

To the Graduate Council:

I am submitting herewith a dissertation written by Mohammad Ibrahim Mirza entitled “Mitigation Strategies of ^{42}K Background and Development of Radiopure Materials for the LEGEND-1000 $0\nu\beta\beta$ Decay Experiment.” I have examined the final paper copy of this dissertation for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Physics.

Yuri Efremenko, Major Professor

We have read this dissertation
and recommend its acceptance:

Yuri Efremenko

Kate Jones

Eric Lukosi

Robert Grzywacz

Accepted for the Council:

Marieke Van Puymbroeck

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(Original signatures are on file with official student records.)

Mitigation Strategies of ^{42}K Background
and Development of Radiopure Materials
for the LEGEND-1000 $0\nu\beta\beta$ Decay
Experiment

A Dissertation Presented for the
Doctor of Philosophy
Degree
The University of Tennessee, Knoxville

Mohammad Ibrahim Mirza

December 2025

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To my family

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“Everything comes to him who knows how to wait.”

— Wolfgang Pauli

Abstract

Neutrinoless double-beta ($0\nu\beta\beta$) decay, if discovered, provides clear evidence of the existence of Majorana neutrinos and constrain or measure effective Majorana neutrino mass. Total lepton number conservation is violated in this decay, leading to the interpretation of matter-antimatter asymmetry in the Universe. Due to the very low probability of such a process, extremely low background conditions are required to observe the $0\nu\beta\beta$ decay signal (monoenergetic peak at 2039 keV for ^{76}Ge). The next-generation LEGEND-1000 experiment aims to observe $0\nu\beta\beta$ decay with 1000 kg of high-purity enriched germanium ^{76}Ge detectors submerged in liquid argon located deep underground. ^{42}K is one of the radioactive isotopes produced in natural liquid argon; its β -decay overlaps the $0\nu\beta\beta$ decay signal region and causes a significant background. Monte Carlo simulations are performed to study the impact of ^{42}K background on the germanium detector. The encapsulation of germanium detectors with low-background (radiopure) material would be an efficient and effective way to suppress the ^{42}K background. In collaboration with the LEGEND group at ORNL, my work focuses on the R&D of the 3D-printing of low-background materials that can be used to encapsulate germanium detectors and also build radiopure detector components. We developed procedures and equipment to build ultra-clean 3D-printable materials. We also developed a vacuum system to measure outgassing rates of the 3D-printed materials. We performed the Monte Carlo simulation for the gamma-ray spectrometer and performed radioassay. Radiopurity results of 3D-printed samples are presented.

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Nomenclature

$0\nu\beta\beta$	Neutrinoless double-beta decay
$2\nu\beta\beta$	Two neutrino double-beta decay
2039 keV	$Q_{\beta\beta}$ value of ^{76}Ge
3D-printing	Three-dimensional printing, additive manufacturing
^{42}Ar	Argon-42 isotope
^{42}K	Potassium-42 isotope
^{76}Ge	Germanium-76 isotope
B.R.	Branching ratio
Background index	Number of counts per keV per kg per year
DI water	Deionized water
Exposure	Mass of isotope times live time
GEANT4	GEometry ANd Tracking
HPGe	High-purity germanium detector
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICPC	Inverted Coaxial Point-Contact
LEGEND-1000	Large Enriched Germanium Experiment for Neutrinoless $\beta\beta$ Decay of 1000 kg ^{76}Ge

LEGEND-200	Large Enriched Germanium Experiment for Neutrinoless $\beta\beta$ Decay of 200 kg ^{76}Ge
$m_{\beta\beta}$	Effective Majorana neutrino mass
ORNL	Oak Ridge National Laboratory
PEN	Polyethylene napthalate
$Q_{\beta\beta}$	Q-value of the neutrinoless double-beta decay
R&D	Research and development
RGA	Residual gas analyzer
ROI	Region of interest
SLA	Stereolithography
SM	Standard Model of particle physics
$T_{1/2}^{0\nu}$	Half-life of neutrinoless double-beta decay

Chapter 1

Introduction

The Standard Model (SM) of particle physics successfully describes elementary particles and their interactions without assigning mass to neutrinos. Observation of the phenomenon of neutrino oscillation confirms the existence of *massive neutrinos*. Neutrinos are the most abundant fermions in the Universe, created during the Big Bang. In addition, they can originate from a wide range of physical systems, including supernova, active galactic nuclei, gamma-ray bursts, the Sun, cosmic-ray interaction, the upper Earth atmosphere, nuclear reactors, particle accelerators, nuclear decay, the decay of other particles, etc. Recently the KM3NeT experiment reported the observation of a neutrino of the ever-highest energy, ~ 220 PeV (10^{15} eV) [1]. This points toward the unexplored source of neutrinos.

In the early Universe, matter and antimatter were created in equal amounts. In the present Universe, matter dominates over antimatter. The Standard Model of cosmology cannot explain the observed asymmetry between matter and antimatter. Recently the LHCb experiment at CERN reported the first observation of charge-parity symmetry violation in baryon decays [2]. These observations indicate the Standard Model is definitely incomplete and requires *new physics* beyond Standard Model.

Another long-standing problem in particle physics is whether neutrinos are Dirac fermions (neutrinos and antineutrinos are different) or Majorana fermions (neutrinos and antineutrinos are the same). Searching for an extremely rare nuclear decay, neutrinoless double-double ($0\nu\beta\beta$) decay (*yet to be observed*), provides an opportunity to explore the fundamental nature of neutrinos, Dirac or Majorana, and provide constrain or measure effective Majorana

neutrino mass. $0\nu\beta\beta$ decay is not allowed in the SM, as it violates total lepton number conservation and therefore provides an avenue to interpret the matter-antimatter asymmetry in the Universe.

Various experiments of the previous generation, e.g., GERDA, MAJORANA, EXO-200, KamLAND-Zen, and CUORE, etc., completed their search for $0\nu\beta\beta$ decay with *null* results. LEGEND-200 is one of the most advanced $0\nu\beta\beta$ decay experiment currently taking data, with the first result published at [3]. Various next-generation experiments, e.g., LEGEND-1000, nEXO, CUPID, KamLAND-Zen2, etc., are being planned.

The next phase of LEGEND (Large Enriched Germanium Experiment for Neutrinoless $\beta\beta$ Decay) is LEGEND-1000 with enriched ^{76}Ge isotope (1000 kg). The experiment aims to achieve very low-background, which is extremely crucial for the observation of $0\nu\beta\beta$ decay. One of the main backgrounds in LEGEND is ^{42}K , a radioactive isotope; its decay hides the $0\nu\beta\beta$ decay signal. To study this background, Monte Carlo simulations are performed. Encapsulation of the germanium detector is under consideration to mitigate the ^{42}K background.

Encapsulation material is required to be low or better free from any radioactivity. 3D-printing techniques are developed to build ultra-pure materials. Outgassing techniques are developed to measure contamination in the materials. Radiopurity measurements are obtained in both ICP-MS and gamma-ray spectroscopy.

This dissertation describes R&D efforts to mitigate ^{42}K background. Monte Carlo simulations on the ^{42}K background are provided, and radiopure materials are developed to encapsulate germanium detectors. This study will be important in achieving the LEGEND background goal and searching for neutrinoless double-beta decay with the LEGEND-1000 experiment.

Outline of the dissertation

Chapter 2: Neutrinoless Double-Beta Decay—describes the introduction of $0\nu\beta\beta$ decay, and the history of germanium-based $0\nu\beta\beta$ decay experiments is discussed. The current best limit on half-life and effective Majorana neutrino mass are mentioned.

Chapter 3: LEGEND Experiment—discusses the brief introduction of GERDA and MAJORANA experiments, and their final results are discussed. The currently operational LEGEND-200 experiment and recent results are discussed. Next-generation LEGEND-1000 experiment are discussed.

Chapter 4: Mitigation Strategies of Argon-42 Background—discusses the cosmogenic impurities ($^{42}\text{Ar}/^{42}\text{K}$) present in atmospheric-sourced liquid argon. Underground-sourced liquid argon with depleted ^{42}K is discussed. Methods for mitigation of the ^{42}Ar background are listed. Simulation results on the effect of germanium dead layer thickness on ^{42}K background are described. Simulation of PEN encapsulation to a germanium detector is described.

Chapter 5: Monte Carlo Simulation for Various Scenarios of Detector Encapsulation—describes the Monte Carlo simulation of the ^{42}K background on the ICPC detector with three scenarios of germanium detector dead layer thickness and liquid argon thickness, and simulation results are summarized.

Chapter 6: R&D of 3D-Printable Low-Background Materials—describes the 3D-printing procedure of radiopure materials; liquid resin, degassing setup, cleanroom, stereolithography 3D-printing, and curing are described. 3D-printing of LEGEND-1000 parts is discussed.

Chapter 7: Outgassing of 3D-Printed Low-Background Materials—describes the outgassing rates and apparatus used to measure outgassing rate. The residual gas analyzer and its functionality are described. Outgassing rates of 3D-printed materials are described, and values are summarized. Outgassing rates of other materials used in the liquid argon cryostat are listed.

Chapter 8: Radioassay of 3D-Printed Low-Background Materials—describes the radioassay techniques with 3D-printed samples. Inductively coupled plasma mass spectrometry results of 3D-printed samples are discussed. Gamma-ray spectrometry results

of 3D-printed samples are discussed. Monte Carlo simulations of a gamma-ray spectrometer are performed, and results are described.

Chapter 9: Conclusion—discusses the prospects of the search $0\nu\beta\beta$ decay with the LEGEND-1000 experiment. Key results on the Monte Carlo simulations for ^{42}K background are summarized. The development of radiopure materials, outgassing rates, mass spectroscopy, gamma-ray spectroscopy, and radioassay simulations are summarized.

Chapter 2

Neutrinoless Double-Beta Decay

The Standard Model (SM) of particle physics $SU(3)_C \times SU(2)_L \times U(1)_Y$ is a relativistic quantum field theory of quarks, leptons and gauge bosons describing strong and electroweak interactions. SM successfully predicted a wide range of particle processes and reactions. The last missing piece of the SM was the Higgs boson, finally discovered at CERN in 2012. SM is well established and no large anomalies are seen.

However, SM cannot explain various observed phenomena including neutrino oscillation, neutrino mass, baryon asymmetry, dark matter, dark energy, etc. It is evident that the SM is incomplete and therefore, theories beyond Standard Model (BSM) must be needed.

Neutrinos are the most abundant matter particle in the Universe. It was first proposed by Wolfgang Pauli in 1930, and the term “neutrino” was coined by Enrico Fermi in 1934. Neutrinos were first detected in 1956 by Frederick Reines and Clyde Cowan. In the SM, electrically charged fermions are both left- and right-handed and obtain their masses from the Yukawa interaction between the Higgs field and fermions. Goldhaber et al. experiment showed that neutrinos are only left-handed [4]. Since right-handed neutrinos do not exist, neutrinos receive no mass term from Yukawa interaction, and therefore left-handed neutrinos remain massless in the SM. The phenomenon of neutrino flavor oscillation ($\nu_\alpha \rightarrow \nu_\beta$, where $\alpha, \beta = e, \mu, \tau$) [5], first observed by Super-Kamiokande [6] and Sudbury Neutrino Observatory [7] leads to the indication of a non-zero value of the neutrino mass. The Standard Model (SM) of elementary particles [8–10] is unable to interpret this anomaly since the SM presumes massless neutrinos. Massive neutrinos are the strongest indication of

the presence of the physics beyond Standard Model (BSM). Due to the quantum mechanical nature of the neutrino, three neutrino flavor eigenstates ν_e, ν_μ, ν_τ should be expressed as a linear combination of neutrino mass eigenstates ν_1, ν_2, ν_3 with neutrino mass values: m_1, m_2, m_3 , as in equation 2.1. The absolute neutrino masses are not yet known, which leads to the two possible scenarios of neutrino mass orderings or hierarchy as normal ($m_3 > m_2 > m_1$) and inverted ($m_2 > m_1 > m_3$) ordering.

$$\begin{pmatrix} \nu_e \\ \nu_\mu \\ \nu_\tau \end{pmatrix} = \begin{pmatrix} U_{e1} & U_{e2} & U_{e3} \\ U_{\mu1} & U_{\mu2} & U_{\mu3} \\ U_{\tau1} & U_{\tau2} & U_{\tau3} \end{pmatrix} \begin{pmatrix} \nu_1 \\ \nu_2 \\ \nu_3 \end{pmatrix} \quad (2.1)$$

The two-flavor neutrino oscillation probability is related to the neutrino mass-squared difference ($\Delta m^2 = m_2^2 - m_1^2$) as

$$P_{e\mu} = \sin^2(2\theta) \cdot \sin^2\left(\frac{\Delta m^2 L}{4E}\right). \quad (2.2)$$

Solar, atmospheric, reactor, and accelerator based neutrino experiments measure, the solar neutrino mass squared difference: $\Delta m_{21}^2 = (7.53 \pm 0.18) \times 10^{-5} \text{ eV}^2$, atmospheric neutrino mass-squared difference: $\Delta m_{32}^2 = (2.455 \pm 0.028) \times 10^{-3} \text{ eV}^2$ (normal ordering), $\Delta m_{32}^2 = (-2.529 \pm 0.029) \times 10^{-3} \text{ eV}^2$ (inverted ordering), solar neutrino mixing angle: $\sin^2\theta_{12} = 0.307 \pm 0.013$, atmospheric neutrino mixing angle: $\sin^2\theta_{23} = 0.553_{-0.024}^{+0.016}$ (inverted order), $\sin^2\theta_{23} = 0.558_{-0.021}^{+0.015}$ (normal order), and reactor neutrino mixing angle: $\sin^2\theta_{13} = (2.19 \pm 0.07) \times 10^{-2}$ [11]. Current neutrino oscillation experiments cannot resolve the correct neutrino ordering, and therefore the next generation experiments: JUNO, Hyper-Kamiokande, and DUNE are planned to make this determination. Neutrino oscillation experiments cannot measure absolute neutrino masses, therefore different techniques are required. From the measurement of β -decay kinematics, the effective electron neutrino mass (m_{ν_e}) can be measured. By measuring tritium β -decay the KATRIN experiment set an upper limit on $m_{\nu_e} < 0.45 \text{ eV}$ [12]. Cosmological measurements provide the sum of neutrino masses ($\Sigma m_\nu = m_1 + m_2 + m_3$). Observations data within the Λ CDM (Lambda Cold Dark Matter) model of cosmology provides the constraints on the upper limit of the

$\Sigma m_\nu < 0.072$ eV [13]. However, Λ CDM is not a direct mass measurement and is subject to interpretation. The search for a yet unobserved nuclear process; neutrinoless double-beta decay ($0\nu\beta\beta$) provides a constraint or measure of the effective Majorana neutrino mass ($m_{\beta\beta}$). Extraction of mass values will need a further much larger detector [14]. All quarks and leptons except neutrinos are electrically charged i.e., particle and its corresponding anti-particle are distinguishable—Dirac fermions. Since the neutrino experiences only the weak interaction and does not have electrical charge, it is therefore yet unknown whether neutrinos and anti-neutrinos are identical or different particles.

$$\begin{aligned} \text{Dirac neutrino: } \nu &\neq \bar{\nu} \quad (\text{distinguishable}) \\ \text{Majorana neutrino: } \nu &= \bar{\nu} \quad (\text{indistinguishable}) \end{aligned} \tag{2.3}$$

Neutrinos are the potential candidate for the Majorana fermions. Neutrino oscillation experiments cannot differentiate between Dirac and Majorana neutrino. Cosmological measurements are not too sensitive to Dirac or Majorana neutrino. β -decay experiments are independent of Dirac or Majorana neutrino. The only practical way to prove neutrinos are Majorana is to search for neutrinoless double-beta decay.

2.1 Introduction to Neutrinoless Double-Beta Decay

Two neutrino double beta ($2\nu\beta\beta$) decay occurs in even-even nuclei when single β -decay is energetically forbidden, as shown in Figure 2.1. It is a second order weak process observed in many even-even nuclei. In $2\nu\beta\beta$ decay, two neutrons simultaneously transform into two protons with the emission of two e^- and two $\bar{\nu}_e$. This decay conserves the total lepton number and is allowed in the SM. $2\nu\beta\beta$ decay cannot distinguish Dirac and Majorana neutrino.

