. Here, only a small yield can be expected for any given crystal-pulling step (20%). The remaining material has to be returned to step "c", and eventually also to step "a".

(e) Detector manufacturing. The yield of this step is not well known, but clearly is also not 100%, so that materials will need to be recycled through the various steps.

Path 2 is the same as Path 1, except that step "a" is skipped. This means a smaller yield in step "c". For maximum yield, it is to be expected that chemical purification facilities will also be necessary.

In addition to the low-purity tails which will need reprocessing, there are also significant amounts of material lost in grinding, etching, etc., which should also be collected and reprocessed in order to make full use of enriched materials. Given the necessity for the constant reprocessing of materials in such a production chain, it is highly desirable that all the production steps described above (a-e) be available in one location. This will greatly simplify the logistics, efficiency and yield of the processing. Given the desire to limit the activation, these facilities should be underground.

The development of such a facility is a long-term effort, and one that will likely be necessary for the success of a 1-ton scale double beta decay experiment (like GERDA Phase III). The development of crystal growing capabilities outside of industry is necessary already today. Having the Ge purification, crystal growth and detector fabrication together in an underground laboratory will speed up production, make it more efficient, and minimize cosmogenic activation

For GERDA Phase II, discussions were held with the Belgian Ge-crystal producer Umicore on the possibilities of placing some of their equipment in the nearby underground laboratory HADES. Although this in principle would be possible the cost for such an undertaking were considered too high for the reasons of (i) HADES has limited space and would require extra galleries for the safe handling of processes involving hydrogen gas (ii) Umicore needed in principle be compensated for the production loss during the time the equipment and their staff were in HADES.

Table 14. Overview on the main features of an underground germanium production facility. The cost estimate are only rough guideline values.

Item		Estimations	
		Size (m^2)	Cost (kEuro)
Germanium Laboratory construction (excluding underground excavation)			1000
Estimated space requirement		**300**	
	Chemical Laboratory	75	
	General testing facility	25	
	Reduction furnace, zone refiners	60	
	Clean room for crystal pulling	25	
	Crystal grinding & mechanics	15	
	Crystal etching	15	
	Clean room for implantation	60	
	Detector contacting & testing	25	
Equipping testing laboratory			500
Equipping Ge-purification			1000
	Main items: reduction furnace; several zone refiners; $GeCl_4$ distillation		
Equipping Ge-crystal growing			2000
	Main items: 2 Czochralsky pullers (1500 k€)		
Equipping Ge-detector fabrication			2000
	Main items: clean room (~500 k€); ion implantation (~200 k€)		
Laboratory operational costs			200 / year
Consumables: e.g. graphite boats, crucibles, pure gases			200 / year
Staff			
	2 scientists + 4 technicians for purification / crystal growth		400 / year
	2 scientists + 1 technician for detector fabrication		250 / year

Table 15. Summery of estimates of costs and time for various HPGe-detector production steps and recommendations.

Production step	Installation cost	Extra cost for underground operation**	Time to carry out this step	Opinion*; number of labs.
Enrichment/depletion	Very high	High	Several weeks	0
Chemical purification using $GeCl_4$	Medium	High		1
Reduction of germanium	High	High	A few days	1
Zone refinement	High	High	Several days	1
Czochralski growth***	High	High	1-2 days	1
Crystal control and characterisation	Low	None	One hour	1
Mechanical machining of crystal	Low	Low	A few hours	1
Li-diffusion	Medium	Low	A few hours	1
Implantation	Medium	Low	A few hours	1
Forming of contact structures	Medium	Low	A few hours	1
Assembling the HPGe-detector***	High	None	A few hours	Some
Pumping and testing of HPGe-detector	Low	None	A few days	Many

*Opinion relates to the authors' recommendation of what production step to place underground and where. "0" Not recommended; "1" Yes in one lab in Europe; "Some" Yes, in a few of the big underground laboratories; "Many" Yes in many underground laboratories, also smaller ones.
** In comparison to an above ground installation. Not relating to duplication of work above ground and underground. In principle all extra costs are safety related.
***Clean room necessary

5. Discussion

It is clear that there is a scientific need for having access to detector grade germanium that is produced underground. There is subsequently also a scientific need for HPGe-detectors that have been produced underground. The cost for implementation of such measures are relatively high as sketched in Tables 14 and 15. The costs referred to in Table 15 would be in the following orders of magnitude: (i) low (~ 10 k€), (ii) medium ~100 k€, (iii) high ~1 M€ (iv) very high ~10 M€.

Table 15 also gives the opinion of the authors regarding the realistic possibilities of implementing a certain production step underground (rightmost column). The costs for underground installation of the production steps should be compared to the upcoming need for such products and the importance of the measurement results obtained using the germanium that was produced underground. From Figure 7 one can estimate the upcoming need for detectors for underground gamma spectrometry within EU to 3 per year having in mind that several new labs are starting up and existing ones are expanding and new applications have arisen. It is likely that neighbouring countries like Russia, Ukraine, Turkey etc. will develop a need to 1 detector per year. This need is clearly too small for any action to be taken towards an underground Ge production facility. It is also clear that for these detectors it is not absolutely necessary with the extra benefits from underground production although they would be welcome, were they present. The driving force for underground production clearly

comes from double beta decay of Ge-76 and astroparticle physics. The GERDA experiment expects that in Phase II some 14 crystals are needed. A likely time for this would be during 2009 or 2010. The American double beta decay experiment MAJORANA could also benefit from the underground production facility provided the transport (without harmful cosmogenic activation) to USA could be arranged (submarine?). It is perhaps more likely that in the new National US underground laboratory that is being planned (DUSEL – Deep Underground Science and Engineering Laboratory) there will be space available for certain production steps. It is, however, clear that the GERDA and MAJORANA collaborations should keep in touch regarding these issues and maybe agree on a common place for underground production of germanium. Contacts towards this direction have already been taken since representatives from two collaborations frequently attend each other's collaboration meetings. In addition, the discussions for a single site with a kilo ton scale experiment - have begun.

Table 16 gives rough estimates on what the needs of potential users of germanium that was produced underground would be. Any user outside the Ge-76 double beta decay community would seriously consider if the extra cost would be necessary to achieve the background that is aimed at. An additional benefit from GERDA Phase II and III is that since germanium enriched in Ge-76 is necessary, there will be a relatively large stock of germanium depleted in Ge-76 available. For experiments not interested in the double beta decay of Ge-76 it is a benefit that this isotope is removed since it actually contributes to the background by the 2 neutrino double beta decay. This background would of course only be important for very long measurement times in deep underground laboratories.

Table 16. Rough estimates on the needs of potential users benefiting from underground produced germanium crystals.

	Crystals per year	**Total number of crystals**	**Importance of underground production**
For underground gamma spectrometry in EU	3	n.a.	Low
For underground gamma spectrometry in countries nearby the EU	1	n.a.	Low
For underground gamma spectrometry in countries far from the EU	2	n.a.	Low
Dark matter	?	?	Low
GERDA phase II		14	High
GERDA Phase III + MAJORANA	50 (for 5 years)	250	High

6. Conclusions

6.1. ILIAS-JRA1 (LBT-DUSL) specific conclusions

- For the strive towards a kilo ton size experiment of the zero neutrino double beta decay in Ge-76, it would be a great improvement if there was possibility of obtaining germanium crystals produced underground.
- The main concerns for underground production are related to safety issues, mainly with respect to the use of hydrogen gas. This calls for big and expensive safety measures
- The production step most suitable to be placed underground is the last step in the crystal manufacturing process, the Czochralski pulling. This step introduces such a great reduction of non-germanium impurities that it fulfils the requirements for GERDA.
- It will take several years to build up the know-how and competence to be able to pull big detector grade crystals of acceptable quality.
- For the deep European underground experiments and measurement facilities, it would be beneficial to have access to underground laboratories for production, refurbishment and maintenance of germanium detectors.
- The main problems with germanium detector production relates to building up know-how and competence as the equipment and safety related issues are not as expensive and complex as for crystal production.

6.2. ILIAS-JRA2 (IDEA) specific conclusions

- The GERDA double beta decay experiment solved its urgent needs for Phase I using optimisation of logistics.
- The crystals for GERDA Phase II can be carried out using a similar scheme of optimised logistics as for Phase I with the addition of the intermediate underground storage during the crystal production.
- For GERDA Phase III it is very important with the possibility of underground pulling of crystals and underground production of the Ge-detectors.
- GERDA has started discussions with MAJORANA regarding optimisation of resources and scientific studies in preparation of a kilo-ton double beta decay experiment.

7. References

Aalseth C E and the Majorana Collaboration, 2005, Nucl. Phys. B. (Proc. Suppl) 138 217-220.

Aalseth C E and the IGEX collaboration, 2002, Phys. Rev. D65.

E.I Abbakumov et al., 1989, Atomnaya Energia 67(4) p. 255 (in Russian).

Avignone F T et al., 1992, "Theoretical and experimental investigation of cosmogenic radioisotope production in germanium", Nucl. Phys. B (Proc. Suppl.) 28A p. 280-285.