Neutrinoless double-beta ($0\nu\beta\beta$) decay is a hypothetical nuclear process in which parent nuclei simultaneously transform into daughter nuclei via the emission of only two electrons. This decay violates lepton number conservation and is therefore forbidden in the Standard Model. The observation of $0\nu\beta\beta$ decay could explain the baryonic asymmetry of the Universe. Baryogenesis explains the observed baryon-to-photon ratio $\eta = 6.04 \times 10^{-10}$ in the Universe [11]. Baryon asymmetry may be generated in the early Universe by satisfying

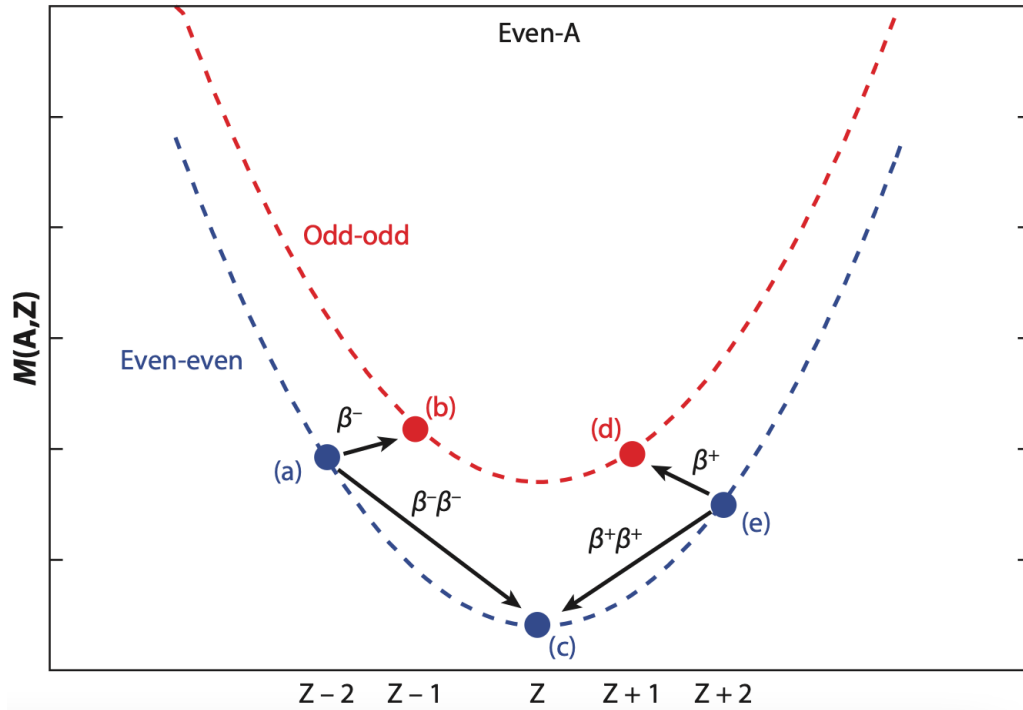


Figure 2.1: Mass parabola of isobars with even mass number (A). β -decay from a to b and e to d is energetically forbidden and $2\nu\beta\beta$ decay a to c and e to c is energetically allowed. Credit [15].

the three Sakharov conditions: (i) violation of baryon number B, (ii) violation of C (charge conjugation) and CP (charge conjugation and parity), and (iii) departure from thermal equilibrium. During $0\nu\beta\beta$ decay, two neutrons each emit a W boson; these W bosons exchange Majorana neutrinos and produce two electrons, as shown in Figure 2.2.

$$(A, Z) \rightarrow (A, Z+2) + e^- + e^- \quad (2.4)$$

The half-life of $0\nu\beta\beta$ decay is finite when neutrinos are massive Majorana particles. In the framework of Majorana neutrino exchange the half-life ($T_{1/2}^{0\nu}$) of the decay is given by,

$$T_{1/2}^{0\nu} = \frac{m_e^2}{\mathcal{G}^{0\nu} g_A^4 |\mathcal{M}^{0\nu}|^2 m_{\beta\beta}^2} \quad (2.5)$$

Where $\mathcal{G}^{0\nu}$ is the phase-space integral, $g_A = 1.27$ is the axial-vector coupling constant, $\mathcal{M}^{0\nu}$ is the nuclear matrix element, $m_{\beta\beta}$ is the effective Majorana neutrino mass, and m_e is the rest mass of the electron.

Phase space factor $\mathcal{G}^{0\nu}$ represents the Columbic interaction between the two emergent electrons and the daughter nucleus during $0\nu\beta\beta$ decay. For ^{76}Ge , $\mathcal{G}^{0\nu} = (2.36 - 2.37) \times 10^{-15} \text{ yr}^{-1}$. The nuclear matrix element $\mathcal{M}^{0\nu}$, is a dimensionless quantity, which describes the second order weak transition from the initial to the final state of the nucleus in $0\nu\beta\beta$ decay. It cannot be measured directly in experiment and has only theoretically calculated values. $\mathcal{M}^{0\nu}$ contains four terms: Gamow-Teller (GT), Fermi (F), tensor (T), contact operator (cont). Nuclear matrix element for $0\nu\beta\beta$ decay is given by,

$$\mathcal{M}^{0\nu} = \mathcal{M}_{\text{GT}}^{0\nu} - (g_V/g_A)^2 \mathcal{M}_{\text{F}}^{0\nu} + \mathcal{M}_{\text{T}}^{0\nu} + (g_\nu^{\text{NN}} m_\pi^2 / g_A^2) \mathcal{M}_{\text{cont}}^{0\nu} \quad (2.6)$$

Where g_V , g_A and g_ν^{NN} are the vector, axial, and hadronic coupling constants respectively. Values of $\mathcal{M}^{0\nu}$ is nuclear model-dependent and ranges from 2.66 to 6.04 for ^{76}Ge [16–27]. This uncertainty in the nuclear matrix element calculations is the reason for the uncertainty in the extraction of effective Majorana neutrino mass limits.

The half-life of $0\nu\beta\beta$ decay calculated from experimentally measurable parameters is,

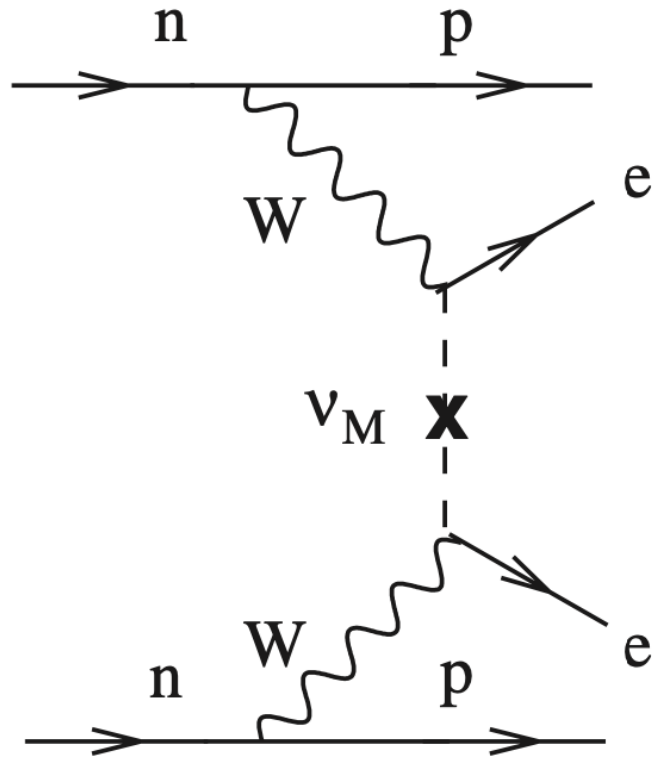


Figure 2.2: Feynman diagram of $0\nu\beta\beta$ decay, where ν_M is the light Majorana neutrino exchange and W is the virtual gauge boson. Credit [28].

$$T_{1/2}^{0\nu} \propto \begin{cases} a \cdot \varepsilon \cdot \sqrt{\frac{\mathcal{E}}{\mathcal{B} \cdot \Delta E}} & (\mathcal{B} \neq 0) \\ a \cdot \varepsilon \cdot \mathcal{E} & (\mathcal{B} = 0) \end{cases}, \quad (2.7)$$

where a is the isotopic abundance, ε is the efficiency of the detection of $0\nu\beta\beta$ signal, $\mathcal{B}$ is the background index, $\mathcal{E}$ is the exposure (= mass of the isotope $\times$ live time), and ΔE is the energy resolution of the detector. For experiments that are background free, $\mathcal{B} = 0$ and non-zero background $\mathcal{B} \neq 0$. Next-generation $0\nu\beta\beta$ decay experiments are planned for longer exposure $\mathcal{E}$ i.e. higher isotope mass and or longer run time, smaller energy resolution ΔE of the detector, and smaller background $\mathcal{B}$. Ultra-low background is extremely important for the search of $0\nu\beta\beta$ decay. Since backgrounds originate from various intrinsic and extrinsic processes, they could overlap the ROI (region of interest near $Q_{\beta\beta}$). Next-generation experiments are developing novel techniques for minimizing the backgrounds. This dissertation work is towards the effort of developing ultra-pure plastics for mitigating the backgrounds sourced from the ^{42}Ar and its daughter ^{42}K ; these are described in Chapter 3.

The effective Majorana neutrino mass ($m_{\beta\beta}$) can be extracted from the measurement of $T_{1/2}^{0\nu}$. $m_{\beta\beta}$ is defined as :

$$m_{\beta\beta} \equiv |U_{e1}^2 m_1 + U_{e2}^2 m_2 + U_{e3}^2 m_3| \quad (2.8)$$

where U_{e1} , U_{e2} , U_{e3} are components of neutrino mixing matrix, and m_1 , m_2 , m_3 are neutrino mass eigenstates. Values of $m_{\beta\beta}$ depends on neutrino mass hierarchy as shown in Figure 2.3.

2.2 History of Neutrinoless Double-Beta Decay

In 1930, Wolfgang Pauli proposed the existence of a new particle the “neutrino” during β -decay. Soon after, in 1935, Maria Goeppert Mayer predicted two neutrino double-beta decay [29]. In 1937, Ettore Majorana proposed that a neutrino could potentially be its own antiparticle (Majorana neutrino) [30]. In 1939, Wendall Fury proposed $0\nu\beta\beta$ by combining Mayer and Majorana ideas [31].

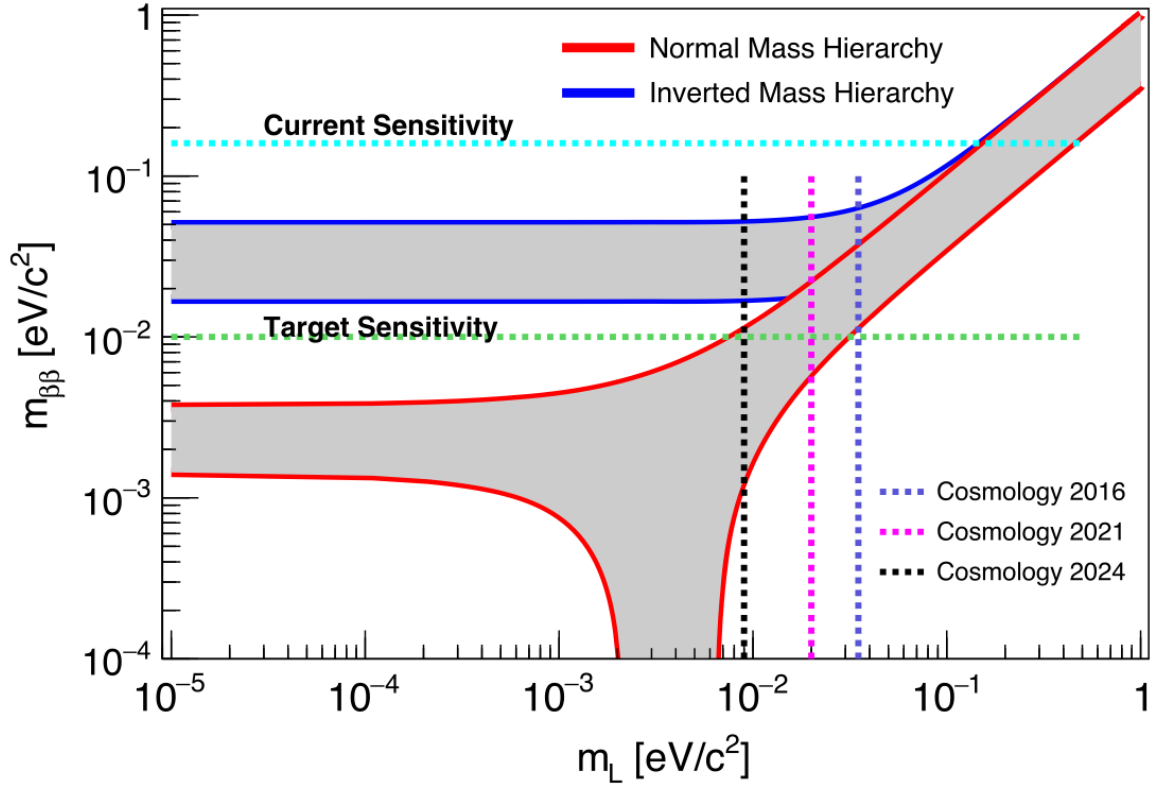


Figure 2.3: Effective Majorana neutrino mass $m_{\beta\beta}$ as a function of the lightest neutrino mass m_L , where the blue band shows inverted mass ordering and the red band shows normal mass ordering. Current sensitivity is achieved by the current generation experiments: KamLAND-Zen, GERDA, and CUORE and LEGEND-200. Target sensitivity will be achieved by next-generation experiments: LEGEND-1000, KamLAND-Zen2, nEXO, and CUPID. Credit [32].

In 1948, the first experiment to search for $2\nu\beta\beta$ decay started and the first observation of $2\nu\beta\beta$ decay in 1950. In 1967, a germanium-based $0\nu\beta\beta$ decay experiment was started. [33].

A highly sensitive counter experiment with ^{76}Ge started in 1973 [34].

In 1982, the Schechter-Valle theorem was proposed, which suggests that the observation of $0\nu\beta\beta$ decay will prove neutrinos are Majorana [35]. In 1986, Vogel and Zirnbauer approximately predicted the $2\nu\beta\beta$ decay rate within the quasiparticle random phase approximation (QRPA) after introducing the particle-particle interaction in nuclear potential.

In 1987, the Institute for Theoretical and Experimental Physics and the Yerevan Physics Institute (ITEP-YePI) experiment was performed 245 m underground at the Avan salt mine at Yerevan, Armenia. The ITEP-YePI was the first experiment that used enriched ^{76}Ge detectors (86% of ^{76}Ge) with a total mass of ~ 1.1 kg and measured the half-life of $0\nu\beta\beta$ decay $T_{1/2}^{0\nu} > 1.3 \times 10^{24}$ yr [36].

In 1991, the International Germanium Experiment (IGEX) was operated at three locations, Homestake gold mine, Canfranc Underground Laboratory and Baksan Neutrino Observatory. IGEX used 86% enriched ^{76}Ge detectors with a total mass of ~ 6.5 kg and measured the half-life of $0\nu\beta\beta$ decay to be $T_{1/2}^{0\nu} > 1.57 \times 10^{25}$ yr [37].

In 1990, the Heidelberg-Moscow experiment started operating at Gran Sasso National Laboratory. The experiment consists of a 11.5 kg coaxial-high-purity ^{76}Ge detectors enriched to 88% and measured the half-life of $0\nu\beta\beta$ decay to be $T_{1/2}^{0\nu} > 1.9 \times 10^{25}$ yr [38].

In 2004, part of the Heidelberg-Moscow experiment, Kleingrothaus, Krivosheina, Dietz, and Chkvorets, (KKDC) claimed evidence of $0\nu\beta\beta$ decay with enriched ^{76}Ge [39].

In 2012, the GERmanium Detector Array (GERDA) experiment started at Gran Sasso National Laboratory. GERDA consists of 44.2 kg enriched ($\sim 87\%$) ^{76}Ge detector. GERDA was the first experiment to operate in the “background-free-regime.” and set the limit on the half-life of $0\nu\beta\beta$ decay of ^{76}Ge , $T_{1/2}^{0\nu} > 1.8 \times 10^{26}$ yr [40].

In 2013, GERDA found no evidence of a $0\nu\beta\beta$ decay signal and refuted the KKDC claim [41].

The MAJORANA DEMONSTRATOR (MJD) was started in 2013 at Sanford Underground Research Facility. MJD consists of 40.4 kg germanium detectors (27.2 kg of $\sim 88\%$ enriched in ^{76}Ge and 13.2 kg of natural). MJD achieved the world-leading energy resolution of 2.52 keV (FWHM) at the 2039 keV for any enriched ^{76}Ge detector. MJD measured the half-life

of $0\nu\beta\beta$ decay of ^{76}Ge , $T_{1/2}^{0\nu} > 8.3 \times 10^{25}$ yr [42].

In 2023, the LEGEND-200 (LEGEND:Large Enriched Germanium Detector for Neutrinoless $\beta\beta$ Decay) experiment started at Gran Sasso National Laboratory. Currently LEGEND-200 consists of 142.2 kg enriched ^{76}Ge detector and currently in data taking phase. First results [3] establishes the best lower limit on $T_{1/2}^{0\nu} > 1.9 \times 10^{26}$ yr.

The history of germanium-based $0\nu\beta\beta$ decay experiments is listed in Table 2.1. The measured lower-limit on the $0\nu\beta\beta$ decay half-life of ^{76}Ge experiments during 55 years, along with the projections of LEGEND-200 and LEGEND-1000 goals are shown in Figure 2.4. In 2024, the KamLAND-Zen experiment set the most stringent lower limit on the half-life of ^{136}Xe , including any $0\nu\beta\beta$ isotope, $T_{1/2}^{0\nu} > 3.8 \times 10^{26}$ yr [43]. KamLAND-Zen finished data-taking in 2024 and rebuild for KamLAND-Zen2. Hence proposed next-generation of $0\nu\beta\beta$ decay experiments are designed to be sensitive for beyond KamLAND-Zen limit.

The history of half-life limits of a few $0\nu\beta\beta$ decay isotopes measured in different years are shown in Figure 2.5. Current experimental limits on half-life of $0\nu\beta\beta$ decay and corresponding limits on effective Majorana neutrino mass are listed in Table 2.2.

Next generation ton-scale experiment e.g., LEGEND-1000, nEXO, CUPID, KamLAND2-Zen, SNO+ are planned. The dependence of the limit of the half-life sensitivity on exposure for various levels on background at the region of interest for the LEGEND-1000 experiment is shown in Figure 2.6.

Table 2.1: Important events in history of germanium-based $0\nu\beta\beta$ decay search.

Year	Timeline
1967	First germanium-based experiment started
1987	IGEX experiment begin with enriched germanium
1990	Heidelberg-Moscow experiment started enriched germanium
2012	GERDA experiment started
2013	MAJORANA experiment started
2023	LEGEND-200 experiment started

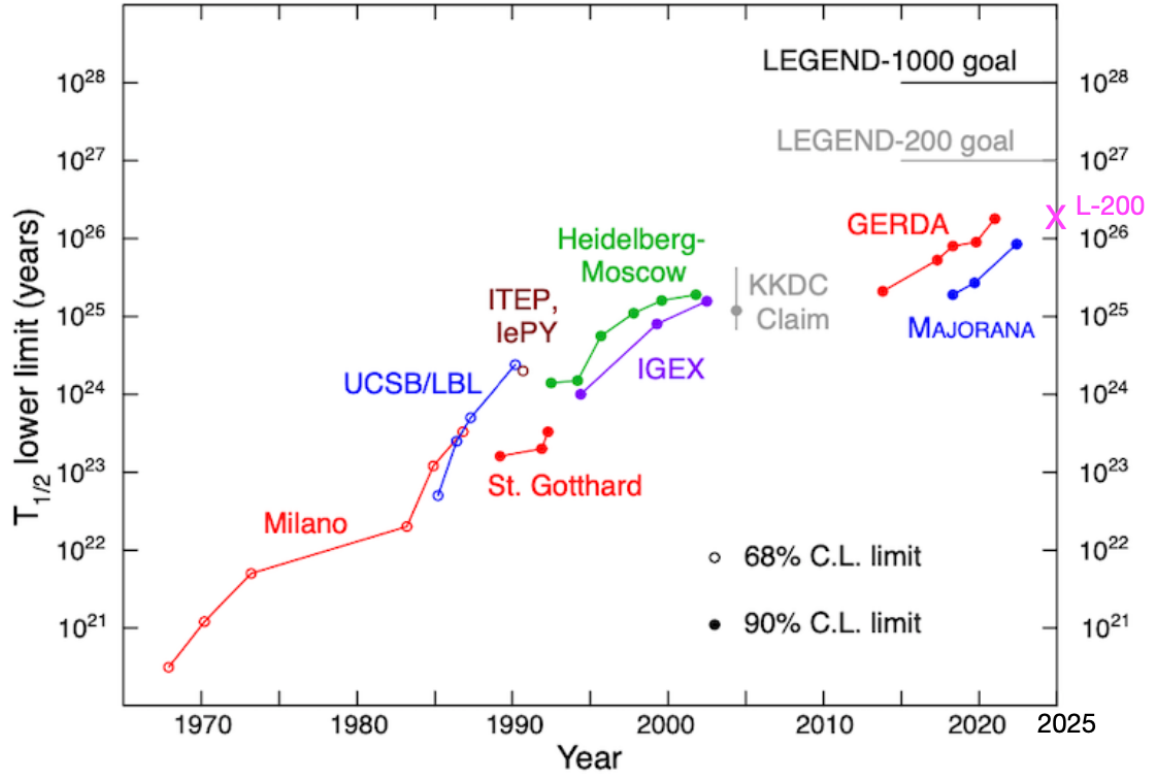


Figure 2.4: Timeline of past, present, and future germanium-76 based $0\nu\beta\beta$ decay experiments, with L-200 (LEGEND-200) is added by the author. Credit [44].

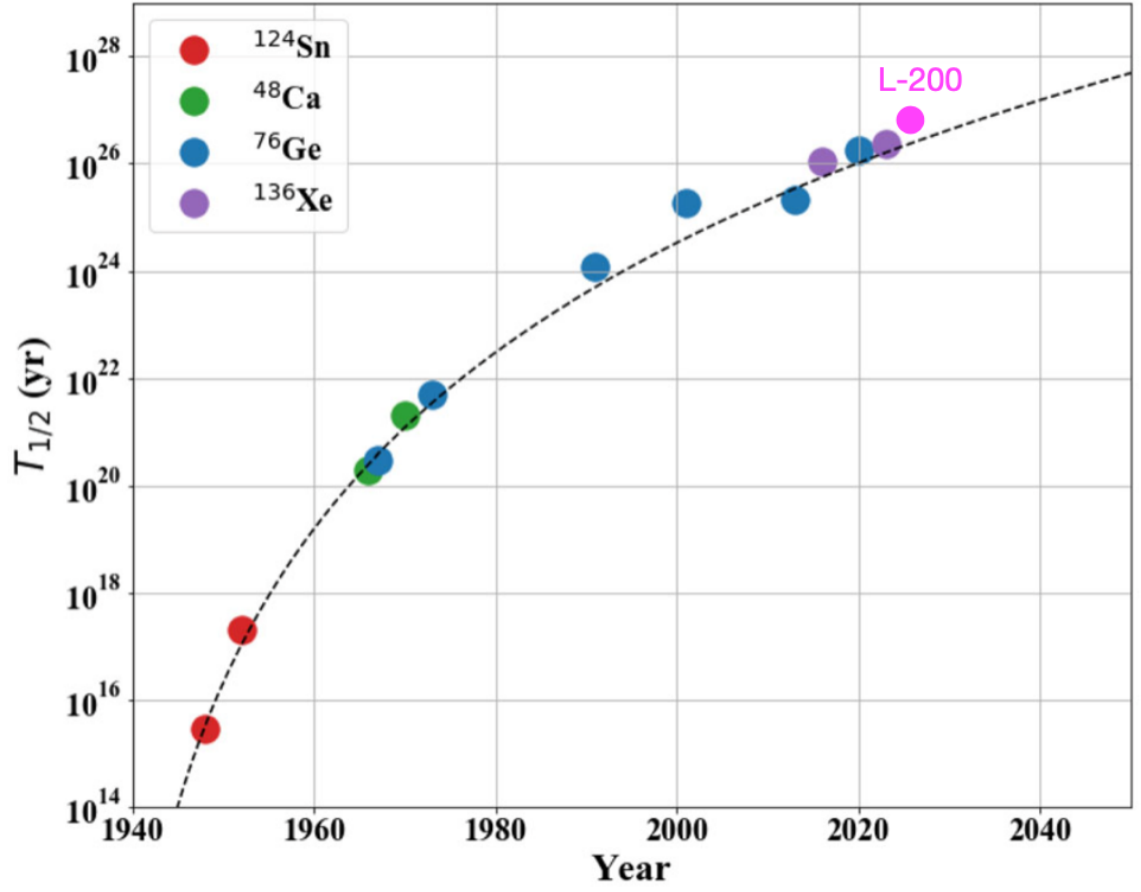


Figure 2.5: Measured $0\nu\beta\beta$ decay half-life limits of various isotopes during 76 years of searches. L-200 (LEGEND-200) is added by the author. Credit [45].

Table 2.2: Current best lower half-life limits $T_{1/2}^{0\nu}$ on $0\nu\beta\beta$ decay and corresponding upper limits on effective Majorana neutrino mass $m_{\beta\beta}$. For LEGEND-200 author adopted the same matrix element values as for GERDA and MAJORANA experiments.

Experiment	Isotope	$T_{1/2}^{0\nu}$ (10^{25} yr)	$m_{\beta\beta}$ (meV)
LEGEND-200	^{76}Ge	19	75 – 178
GERDA	^{76}Ge	18	79 – 180
MAJORANA	^{76}Ge	8.3	113 – 269
KamLAND-Zen	^{136}Xe	38	28 – 122
EXO-200	^{136}Xe	3.5	93 – 286
CUORE	^{130}Te	2.2	90 – 305

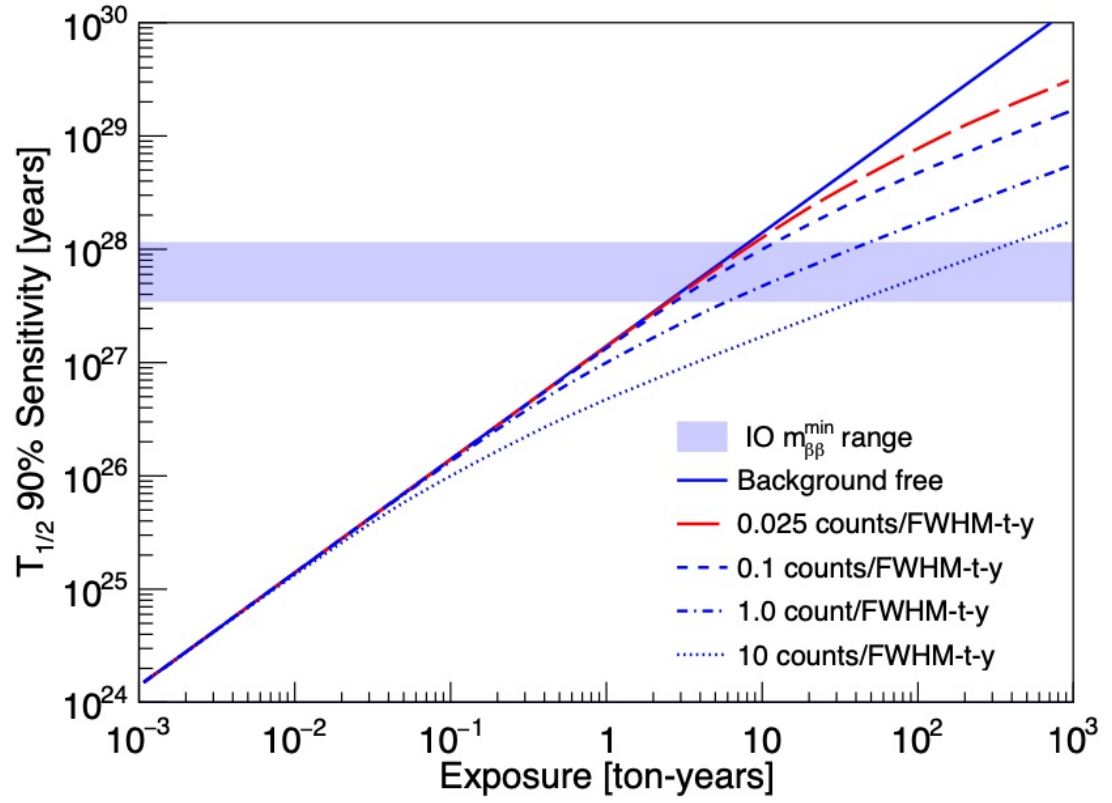


Figure 2.6: Variation of exclusion sensitivity with exposure for LEGEND-1000 experiment. Credit [44].

Chapter 3

LEGEND Experiment

The Large Enriched Germanium Experiment for Neutrinoless $\beta\beta$ Decay (LEGEND) is a next-generation $\beta\beta$ decay experiment. LEGEND aims to search for a Majorana neutrino, demonstrate the violation of lepton number conservation, and constrain or measure the effective Majorana neutrino mass. LEGEND adopts a phased approach to build the experiment, the first phase LEGEND-200 and next phase LEGEND-1000, as the collaboration merges the two previous-generation experiments, MAJORANA DEMONSTRATOR and GERmanium Detector Array (GERDA), in one single experiment. The MAJORANA DEMONSTRATOR completed the search for $0\nu\beta\beta$ decay with 27.2 kg of enriched ^{76}Ge detectors. An array of enriched germanium detectors was deployed in a vacuum cryostat located deep underground in the Sanford Underground Research Facility, Figure 3.1 (left). The MAJORANA collaboration developed underground electroformed copper and achieved world-leading energy resolution for a germanium detector at 2.52 keV. MAJORANA set the lower limit of the half-life of $0\nu\beta\beta$ decay in ^{76}Ge at $T_{1/2}^{0\nu} > 8.3 \times 10^{25}$ yr and corresponding upper limits on effective Majorana neutrino mass $m_{\beta\beta} < 113\text{--}269$ meV [42]. The final energy spectrum of MAJORANA with the exposure of 64.5 kg·yr is shown in Figure 3.2. MAJORANA’s key innovations in the development of an excellent energy resolution detector, were ultra-pure copper, radiopure components—clean dielectrics, cables, connectors, coating and low mass front-end electronics. GERDA completed a search for $0\nu\beta\beta$ decay with 44.2 kg of enriched ^{76}Ge detectors. Figure 3.1 (right) shows an array of enriched germanium detectors deployed in a liquid

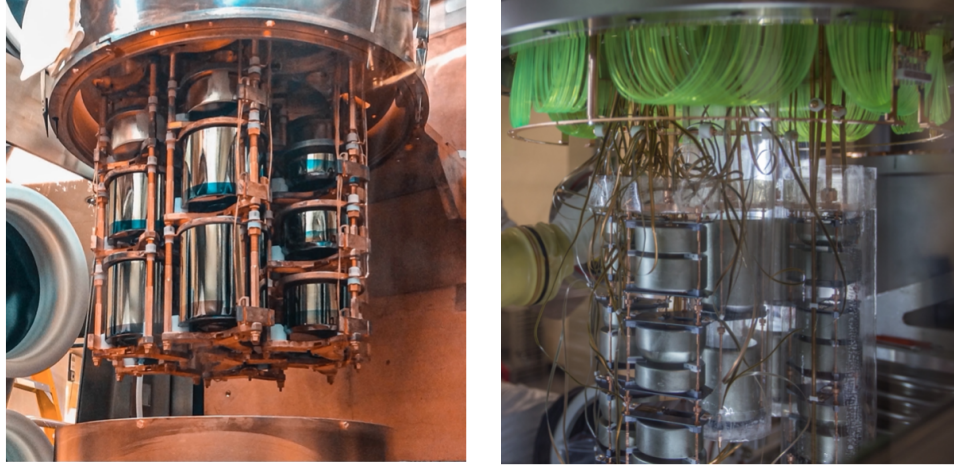


Figure 3.1: (Left) MAJORANA DEMONSTRATOR—an array of high purity enriched germanium detector supported by radiopure copper components in a vacuum cryostat. Detector modules surrounded by low-background shields consist of underground electroformed copper, commercially sourced copper, and high-purity lead. Strings are kept in an aluminum radon exclusion box, which is surrounded by plastic scintillating panels to veto cosmic muons and massive polyethylene shielding to protect from environmental neutrons. (Right) GERDA—an array of enriched germanium detectors surrounded by optical fibers coated with tetraphenyl butadiene. The string of detectors are surrounded by a nylon cylinder to limit the liquid argon volume to reduce the $^{42}\text{K}^+$ ions reaching the detectors. To protect the detectors from external radioactivity, the strings are kept in a liquid argon cryostat. Neutrons are stopped by further surrounding the cryostat with a water tank equipped with PMT to veto cosmic muons along with scintillator panels installed at the top.

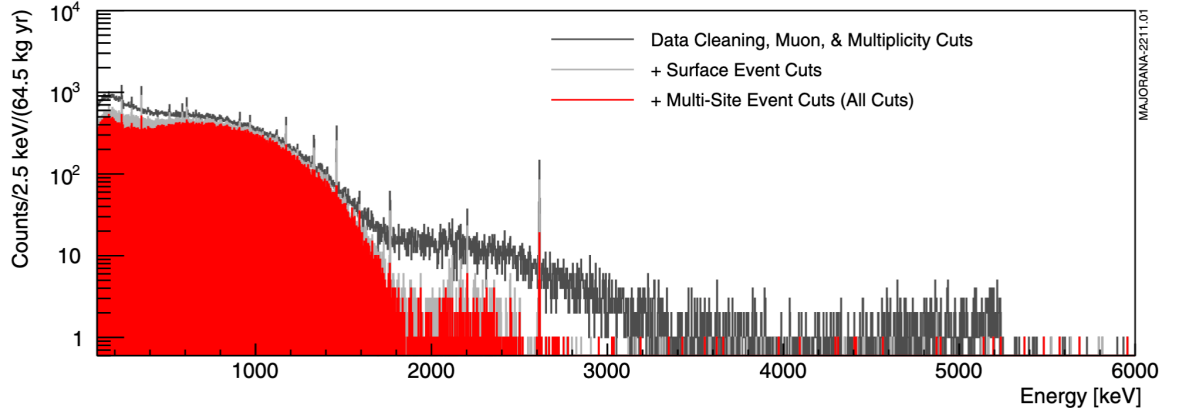


Figure 3.2: Final results of MAJORANA DEMONSTRATOR search for $0\nu\beta\beta$ decay in ^{76}Ge . Measured energy distribution and applying various cuts with the total exposure of 64.5 kg.yr. $2\nu\beta\beta$ decay continuum of ^{76}Ge is observed below $Q_{\beta\beta}$. After unblinding and all cuts, 4 events are found in the 10 keV window near $Q_{\beta\beta}$. MAJORANA set the lower limit on $T_{1/2}^{0\nu} > 8.3 \times 10^{25}$ yr and corresponding upper limits on $m_{\beta\beta} < 113\text{--}269$ meV. Credit[42].

argon cryostat located deep underground in Laboratori Nazionali del Gran Sasso. GERDA developed a liquid argon veto system and achieved a quasi-background-free regime. The GERDA collaboration set the lower limit on $T_{1/2}^{0\nu} > 1.8 \times 10^{26}$ yr and corresponding upper limits on $m_{\beta\beta} < 79\text{--}180$ meV [40]. The final energy spectrum of GERDA phase II with the exposure of 103.7 kg·yr is shown in Figure 3.3. GERDA’s key innovation is argon shielding to protect the germanium detectors from radioactivity and provide a veto signal because argon is a bright scintillator.