Barabanov I et al., 2006, "Cosmogenic activation of germanium and its reduction for low background experiments" Nucl. Instr. Meth. B251, p. 115-120.

the BOREXINO collaboration, 2002, Astropart. Phys. 18, p. 1-25.

V.D. Borisevich et al., 2000, Nucl. Instr. Meth. A450, p. 515-521.

S. Cebrian, 2004, Rev. Real Academia de Ciencias. Zaragoza 59 p. 7-46.

S. Cebrian, 2005, "Cosmogenic activation in germanium Double Beta Decay experiments", presentation at TAUP05 conference.

Caryacademy, 2007, (accessed November 2007) http://web1.caryacademy.org/chemistry/rushin/StudentProjects/ElementWebSites/germanium/historical_background.htm.

Cazala C, Reyss J L, Decossas J L and Royer A, 2003, Environ. Sci. Technol. 37, p. 4990-4993.

Chapellier and the EDELWEISS Collaboration, 2003, Physica B: Condensed Matter Volumes 329-333, Part 2, p. 1592-1593.

S. Charalambus S, 1971, Nucl. Phys. A166, p.145-161.

Claeys C and Simoen E (Eds.), 2007, "Germanium-based Technologies", Elsevier, Oxford, UK.

Cocconi G, Cocconi-Tongiorgi V, 1951, Phys. Rev 84, p.29.

Dementyev A V, Sobolevsky N M, 1999, "SHIELD – universal Monte Carlo hadron transport code: scope and application", Radiation Measurements 30, p. 553.

DDEP, 2007, http://www.nucleide.org/DDEP_WG/DDEPdata.htm, (accessed, November 22, 2007).

Freck D W and Wakefield J 1962 Nature, p. 4816 669/

Gorshkov G V et al., 1966, "Estestveny fon atmosfery y zemnoy kory", Atomizdat. M. (in Russian).

Gurriaran R, Barker E, Bouisset P, Cagnat X and Ferguson C, 2004, Nucl. Instr. Meth. A524, p. 264-272.

Hall R N and Soltys T J, 1971, IEEE Trans. Nucl.. Sci. NS-18, p. 160-165.

Horuguchi T et al., 1983, Int. J. Appl. Rad. Isot. 34, p. 1531.

Haller E E, Hansen W and Goulding F S, 1981, "Physics of ultra-pure germanium Advances in Physics 30(1), p. 93-138.

Hayakawa S, 1969, "Cosmic Ray Physics, nuclear aspect", J. Willey & sons.

Heusser G, 1996, Ann. Rev. of Nucl. and Part. Sci. 45, p. 543-590.

Hult M, 2007, “Low-level gamma-ray spectrometry using Ge-detectors” Metrologia 44 p.S87-S94. Erratum at Metrologia 44, p. 425.

Hult M, Preusse W, Gasparro J and Köhler M, 2006, Acta Chim. Slov. 53, p. 1-7.

Hult M, Gasparro J, 2007 Int. Rep. on FoM, in manuscript.

ILIAS, 2007, http://www-ilias.cea.fr/scripts/home/publigen/content/templates/show.asp?P=293&L=EN&ITEMID=2, accessed December 6, 2007.

Klapdor-Kleingrothaus H V et al., 2001, European Phys. J. A 12(2), p. 147-154.

Kornoukhov V N, 2003a, "Man made neutrinos sources & enriched isotopes", LowNu2003, 4th International Workshop on Low Energy and Solar Neutrinos May 19-21, 2003 (http://www.apc.univ-paris7.fr/Conference/LowNu03).

Kornoukhov V N, 2003b, "Stable Isotopes Production in Russia", Report for IV International Neutrino Conference on Non-Accelerated New Physics, Dubna, Russia, June 23-28, 2003 (http://nanp.dubna.ru/2003).

Kornoukhov V N, 2005, "New requirements on enriched isotopes for experiments studying neutrinoless double β-decay (GERDA experiment)", Proc. of 10th Intern. Scientific Conference «Physical-Chemical Processes on Selection of Atoms and Molecules». October 3-7, 2005, Zvenigorod.Edited by Dr.Yury A.Kolesnikov Moscow, Atominform, Moscow region, Troitsk, SRC RF TRINITI, pp. 148-153.

Laubenstein M, Hult M, Gasparro J, Heusser G, Köhler M, Neumaier S, Arnold D, Povinec P, Reyss J L, Schwaiger M and Theodorsson P, 2004, Appl. Radiat. Isotopes 61(2-3) p. 167-172.