3.1 LEGEND-200

LEGEND-200 is the most advanced current-generation $0\nu\beta\beta$ decay experiment and started taking data in March 2023. 142 kg of high-purity germanium detectors enriched in ^{76}Ge are deployed in the existing GERDA liquid argon cryostat located deep underground in Laboratori Nazionali del Gran Sasso. LEGEND-200 germanium detector consists of 22.1 kg from MAJORANA DEMONSTRATOR, and 33.7 kg from GERDA, and a newly produced 86.7 kg. Along with the best features of MAJORANA DEMONSTRATOR and GERDA, the LEGEND-200 has new components with enhanced features, including larger germanium mass, radiopure components, better scintillation readout from the active argon veto, high-purity liquid argon, enhanced photon yield, low-mass front electronics, a charge sensitive amplifier, and a PEN baseplate to germanium. An array of enriched germanium detectors mounted with the use of copper rods in the central portion of the cryostat. Germanium detectors are surrounded by wavelength-shifting fibers connected to silicon photomultipliers for light collection and readout. The cryostat is filled with liquid argon, which acts as a radiation protection shield, a coolant for absorbing heat generated by germanium detectors, and a detection of scintillation light produced by background particles. Liquid argon emits a 128 nm photon (vacuum ultraviolet) from the interaction of external background particles, which cannot be directly detected, wavelength shifting fiber coated with tetraphenyl butadiene converts the energy of the 128 nm photon to around 500 nm (green), hence readout by silicon photomultipliers. The cryostat is then followed by the water tank with photomultiplier tubes distributed throughout the tank, which acts as a veto to incident muons. The LEGEND-200 detector is shown in Figure 3.4.

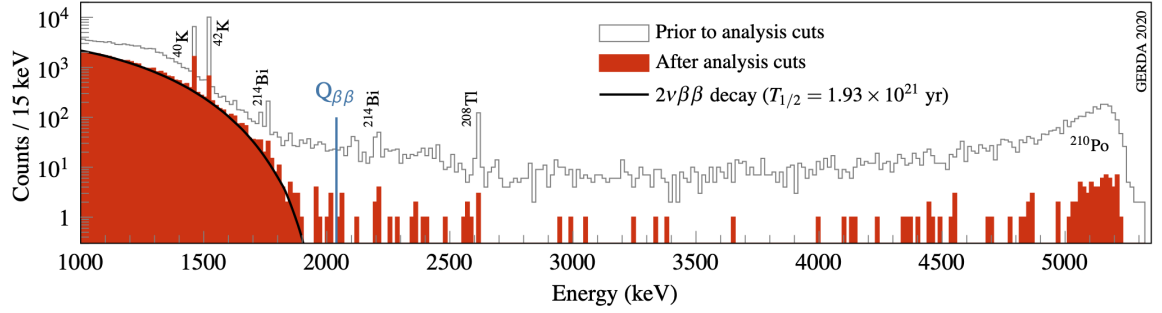


Figure 3.3: Final results of GERDA on the search for $0\nu\beta\beta$ decay in ^{76}Ge . Measured energy distribution before and after the analysis cuts with the total exposure of 103.7 kg·yr. The $2\nu\beta\beta$ decay continuum of ^{76}Ge is observed below $Q_{\beta\beta}$. After unblinding and all cuts, 13 events are found in the $0\nu\beta\beta$ decay analysis window 1930 to 2190 keV. The blue line depicts the expected $Q_{\beta\beta}$ at 2039 keV. GERDA achieved the low background index 5.2×10^{-4} counts/(keV·kg·yr) and set the lower limit on $T_{1/2}^{0\nu} > 1.8 \times 10^{26}$ yr and corresponding upper limits on $m_{\beta\beta} < 79\text{--}180$ meV. Credit [40].

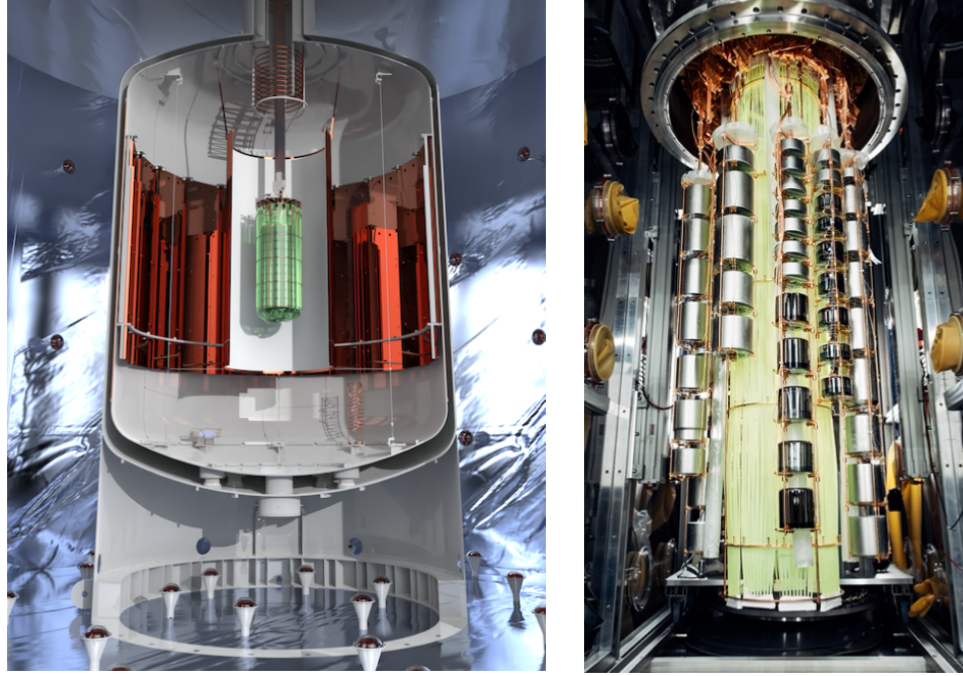


Figure 3.4: (Left) Rendering of the LEGEND-200 detector located at Gran Sasso National Laboratory. (Right) An array of 142 kg high-purity enriched germanium detectors deployed in a GERDA cryostat filled with liquid argon. Each detector is placed on a scintillating polyethylene naphthalate plate supported by copper (underground electroformed) rods. To limit $^{42}\text{K}^+$ ions reaching the germanium, the strings are surrounded by nylon cylinders coated with tetraphenyl butadiene (TPB), which are further surrounded by TPB-coated wavelength-shifting optical fibers connected with silicon photomultipliers. The cryostat is kept in a water tank equipped with photomultiplier tubes to veto cosmic muons. Credit [44].

LEGEND-200 phase-I physics data taking has been completed, with the background index $(5.3 \pm 2.2) \times 10^{-4}$ cts/(keV·kg·yr) and observed lower limit on $T_{1/2}^{0\nu} > 1.9 \times 10^{26}$ yr [3]. The first results of LEGEND-200 are shown in Figure 3.5. After background characterization, maintenance, and adding more germanium detectors, physics data taking will continue in 2025. LEGEND-200 aims to collect data for 5 years with a total exposure of 1 ton-year in a quasi-background-free regime at background index 2×10^{-4} cts/(keV·kg·yr). The experiment will set the lower limit on $T_{1/2}^{0\nu} > 9.7 \times 10^{26}$ yr and corresponding upper limits on $m_{\beta\beta} < 33\text{--}78$ meV [44]. LEGEND-200 will continue to be the leading experiment in germanium until the next-generation LEGEND-1000 starts collecting data. One year exposure of LEGEND-200 data taking will be better than five years exposure of GERDA or MAJORANA.

3.2 LEGEND-1000

“The collaboration aims to develop a phased, ^{76}Ge based double-beta decay experimental program with discovery potential at a half-life beyond 10^{28} years, using existing resources as appropriate to expedite physics results.”- LEGEND Collaboration.

The LEGEND-1000 is a next-generation $0\nu\beta\beta$ decay experiment currently in its R&D phase. This will be a new experiment with a new design and located in a new place. It will overcome the shortcoming of LEGEND-200, namely low detector mass, better background, and ^{42}Ar isotope. LEGEND-1000 specifically focuses on the mitigation of the ^{42}Ar isotope present in liquid argon. ^{42}Ar mitigation strategies include the application of underground-sourced liquid argon, encapsulation of germanium detectors, etc. The experiment requires 336 enriched ^{76}Ge detectors with a total mass of 1000 kg located in Hall C at LNGS. The conceptual design of the experiment is shown in Figure 3.6. The experiment uses high-purity p-type ICPC Ge semiconductor enriched $> 90\%$ in ^{76}Ge . The required 870 kg of ICPC detector will be newly fabricated, and the remaining 130 kg will be given by LEGEND-200. Bare ICPC detectors are mounted with copper (underground electroformed) rods, which are surrounded by a liquid argon veto system (optical fibers connected to SiPM). The strings of detectors are kept in a reentrant tube filled with underground-sourced liquid argon

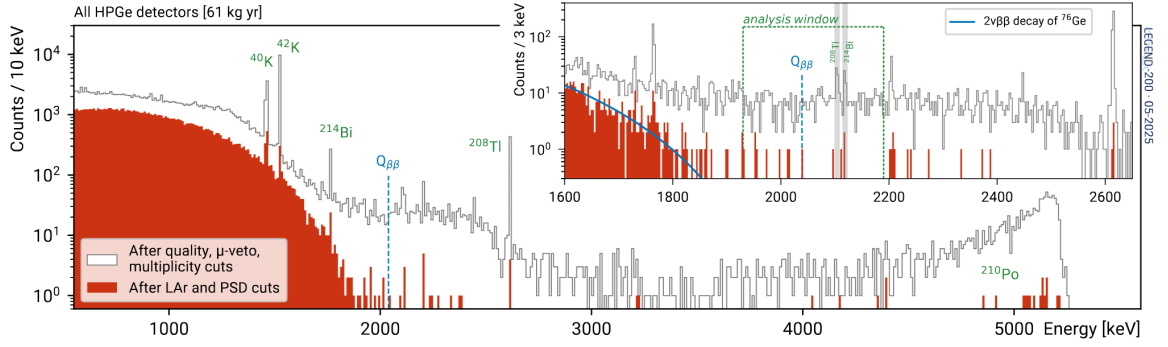


Figure 3.5: First results of LEGEND-200 for the search of $0\nu\beta\beta$ decay in ^{76}Ge . Measured energy distribution of LEGEND-200 after the analysis cuts with the exposure of 61 kg·yr. The $2\nu\beta\beta$ decay continuum of ^{76}Ge is observed below $Q_{\beta\beta}$. After unblinding and all analysis cuts, 11 events are found in the $0\nu\beta\beta$ decay analysis window of 1930 to 2190 keV. The dashed red line depicts the expected $Q_{\beta\beta}$ at 2039 keV. LEGEND-200 estimated the background index $0.54^{+0.25}_{-0.19}$ cts/(keV·tonne·yr) and set the lower limit on $T_{1/2}^{0\nu} > 1.9 \times 10^{26}$ yr and corresponding upper limits on $m_{\beta\beta} < 75\text{--}200$ meV. Credit [3].

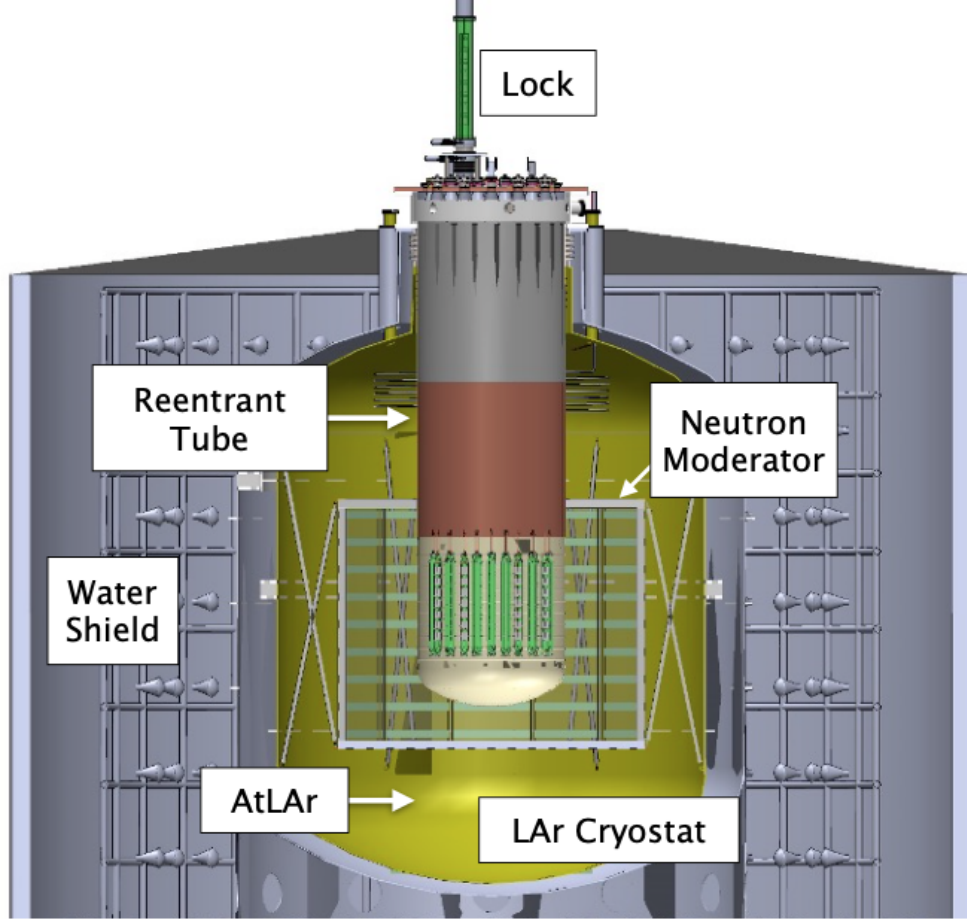


Figure 3.6: Conceptual design of the Large Enriched Germanium Experiment for Neutrinoless $\beta\beta$ Decay (LEGEND-1000) experiment. The inverted coaxial point contact high-purity germanium (1000 kg, enriched ^{76}Ge) detector strings are surrounded by waveshifting optical fibers. Detector arrays are immersed in an underground-sourced liquid argon (UGLAr) kept in a reentrant tube. UGLAr is an expensive argon and is expected to have a depleted ^{42}Ar background. The reentrant tube is kept in an atmospheric-sourced liquid argon cryostat (AtLAr). Polymethyl methacrylate panels are placed in AtLAr to reduce the environmental neutrons. Cryostat is surrounded by a water tank equipped with photomultiplier tubes. Credit [46].

(UGLAr); this can significantly reduce ^{42}Ar background. UGLAr is an expensive argon, and LEGEND-1000 cannot afford all the UGLAr for its full argon requirement. Therefore, the reentrant tube is further kept in a cryostat filled with atmospheric-sourced liquid argon (AtLAr). The cryostat is surrounded by a water tank consisting of PMTs. In case UGLAr is unavailable, then germanium detectors will require to be encapsulated with the radiopure (low-radioactivity) material in order to protect from the ^{42}Ar background.

The LEGEND-1000 is not an upgrade of the LEGEND-200 but an absolutely new experiment built with new specifications at the other part of the LNGS underground campus. The live time of LEGEND-1000 is 10 years with a total exposure of 10 ton-year and achieves a quasi-background-free regime with a background index $< 1 \times 10^{-5}$ cts/(keV·kg·yr). The discovery sensitivity on the lower limit of half-life is $T_{1/2}^{0\nu} > 1.3 \times 10^{28}$ yr and corresponding upper limits on $m_{\beta\beta} < 9\text{--}21$ meV [44]. After 10 years of data taking, the experiment can observe 3–4 events of $0\nu\beta\beta$ decay within the span of the inverse neutrino mass hierarchy band; see Figure 2.3. LEGEND-1000 will be 10 times more sensitive than its predecessor, LEGEND-200, and 100 times more sensitive than GERDA.

Design and review of LEGEND-1000 is currently ongoing. Critical Decision 1 is scheduled in 2025, and after the approval from the Department of Energy, LEGEND-1000 construction will begin at LNGS. The first physics data of LEGEND-1000 is expected to start in 2030. The expected total energy spectrum, including all backgrounds, is shown in Figure 3.7, and on the same plot, the expected signal of $0\nu\beta\beta$ decay with $T_{1/2}^{0\nu}$ of 10^{28} years is shown. The detector parameters of LEGEND-1000 are listed in Table 3.1. Ultra-low background is extremely important for the observation of $0\nu\beta\beta$ decay. My work is focused on the mitigation of the ^{42}Ar background, which is discussed in detail in Chapter 4.

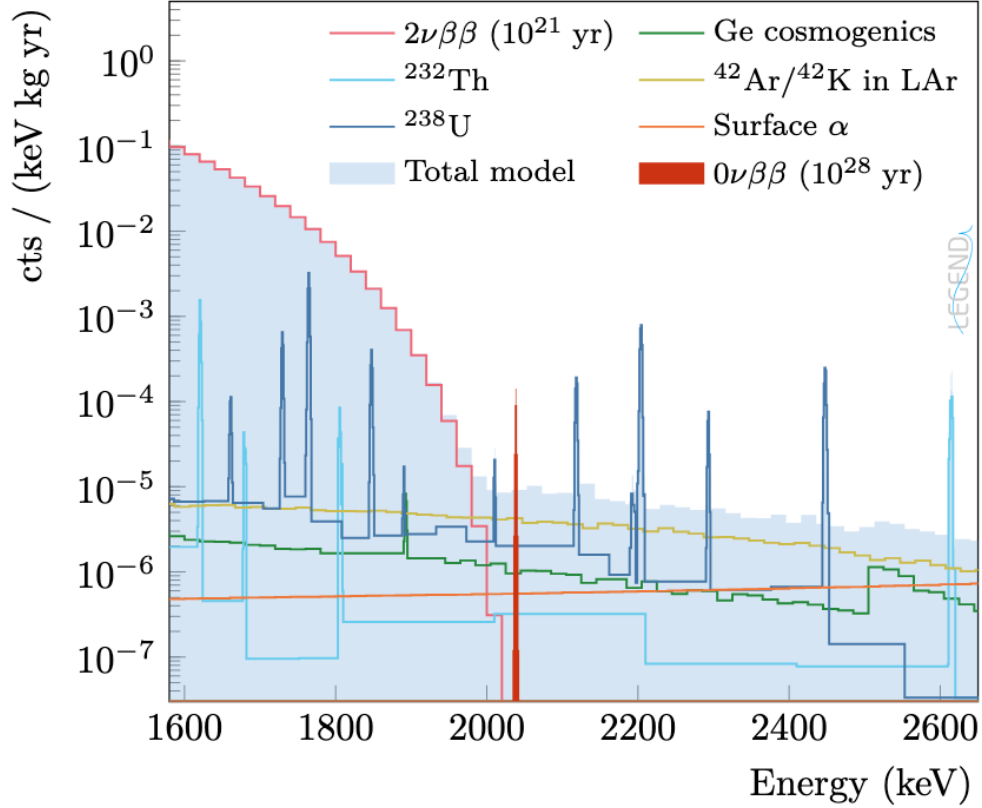


Figure 3.7: LEGEND-1000 expected background spectrum. The expected number of $0\nu\beta\beta$ decay events is 3.6 counts at the total exposure 10 ton·year with $T_{1/2}^{0\nu}$ of 10^{28} years. The $0\nu\beta\beta$ decay peak is depicted by red and separated from the continuum of $2\nu\beta\beta$ decay background. The estimated background index $< 1 \times 10^{-5}$ cts/(keV·kg·yr). As can be seen even in the optimal scenario $^{42}\text{Ar}/^{42}\text{K}$ continuum to germanium is the major source of background. Credit [44].

Table 3.1: LEGEND-1000 experimental program, [44].

Status	R&D phase
Location	Hall C at LNGS
Isotope	^{76}Ge
Goal	$^{76}\text{Ge} \rightarrow ^{76}\text{Se} + 2e^-$
$Q_{\beta\beta}$	2.039 MeV
Number of germanium detector	336
Average detector mass	3 kg
Total germanium mass	1000 kg
Live time	10 yr
Total exposure	10 tonne-year
Atmospheric LAr volume	205 m ³
Underground LAr volume	15.6 m ³
Background index	$< 1 \times 10^{-5}$ cts/(keV·kg·yr)
Half-life sensitivity	$> 1.3 \times 10^{28}$ yr
$m_{\beta\beta}$ sensitivity	$< 9\text{--}21$ meV
Expected $0\nu\beta\beta$ events for $T_{1/2}^{0\nu}$ of 10^{28} yr	3–4

Chapter 4

Mitigation Strategies of Argon-42 Background

The energy spectrum measured in the germanium detector by the LEGEND-1000 experiment is populated by various background radiations, some of which overlap with $0\nu\beta\beta$ decay signal at 2.039 MeV. Hence the $0\nu\beta\beta$ decay signal (a mono-energetic peak) could be hidden by the presence of background events. Background could originate from uranium and thorium decay chains, α , β and γ particles reaching germanium detector surfaces, radioactive isotopes present in liquid argon, and cosmogenic production of radioactive isotopes at the underground location of the detector. Such background signals are essential to minimize for the observation of the $0\nu\beta\beta$ decay signal. The underground location of the detector stops most of the external cosmogenic background events. However, internal background events could originate from the detector components, such as liquid argon, germanium, detector mounts, front-end electronics, cables, optical fibers, re-entrant tube, cryostat, and water. LEGEND-1000 aims for a “quasi-background-free” experiment with a background goal of $< 1 \times 10^{-5}$ cts/(keV·kg·yr) around 2.039 MeV. In the background-free case, sensitivity (half-life value at which there is a 50% chance of a 3σ discovery) rises linearly with experiment live time (exposure), and in the background-limited case, sensitivity rises as the square root of exposure, as shown in Figure 2.6. The collaboration is developing new techniques to minimize the effect of such backgrounds. In this work I am involved in the

R&D to mitigate the background present in the liquid argon; these efforts are described in the subsequent sections and chapters.

4.1 Cosmogenic Impurities in Atmospheric Liquid Argon

High-purity germanium detectors are operated at cryogenic temperatures to avoid the electronic noise that adversely affects the energy resolution. In LEGEND-1000, arrays of germanium diodes are immersed in a cryogenic liquid argon, which has several advantages as a (i) coolant: liquid argon extracts the heat generated from the germanium detector as it is applied to high voltage in reverse bias. (ii) Veto background particles: liquid argon is a bright scintillator; therefore, by detecting scintillation light near germanium detectors, some background signals can be effectively suppressed. (iii) Shield: liquid argon provides additional shielding to prevent the external particles that could cause a background from reaching the detector as shown in Figure 4.1 (left). The primary drawback of liquid argon, which is atmospherically sourced, contains a small amount of ^{42}Ar with a half-life of 32.9 yr. This in turn produces ^{42}K , which can cause background for the $0\nu\beta\beta$ signal in ^{76}Ge . Figure 4.1 (right) represents the decay of ^{42}Ar and its daughter ^{42}K in liquid argon; emitted β from ^{42}K can penetrate into the germanium detector and cause a background. The decay scheme of ^{42}Ar and ^{42}K is shown in Figure 4.2. Production of ^{42}Ar and ^{42}K background is described as follows:

(a) Cosmogenically produced from ^{42}Ar : gaseous argon in the upper atmosphere is constantly bombarded with cosmic rays. When cosmogenically generated α particles interact with atmospheric argon and produce ^{42}Ar , as in reaction 4.1. The liquid argon is extracted from the atmosphere, so ^{42}Ar is also present in natural argon.



(b) Production of ^{42}K : the ^{42}Ar is a long-lived radioactive isotope of argon, which undergoes β^- decay, as in reaction 4.2, with a Q -value (Q_β) = 0.599 MeV, since $Q_\beta < Q_{\beta\beta} = 2.039$ MeV (for ^{76}Ge), the ^{42}Ar does not cause a background for $0\nu\beta\beta$ signal. The ^{42}Ar β -decay spectrum is shown in the Figure 4.3.

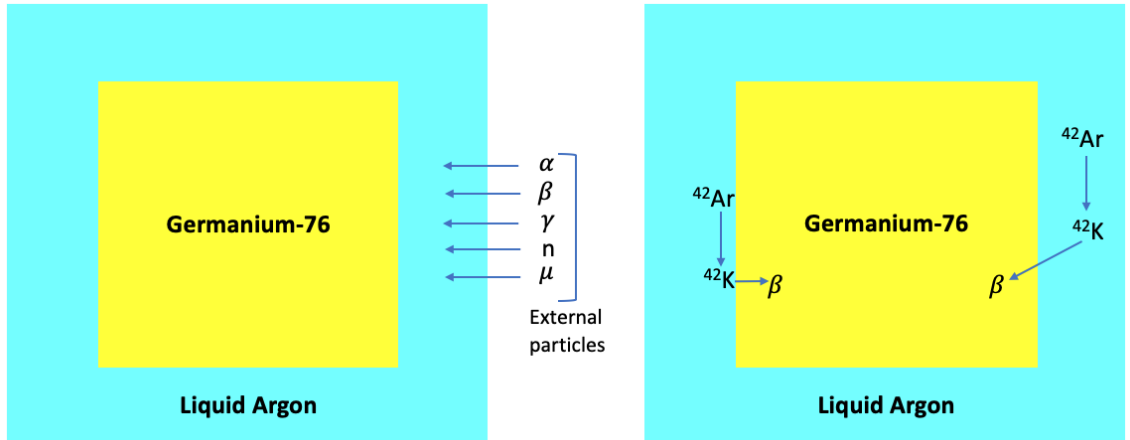


Figure 4.1: (Left) liquid argon as a shield to external particles, coolant, and veto background particles. (Right) natural argon consists of ^{42}Ar (radioactive argon), which decays to ^{42}K (radioactive), producing a background.

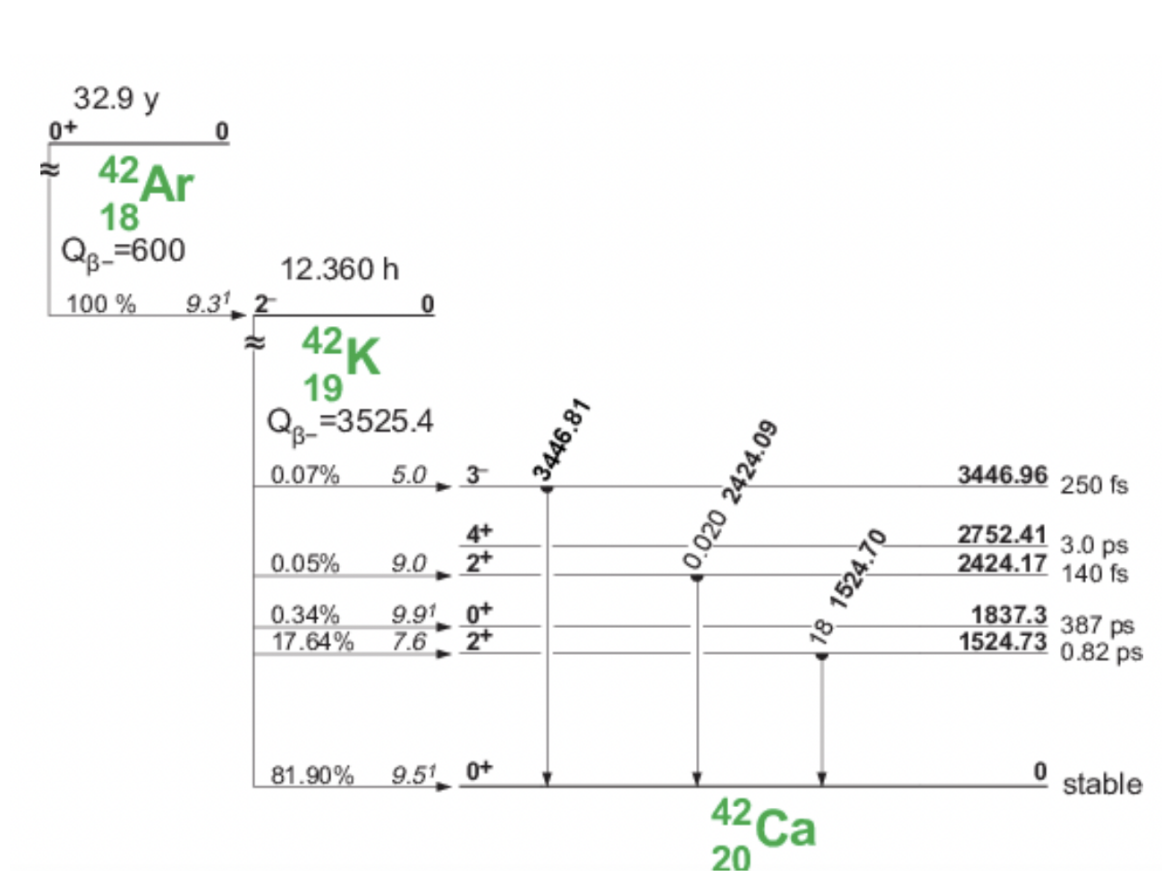


Figure 4.2: Decay scheme of ^{42}Ar and ^{42}K .

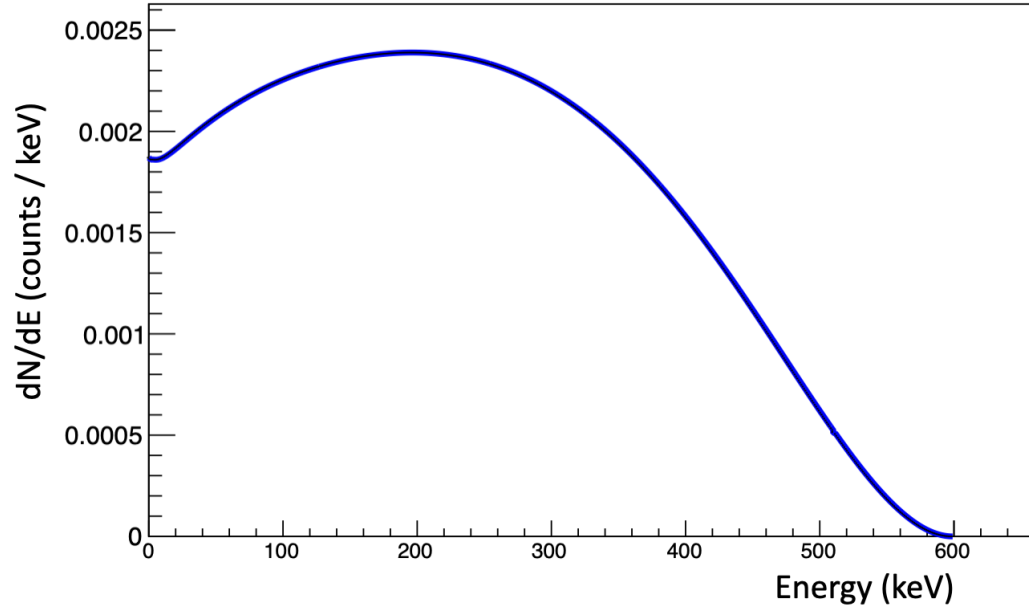


Figure 4.3: Energy continuum from ^{42}Ar decay, $^{42}\text{Ar} \rightarrow ^{42}\text{K} + \beta^- + \bar{\nu}_e$ with a $Q_\beta = 0.599$ MeV, where $Q_\beta < Q_{\beta\beta}$. ^{42}Ar is not a background for the $0\nu\beta\beta$ signal. Data is taken from [47].



(c) Decay of ^{42}K : the produced daughter isotope ^{42}K is also radioactive and has a relatively short half-life of 12.36 h and undergoes β^- decay, as in reaction 4.3, with a $Q_\beta = 3.525$ MeV, since $Q_\beta > Q_{\beta\beta} = 2.039$ MeV, and overlaps the signal region the ^{42}K can be a background for the $0\nu\beta\beta$ signal. The ^{42}K β -decay spectrum is shown in the Figure 4.4.



It been speculated that $^{42}\text{K}^+$ produced in the entire argon volume has a tendency to drift and accumulate on the surface of the germanium detectors. The emitted electron from ^{42}K decay has an energy continuum from 0 MeV to 3.525 MeV, and its energy deposition in the germanium detector can overshadow the $0\nu\beta\beta$ signal at 2.039 MeV. It is essential to reduce the concentration of ^{42}Ar in liquid argon.

4.2 Underground-Sourced Liquid Argon

The most promising method to eliminate ^{42}Ar is the use of underground-sourced liquid argon (UGLAr), i.e., argon extracted from deep underground sources. UGLAr is exposed to a much lower flux of cosmic particles; hence the lower production rate of ^{42}Ar in UGLAr is expected. The DarkSide experiment measured the cosmogenically induced isotope, ^{39}Ar , in UGLAr is 1400 times suppressed that compared with in atmospheric liquid argon [48]. The LEGEND-1000 is planned to immerse all germanium detectors in a reentrant tube (volume that contains the UGLAr) filled with UGLAr at the inner volume of the cryostat, as shown in Figure 3.6. The Argon Extraction Facility plant produces UGLAr extracted from CO_2 well in Colorado, then transferred to the ARIA plant in Sardinia for additional chemical purification. LEGEND-1000 requires approximately 22 tons of UGLAr and envisions obtaining UGLAr in 2026. However, this is a very expensive material, and therefore extra infrastructure for the experiment is needed to recover and store all argon in the case of emergency. Regular argon is relatively cheap and can be just vented out into the atmosphere. The availability of UGLAr is presently highly uncertain; therefore,

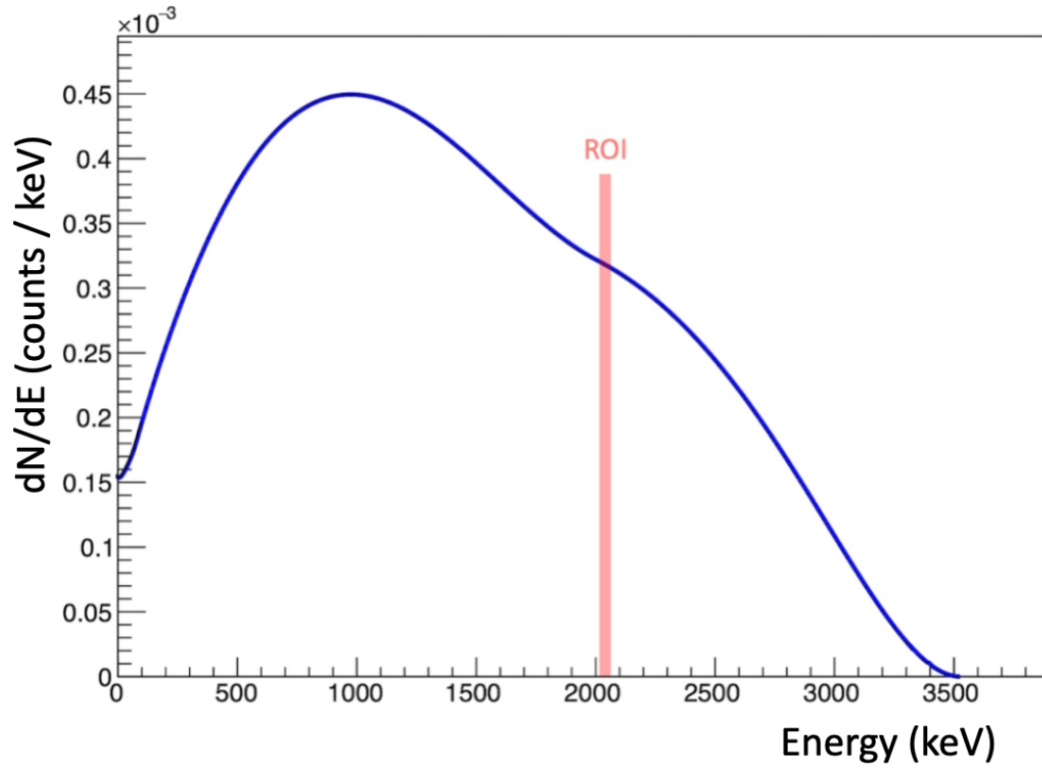


Figure 4.4: Energy continuum from ^{42}K decay, $^{42}\text{K} \rightarrow ^{42}\text{Ca} + \beta^- + \bar{\nu}_e$, with a $Q_\beta = 3.525$ MeV, $Q_\beta > Q_{\beta\beta}$ (depicted by the red band). ^{42}K is a background for the $0\nu\beta\beta$ signal. Credit [49].