E.Nikitina, Report for International Conference TAUP 2007, Sendai, Japan (http://www.awa.tohoku.ac.jp/taup2007/).

NIST, 2006, http://physics.nist.gov/cgi-bin/cuu/Value?na, accessed April 30, 2006.

NIST, 2007, http://physics.nist.gov/PhysRefData/Compositions/index.html, accessed November 22, 2007.

Rosman K J R, Taylor P D P, 1998, "Isotopic compositions of the elements 1997", Pure & Appl. Chern., 70(1), p. 217-235.

Sangsingkeow P, Berry K D, Dumas E J, Raudorf T W and Underwood T A, 2003, Nucl. Instr. Meth. A505, p. 183–186.

S. Schönert and the GERDA collaboration, 2005, Nucl. Phys. B145, p. 142-245.

Seilnacht, 2007, http://www.seilnacht.com/Lexikon/32German.htm, accessed November 2007.

Shubin A N, Gilev A N, Kononov D B, Mis'kov A A, Nikitina E A, Skorynin G M, Barabanov I R, Bezrukov L B, Belogurov S G, Denisov A N, Kornoukhov V N, Sobolevsky N M, Altmann M and Caldwell A, 2006, "New requirements on enriched isotopes for study of neutrinoless double beta decay (experiment GERDA), Atomic Energy 101(2).

Sobolevsky N M, 2004, "Computer Study of Interactions of Hadrons and Nuclei with Complex Media", Dr. hab. Thesis, Institute for Nuclear Research RAS, Moscow, 2004 (in Russian).

US Geological Survey, 2007, http://minerals.usgs.gov/minerals/pubs/commodity/germanium/ accessed December 5, 2007.

Vanhellemon J, Simoen E, Romandic I and Theuwis A, 2007, Chapter 2 in Claeys and Simoen (Eds.), "Germanium-based Technologies", Elsevier, Oxford, UK.

Vasenko A A et al., 1989, Prybory y Technika Experimenta, 2, p. 56-63 (in Russian).

Vasenko A A et al., 1990, Modern Physics Letters A5(17), P. 1299-1306.

Winkler H C A, 1886, J. Prak. Chemie 34, p.177.

WWW Table of Isotopes, 2007, http://nucleardata.nuclear.lu.se/nucleardata/toi/ (Accessed November 22, 2007).

Wyttenbach A et al., 1978, Nucl. Phys. A294, p. 278.

Yu G and Grigoriev, 2003a, Report for Grand meeting in honor of 65th Anniversary of Kurchatov Institute, 11 of April 2003.

Yu G and Grigoriev, 2003b, "Production of stable isotope in RCC Kurchatov Institute", Report for IV International Neutrino Conference on Non-Accelerated New Physics, Dubna, Russia, June 23-28, 2003 (http://nanp.dubna.ru/2003).

Ziegler J F and Sellschop J P F, 1981, Nucl. Instrum. Meth. 191, p. 419.

Ziegler J F, 1996, IBM J. res. develop. 40, p. 19.

European Commission

EUR 23237 EN– Joint Research Centre – Institute for Reference Materials and Measurements
Title: On the Underground Production of High Purity Germanium Detectors
Author(s): Mikael Hult, Sergey Belogurov, Allan Caldwell, Josef Janicsko, Vasiliy Kornoukhov, Stefan Schönert
Luxembourg: Office for Official Publications of the European Communities
2008 – 44 pp. – 21 x 29.7 cm
EUR – Scientific and Technical Research series – ISSN 1018-5593
ISBN 978-92-79-08276-4
DOI 10.2787/13569

Abstract
Detectors based on high purity germanium (HPGe) are used in numerous deep underground experiments world-wide aiming at detecting rare events like double beta decay and interactions of dark matter. These detectors require the lowest possible background. A significant part of the background is due to radionuclides produced by cosmic-ray interactions with the germanium crystal. This report gives quantitative data on this activation and discusses the possible solutions. The first solution is to optimise the logistics during the crystal and detector fabrication so that the germanium spends a minimum time above ground. The second solution is to implement one or several (up to 12) production steps underground. The report also makes estimates on the future needs for germanium produced underground and the costs involved.

The mission of the JRC is to provide customer-driven scientific and technical support for the conception, development, implementation and monitoring of EU policies. As a service of the European Commission, the JRC functions as a reference centre of science and technology for the Union. Close to the policy-making process, it serves the common interest of the Member States, while being independent of special interests, whether private or national.

LA-NA-23237-EN-C

ISBN 978-92-79-08276-4