LEGEND-1000 is developing new alternative techniques to mitigate ^{42}Ar in atmospheric liquid argon.

4.3 Methods to Mitigate ^{42}Ar Background

Several techniques to suppress the ^{42}Ar background from regular atmospheric argon are being studied as

- (i) Producing germanium detectors with thicker n+ dead layers (passivated insulating surfaces). Simulation of dead layer thickness shows a big loss of sensitive volume of germanium mass.
- (ii) Enclosing the detector strings with high-purity nylon mini-shrouds to reduce the liquid argon volume similar to the LEGEND-200 design.
- (iii) Improving the pulse-shape discrimination of ^{42}K surface events.
- (iv) Doping the liquid argon with a non-quenching impurity to neutralize the ^{42}K ions. However, as it is shown in my simulations below, even small amounts of liquid argon produce sizable ^{42}K background, so quenching does not help.
- (v) Enclosing the germanium detectors with wavelength-shifting and scintillating high-purity polyethylene naphthalene (PEN) or other low radioactive 3D printable material.

The focus of my dissertation is research and development of the germanium detectors using ultra-pure scintillating material, which can be custom 3D printed around each individual detector to prevent ^{42}K atoms from coming to the detectors. In addition, I run simulations on the effect of a dead layer on background reduction.

4.4 Effect of Dead Layer Thickness on ^{42}K Background

Germanium has a passivated insulating region at the surface known as the dead layer, where the detector is insensitive to particle detection. Such a dead layer can be used as a shield to reduce the ^{42}K electron energy. We use GEANT4 simulations to study the effect of the dead layer on the ^{42}K background. GEANT4 is a Monte Carlo simulation package [50–52] with an ability to replicate the experimental setup in nuclear and particle physics and perform the interaction of radiation with the detector. In this simulation, we built

an Inverted Coaxial Point-Contact (ICPC) germanium detector (88% enriched ^{76}Ge). To study the effect of dead layer on ^{42}K background, we include 4 dead layers (each 0.5 mm thick) on the germanium detector. Liquid argon is filled surrounding the detector, including the bore-hole. The detector geometry is shown in Figure 4.5. The ^{42}K isotope (radioactive) is distributed uniformly in the liquid argon, including the bore-hole.

First we simulated how much argon thickness is required for simulation. To compute the lifetime exposure (T_{Ar}) for 10^6 events of $^{42}\text{Ar}/^{42}\text{K}$ events in a given liquid argon volume V (cm^3) per detector is given by,

$$T_{Ar} = \frac{10^6}{\pi \cdot 1.396 \cdot V} \quad \text{years} \quad (4.4)$$

Total backgrounds events per 40 keV in 2.00–2.04 MeV at $Q_{\beta\beta}$ is collected in germanium (per detector). Background events per 10 keV (± 5 keV at $Q_{\beta\beta}$) is calculated and divided by the lifetime of ^{42}Ar in a given volume (calculated using equation 4.4). Background events per detector per year per 10 keV as a function of liquid argon volume (for one 3 kg ICPC germanium detector with no dead layer) are shown in Figure 4.6. Simulation shows the number of ^{42}K background events increases up to 10 mm liquid argon layer thickness around the detector and saturates after 10 mm. Therefore, adding further liquid argon does not affect results of our simulations.

Energy deposition is calculated in a germanium detector with four dead layers. The sum of energy deposition of 10^6 events in different dead layers of the detector is shown in Figure 4.7. The 1.525 MeV gamma line from ^{42}K is clearly visible over the continuum. Simulation demonstrates the effectiveness of dead layers on reducing the ^{42}K background. However, there will be a loss in germanium mass of dead layers. A 2 mm dead layer reduces 335 times the ^{42}K background (within the goal). Dead layer thickness corresponding to germanium sensitive mass with relative values of ^{42}K background reduction are summarized in Table 4.1. Background events per detector per year per 10 keV as a function of dead layer thickness in germanium (one 3 kg ICPC detector) for 10 mm of liquid argon layer is shown in Figure 4.8. Simulation shows the ^{42}K background is largest at full detector (no dead layer) and decreases with increasing thickness of the dead layer. Background events per detector per year per 10 keV at $Q_{\beta\beta}$ in different dead layer thicknesses as a function of argon volume

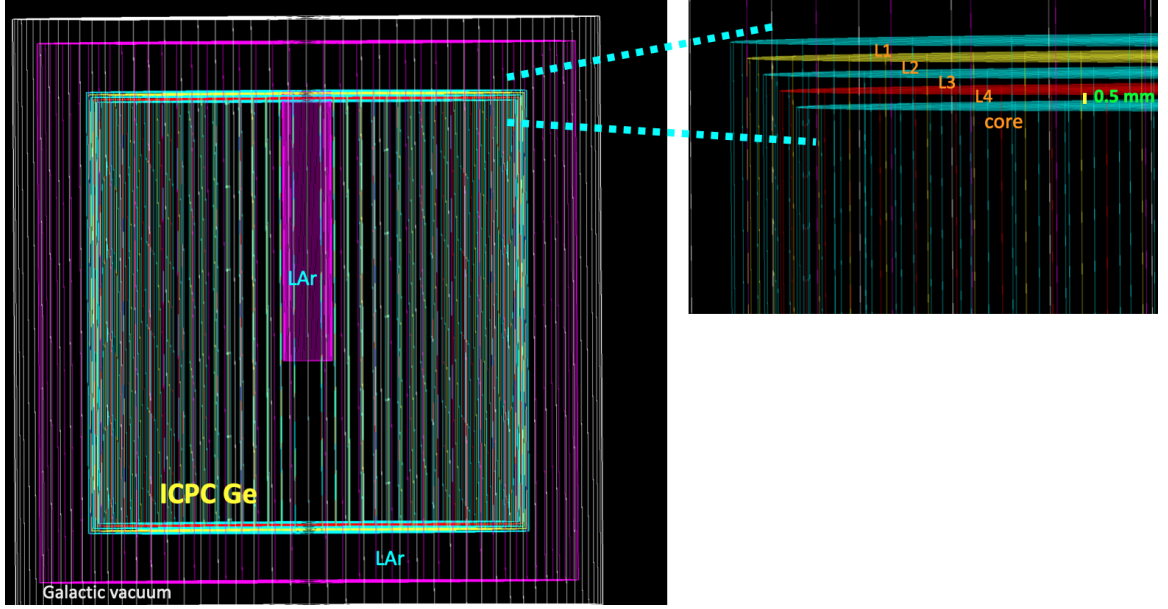


Figure 4.5: GEANT4 geometry to the simulation on the effect of dead layer. Inverted Coaxial Point-Contact (ICPC) germanium detector enriched to 88% ^{76}Ge . Detector dimensions: diameter = 90 mm and height = 90 mm; bore-hole dimension: diameter = 10 mm and height = 55 mm [53]; mass = 3.02 kg. Four dead layers (L1, L2, L3, and L4), each 0.5 mm thick, are included on the detector. The detector is surrounded by 10 mm of liquid argon (LAr), and the bore-hole is also filled with LAr. Radioactive ^{42}K is uniformly distributed in the LAr. Ultrahigh (galactic) vacuum is set outside the LAr.

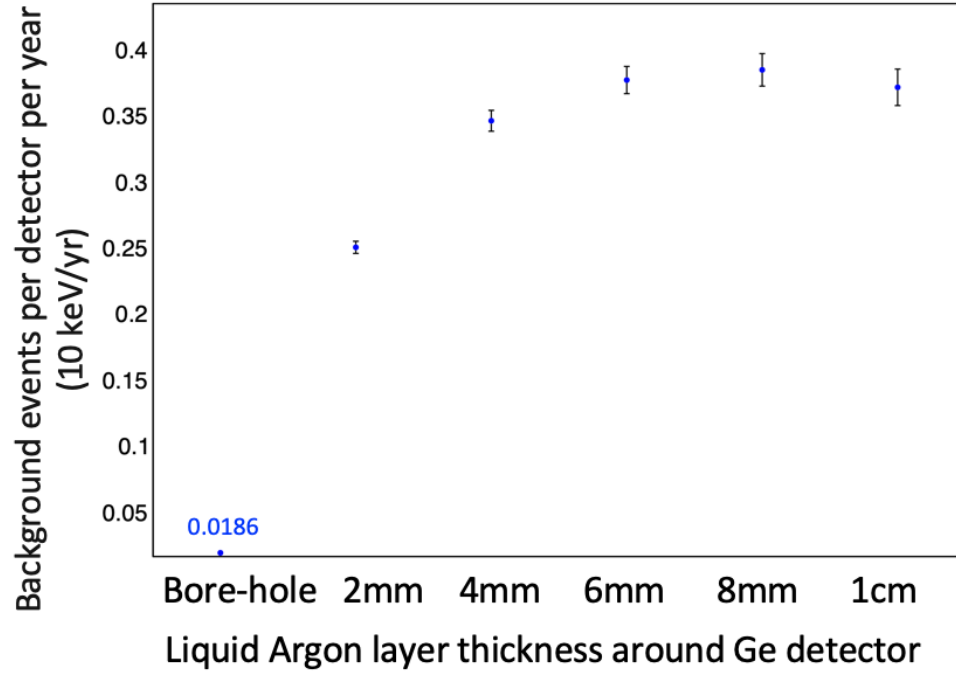


Figure 4.6: Background events per detector per year per 10 keV as a function of liquid argon volume (for one 3 kg ICPC germanium detector with no dead layer). Total background events per 40 keV in 2.00–2.04 MeV at $Q_{\beta\beta}$ is collected in the germanium. Simulation shows the number of ^{42}K background events increases up to 10 mm liquid argon layer thickness around the detector and saturates after 8 mm.

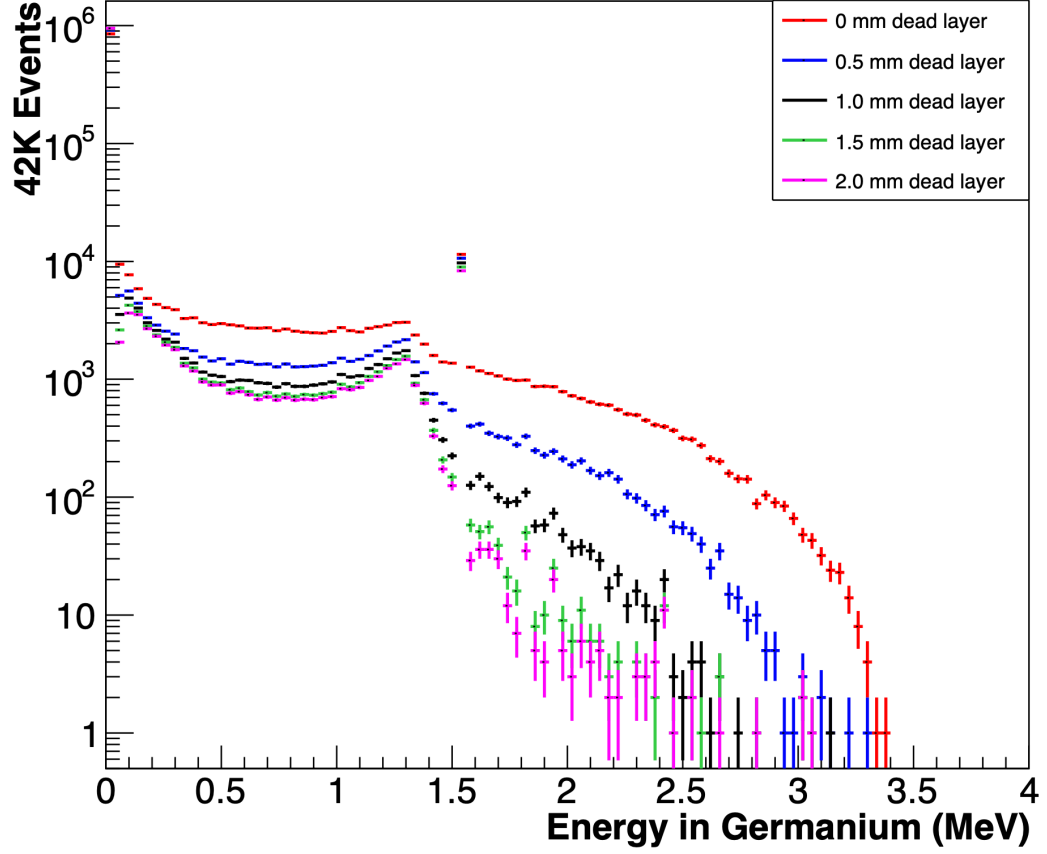


Figure 4.7: The sum of energy deposition 10^6 events of ^{42}K for different dead layers thicknesses (in 0.5 mm steps) of the germanium detector. The 1.525 MeV gamma line from ^{42}K is clearly visible over the continuum. Radioactive ^{42}K is deposited in 10 mm of liquid argon surrounding the germanium detector.

Table 4.1: Dead layer thickness corresponding to germanium sensitive mass with relative values of ^{42}K background reduction.

Dead Layer	0 mm	0.5 mm	1 mm	1.5 mm	2 mm
Germanium Sensitive Mass	100%	96%	93%	89%	85%
^{42}Ar Background Reduction Factor	1	3.9	18.9	95.8	335

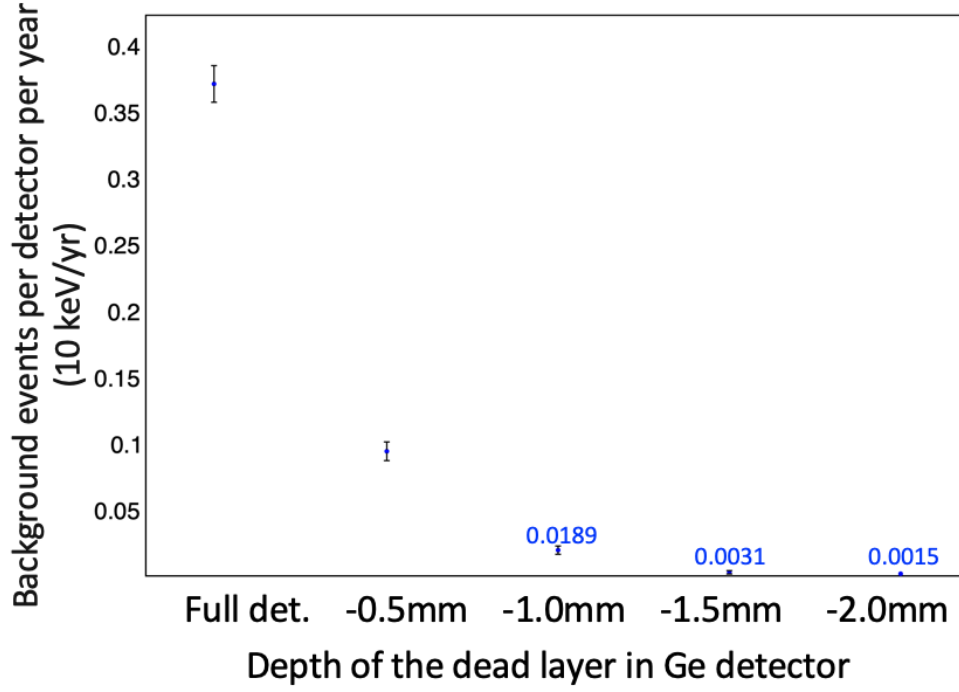


Figure 4.8: Background events per detector per year per 10 keV as a function of dead layer thickness in germanium (one 3 kg ICPC detector) for 10 mm of liquid argon layer. Total backgrounds events per 40 keV in 2.00–2.04 MeV at $Q_{\beta\beta}$ is collected in germanium. Simulation shows the ^{42}K background is largest at full detector (no dead layer) and decreases with increasing thickness of the dead layer.

are summarized in Table 4.2. Required radiation of background factor > 100 requires 1.5 mm dead layer which will result in $> 10\%$ loss of expensive ^{76}Ge material. More detailed simulations on the ^{42}K background on a germanium detector with a dead layer are described in Chapter 5.

In conclusion, there will be large mass loss, and this approach is very expensive, since the germanium detector is very costly and the production of enriched germanium ^{76}Ge detector is a long process, so we do not intend to reduce the sensitive volume of the germanium. Due to the germanium mass loss, this method also reduces the exposure of the $0\nu\beta\beta$ decay half-life sensitivity measurements since $\text{half-life} \propto \text{exposure}^{1/2}$, in the presence of background. We are studying the alternative ways to germanium dead layer thickness for reducing the ^{42}K background events. One of the techniques under consideration is to encapsulate the germanium detector with low-radioactive material, which could potentially reduce the ^{42}K - β^- energy below the ROI, which are described in the next section.

4.5 Encapsulation of Germanium Detectors

An alternative technique to prevent the detector from ^{42}Ar background is to develop the enclosure around the germanium detector. The encapsulation of detectors with low-radioactive (radiopure) material would be efficient and effective to reduce the ^{42}Ar background by absorbing the ^{42}K isotope before it can reach the sensitive volume of germanium.

Initially we consider polyethylene naphthalene (PEN) as the most promising material of encapsulation. PEN is a transparent scintillating plastic that has proven its capability as a wavelength shifter from vacuum ultraviolet (128 nm) into the blue region of the visible spectrum [54, 55]. It is a scintillator and has the same mechanical strength as copper at cryogenic temperatures. The peak emission wavelength of PEN (445 ± 5 nm) is greater than other available plastic scintillators, e.g., BC-408 (425 ± 5 nm) and polymethylmethacrylate (PMMA) (380 ± 5 nm). LEGEND-200 employs PEN as a baseplate for the germanium detector. PEN is also being considered to be a baseplate material for detector mounting for LEGEND-1000, and the radiopurity level of PEN is adequate to the LEGEND-1000 background requirements. Using GEANT4, we simulate the response of a detector with

Table 4.2: Background events per detector per year at 10 keV at $Q_{\beta\beta}$ in different dead layer thicknesses in germanium (one 3 kg ICPC detector) as a function of argon volume.

Background =Events/Lifetime (10 keV/yr)	Bore-hole argon	2 mm argon	4 mm argon	6 mm argon	8 mm argon	10 mm argon
0 mm dead layer	(1.861±0.030) ·10 ⁻²	(2.503±0.046) ·10 ⁻¹	(3.459±0.079) ·10 ⁻¹	(3.711±0.103) ·10 ⁻¹	(3.848±0.123) ·10 ⁻¹	(3.711±0.138) ·10 ⁻¹
0.5 mm dead layer	(5.218±0.157) ·10 ⁻³	(7.969±0.257) ·10 ⁻²	(1.049±0.043) ·10 ⁻¹	(9.786±0.525) ·10 ⁻²	(9.945±0.625) ·10 ⁻²	(9.651±0.704) ·10 ⁻²
1.0 mm dead layer	(1.137±0.073) ·10 ⁻³	(1.738±0.120) ·10 ⁻²	(1.655±0.172) ·10 ⁻²	(1.997±0.237) ·10 ⁻²	(1.729±0.261) ·10 ⁻²	(1.899±0.312) ·10 ⁻²
1.5 mm dead layer	(2.036±0.310) ·10 ⁻⁴	(2.069±0.414) ·10 ⁻³	(2.491±0.666) ·10 ⁻³	(3.937±1.052) ·10 ⁻³	(3.145±1.112) ·10 ⁻³	(3.080±1.257) ·10 ⁻³
2.0 mm dead layer	(8.050±1.952) ·10 ⁻⁵	(6.620±2.240) ·10 ⁻⁴	(8.897±3.979) ·10 ⁻⁴	(1.125±0.563) ·10 ⁻³	(1.179±0.681) ·10 ⁻³	(1.540±0.889) ·10 ⁻³

PEN encapsulation exposed to ^{42}K background. For this purpose, we input the geometry of the germanium detector in GEANT4, having the dimensions of 9cm height and 9cm diameter; these are the same dimensions as the detector at ORNL. Thereafter, layer the PEN material over the top cross-section of the detector and set the surrounding environment to be filled with liquid argon, as shown in Figure 4.9. The β^- decay of ^{42}K has a Q -value of 3.525 MeV. So we input the source of the electron with the energy of 3.525 MeV at the center of the top cross-section of PEN material and set the momentum of β^- along the perpendicularly inward direction, i.e., all β^- travel inside the PEN material and deposit their energy inside the PEN. In the Figure 4.9, red represents the electron distribution in the PEN. Because of the bremsstrahlung, energetic photons were generated, and these photons further propagated into the adjacent germanium detector, and their trajectories are shown as green in Figure 4.9.

We calculated the energy deposition in the germanium detector by varying the encapsulated thickness of PEN material from 2 mm to 12 mm by the incident of 3.525 MeV β^- particles, obtained energy spectra as shown in Figure 4.10, and each simulation run with 10,000 events. It is clear from the Figure 4.10 that the thicker the encapsulation, the lesser the energy deposition in the germanium detector. The simulation shows that 6 to 8 mm of PEN encapsulation would be sufficient to push the energy of the β^- below the 2039 keV.

However, encapsulation of detectors with 6 to 8 mm passive PEN would not be viable because of radio-purity considerations, since PEN has some radio-impurities by itself. The more external material in the vicinity of the germanium detector, the more potential there is for other sources of background. However, encapsulation with 2 mm optically active PEN along with LAr veto would be a viable option for the reduction of the background caused by ^{42}K at the region of interest.

Additionally, Figure 4.11 shows the simulated spectra of the energy distribution in a germanium detector of 9 cm thick and the energy distribution in PEN of 2 mm thick. 2 mm PEN will absorb more than 0.25 MeV of electron energy. This should be enough to produce scintillation light, which can effectively veto all electrons moving toward the detector. As a result, encapsulation of detectors by PEN is a promising approach. The one issue is to build efficient light collection from PEN scintillation. The other issue is that ultra-clean PEN is difficult to build, and this is not 3D-printable material. Because all

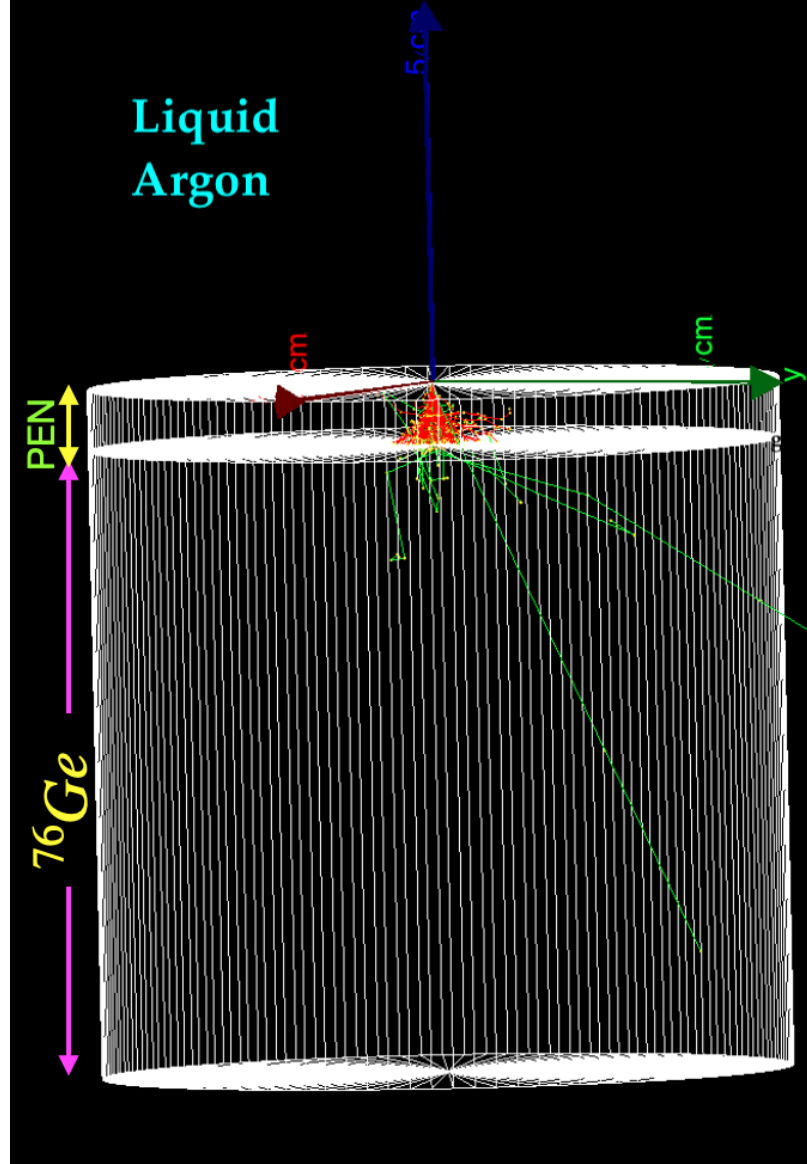


Figure 4.9: GEANT4 simulation of the energy deposition of 3.525 MeV electrons (Q -value of ^{42}K) in PEN-encapsulated germanium detector kept in liquid argon. The source is located at the top center of PEN. Red represents the electron density in the PEN, and green represents the photon trajectories in germanium.

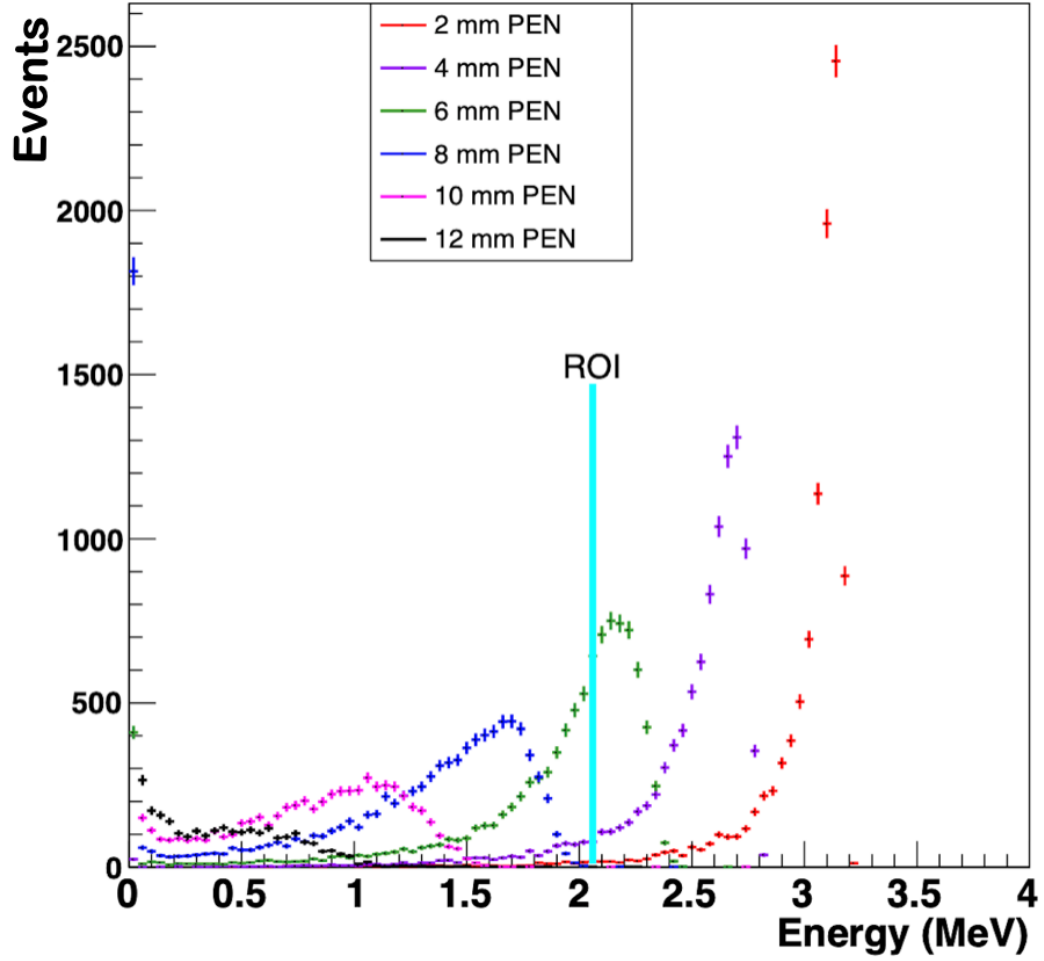


Figure 4.10: GEANT4 result of energy deposition in germanium at various PEN thicknesses for the incident electrons of 3.525 MeV (Q -value of ^{42}K). Simulation shows 6 to 8 mm PEN encapsulation to germanium would be sufficient to lower the energy of electrons below the 2039 keV ($Q_{\beta\beta}$ of ^{76}Ge). Cyan band depicts the ROI. Credit [49].

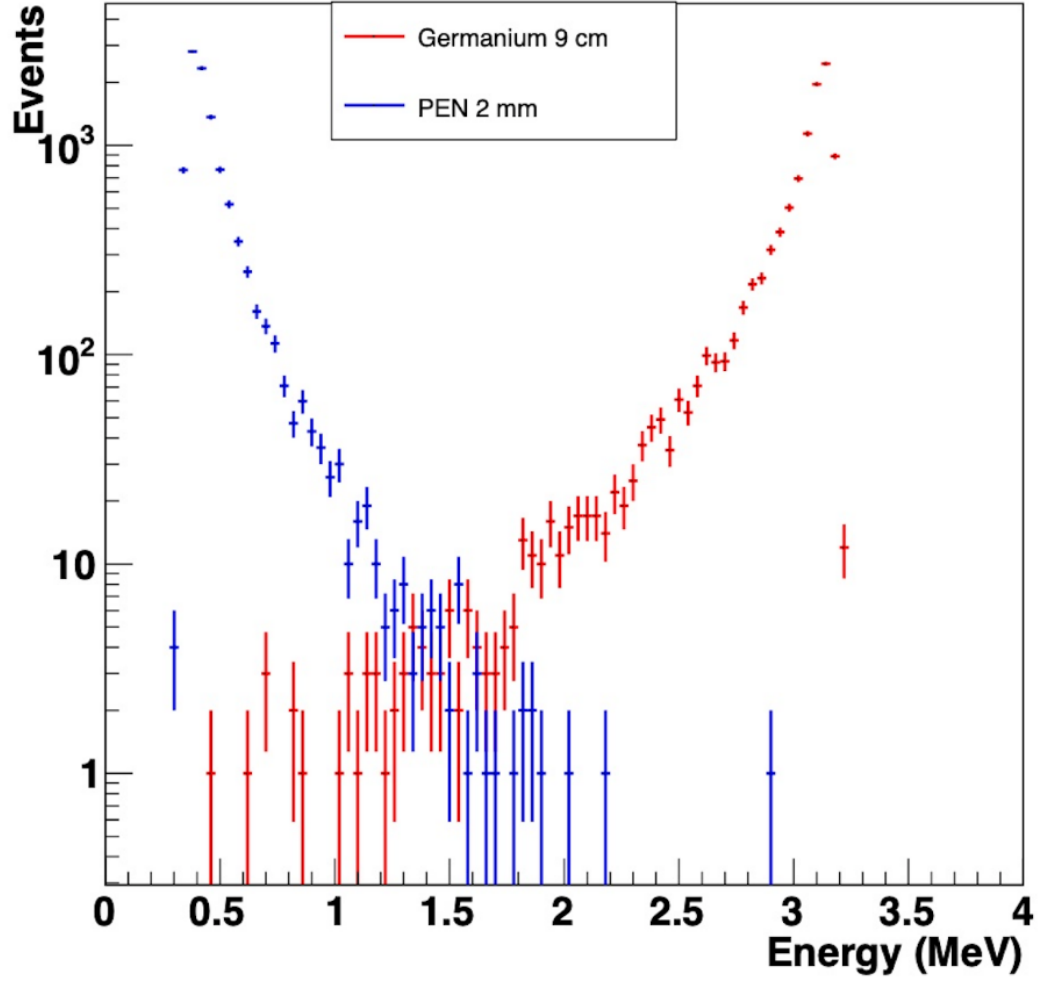


Figure 4.11: GEANT4 result of energy deposition in germanium (9 cm thick) and PEN (2 mm thick) for the incident electrons of 3.525 MeV. Simulation shows 2 mm PEN will absorb more than 0.25 MeV of incident electron energy. LEGEND-200 PEN scintillator batch measured the light yield (5440 ± 220) photons/MeVee [56]. Credit [49].

detectors have different sizes, customization of PEN encapsulation could be a long and expensive approach.

As a result, we focused on the materials that are 3D printable and can be used for encapsulation in a clean, hands-free environment.

Chapter 5

Monte Carlo Simulation for Various Scenarios of Detector Encapsulation

The LEGEND-1000 experiment is currently an extensive R&D program on mitigating the $^{42}\text{Ar}/^{42}\text{K}$ background. One of the techniques to suppress the ^{42}Ar background from regular atmospheric argon is to produce germanium detectors with thicker n+ dead layers. To study the impact of ^{42}K background on a germanium detector with a thin dead layer and various encapsulation scenarios, we performed the following simulations using GEANT4 [50–52].

An inverted-coaxial point-contact (ICPC) germanium detector enriched (88% ^{76}Ge) (diameter = 9 cm and height = 9 cm) with a bore-hole (diameter = 10 mm and height = 55 mm) having a mass of 3.02 kg is simulated. A germanium dead layer is included on all surfaces, and the detector is immersed in liquid argon. Interaction of the ^{42}K isotope with the germanium, dead layer, and liquid argon are simulated in different scenarios. Generated events in each part of the detector are collected. Energy deposited by β^- particles called primary events (point like) and by γ rays are secondaries (fuzzy). The sum of primary and secondary energy deposited is referred to as total. Primary electrons and secondary particles are separately recorded for each segment of the setup. Simulations are performed for three different configurations as follows:

- (i) ICPC detector with 0.5 mm dead layer surrounded by 1 mm liquid argon layer; ^{42}K is uniformly distributed in liquid argon.
- (ii) ICPC detector with 1 mm dead layer surrounded by 1 mm liquid argon layer; ^{42}K is uniformly distributed in liquid argon.
- (iii) ICPC detector with 0.5 mm dead layer surrounded by 0.2 mm liquid argon layer; ^{42}K is uniformly distributed in liquid argon.

These configurations and simulation results of the interaction of ^{42}K decay with liquid argon and germanium detector are described below.

5.1 ICPC detector with 0.5 mm dead layer surrounded by 1 mm liquid argon layer; ^{42}K is uniformly distributed in liquid argon

This study is to understand how much liquid argon can be allowed between encapsulation and detector. In this simulation, a 0.5 mm thick dead layer is included on all surfaces of an Inverted Coaxial Point-Contact (ICPC) germanium detector. Surrounding the detector, a 1 mm layer of liquid argon is filled, including the bore-hole. The schematic of the setup is shown in Figure 5.1. Radioactive ^{42}K isotopes (10^6 events) are uniformly distributed in liquid argon.

In a liquid argon medium, radioisotope ^{42}K decays ($^{42}\text{K} \rightarrow ^{42}\text{Ca} + \beta^- + \bar{\nu}_e$), producing β^- with an energy continuum up to 3.525 MeV; these β^- are counted as primary electrons. Such β^- propagates and deposits energy in liquid argon primarily by ionization.

Energy distribution of primary electrons in liquid argon is shown in Figure 5.2. As a result of nuclear de-excitation of ^{42}K released a 1.525 MeV γ -line and the interaction of primary electrons with the liquid argon medium produced additional particles counted as secondaries. Some of these secondary particles proceed toward the detector and, during propagation, deposit energy in the liquid argon. Figure 5.3 represents the total (primary+secondary) energy distribution in 1 mm liquid argon. After traversing liquid argon layer, energetic primary electrons and secondary particles can penetrate the 0.5 mm dead layer deposited on the ICPC detector. The dead layer acts as a shield to the germanium

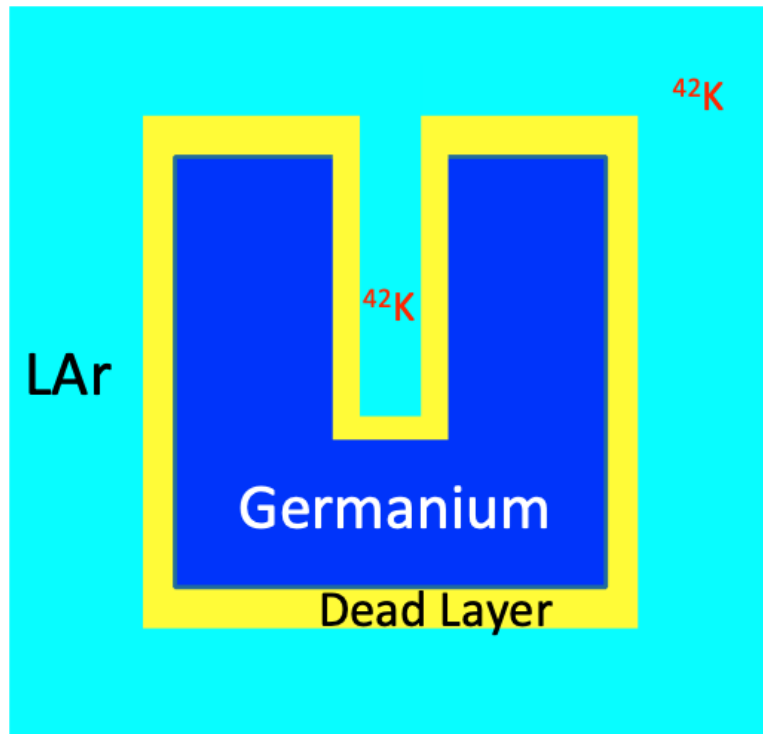


Figure 5.1: Schematics of ICPC detector with 0.5 mm dead layer surrounded by 1 mm liquid argon layer. The ^{42}K isotope is uniformly distributed in liquid argon. *Not drawn to scale to enhance the visibility for showing dead layer and liquid argon.*

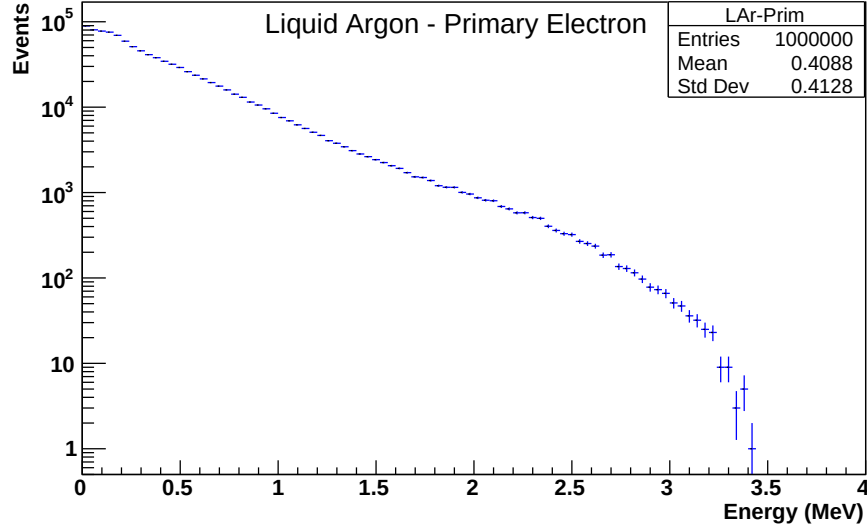


Figure 5.2: 0.5 mm dead layer, ^{42}K in 1 mm liquid argon: energy distribution in 1 mm liquid argon by primary ($^{42}\text{K}-\beta^-$ only) events. $^{42}\text{K} \rightarrow ^{42}\text{Ca} + \beta^- + \bar{\nu}_e$, producing β^- with an energy continuum up to 3.525 MeV; these β^- are counted as primary electrons.

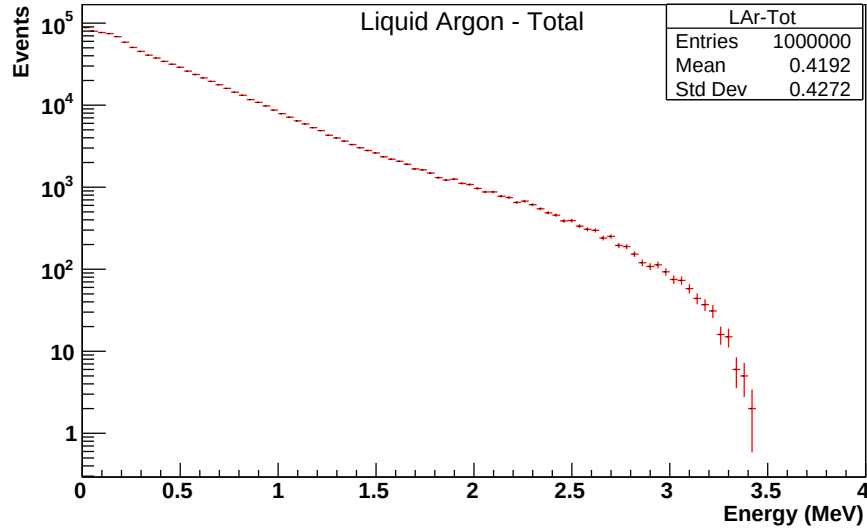


Figure 5.3: 0.5 mm dead layer, ^{42}K in 1 mm liquid argon: energy distribution in 1 mm liquid argon by total (primary+secondary) events. 1.525 MeV γ -line from nuclear de-excitation of ^{42}K and other particles produced after interaction of primary electrons in liquid argon are secondaries.

detector and reduces some energy of both primary electrons and secondary particles. Such energetic particles enter a sensitive volume of the germanium region where $0\nu\beta\beta$ decay would occur, where these primary electrons and secondary particles could overlap the region of interest and contribute to the background. Primary electrons deposit energy in germanium mostly by bremsstrahlung and produce additional gamma photons as a result of energy loss of β^- in germanium. Energy distribution of primary electrons in germanium is shown in Figure 5.4. Incoming secondary particles in a germanium medium also lose energy by bremsstrahlung. 1.525 MeV photons lose energy in germanium by photoelectric absorption, Compton scattering, and Rayleigh scattering; such photons are identified as a distinct sharp γ -line in the energy continuum. Energy distribution of secondary (1.5 MeV γ -line + all secondaries produced) in germanium is shown in Figure 5.5. Good pulse shape discrimination in a germanium detector potentially can identify these events. This is a subject of future studies. The correlation between total energy in germanium with total energy in liquid argon is shown in figure 5.6. We have events in the region of interest. By applying a veto cut in argon, we can reduce the ^{42}K background in the germanium detector. The total energy in liquid argon when energy deposition in germanium ± 50 keV at $Q_{\beta\beta}$ (1989–2089 keV) is shown in Figure 5.7. The simulation shows below 0.1 MeV approximately 2000 events in liquid argon in 100 keV at $Q_{\beta\beta}$. 0.1 MeV is the estimated threshold of energy deposition detection in liquid argon. These 2000 events in the liquid argon are undetected and can not be vetoed by the liquid argon veto system. Such events contribute to the ^{42}K background in the germanium detector. To compute the lifetime exposure (T_{Ar}) of N events of $^{42}\text{Ar}/^{42}\text{K}$ background in a given liquid argon volume V (cm^3) is given by,

$$T_{Ar} = \frac{N}{\pi \cdot 1.396 \cdot V} \quad \text{years} \quad (5.1)$$

In this simulation, 1 mm layer of liquid argon surrounding the ICPC detector, including the bore-hole with a volume $V = 43.34 \text{ cm}^3$ and ^{42}K events $N = 10^6$. Using equation 5.1, the lifetime of 10^6 events of ^{42}K in 1 mm liquid argon corresponds to $T_{Ar} = 5261$ years/detector. There are 336 detectors in LEGEND-1000 experiment with a mass of 3.02 kg per detector and the total mass of 1 ton. Therefore the simulated lifetime for LEGEND-1000 is $\sim 5261/336$ years ~ 15 years.

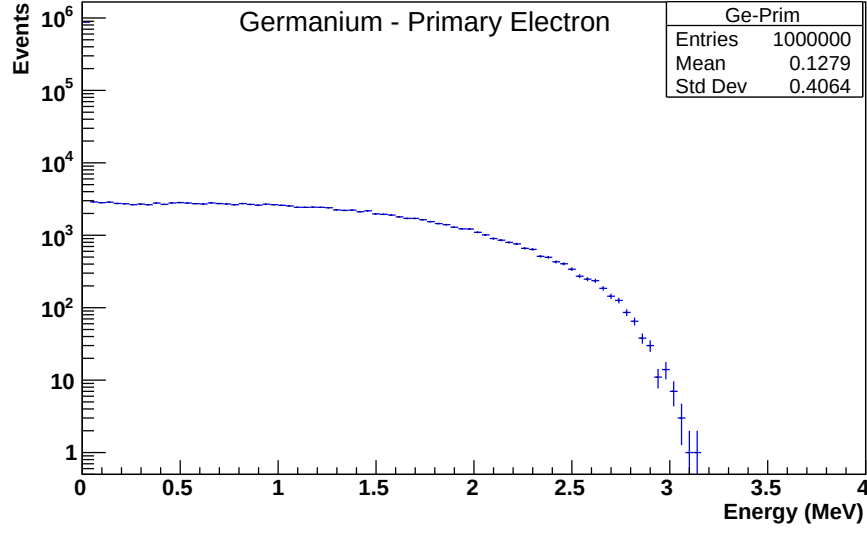


Figure 5.4: 0.5 mm dead layer, ^{42}K in 1 mm liquid argon: energy distribution in germanium by primary ($^{42}\text{K}-\beta^-$ only) events. Primary electrons deposit energy in germanium mostly by bremsstrahlung and produce additional gamma photons as a result of energy loss of β^- in germanium.

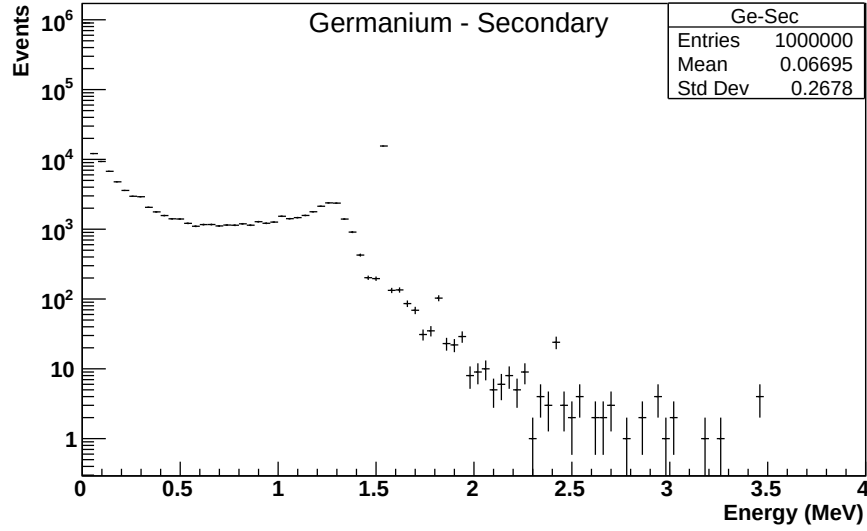


Figure 5.5: 0.5 mm dead layer, ^{42}K in 1 mm liquid argon: energy distribution in germanium by secondary (1.5 MeV gamma + all secondaries produced). The γ -line from the nuclear de-excitation of ^{42}K is clearly visible at 1.525 MeV. Secondary particles lose energy by bremsstrahlung, and 1.525 MeV photons lose energy in germanium by photoelectric absorption, Compton scattering, and Rayleigh scattering. Good pulse shape discrimination in a germanium detector potentially can identify these events.

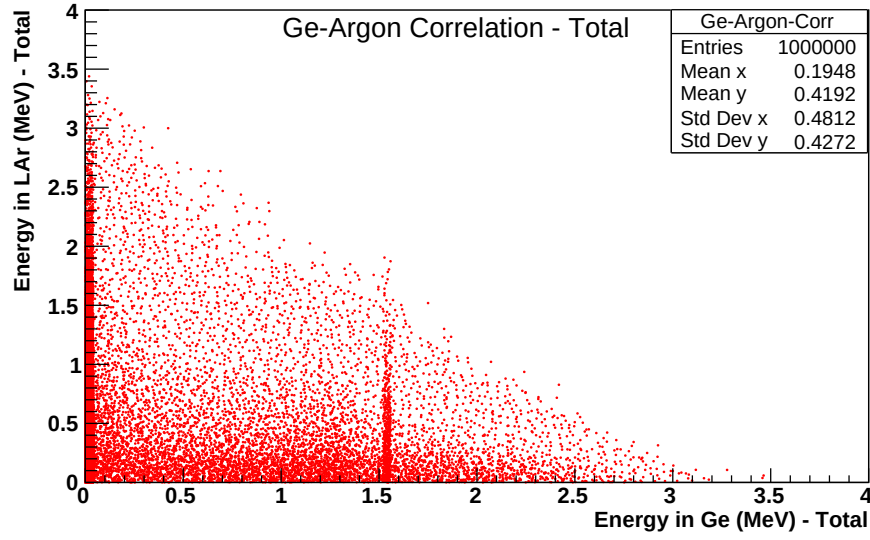


Figure 5.6: 0.5 mm dead layer, ^{42}K in 1 mm liquid argon: correlation of total energy in germanium with total energy in liquid argon. We have events in the region of interest. By applying a veto cut in argon, we can reduce the ^{42}K background in the germanium detector.

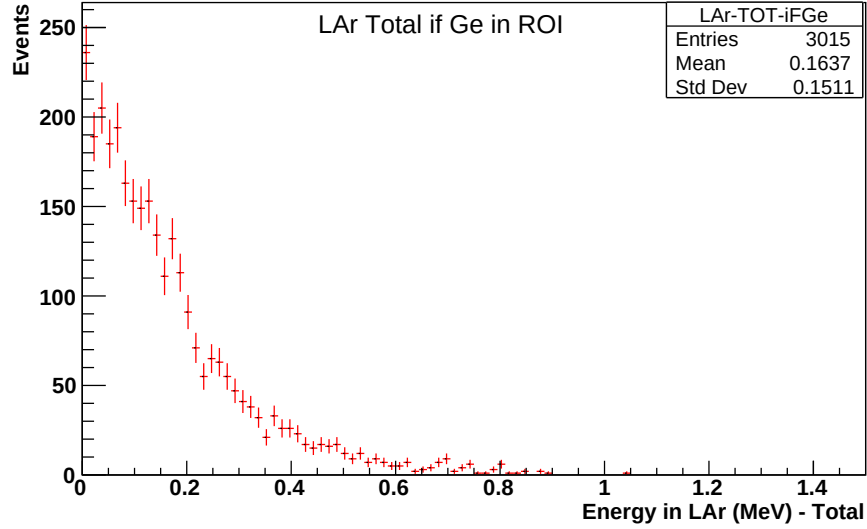


Figure 5.7: 0.5 mm dead layer, ^{42}K in 1 mm liquid argon: total energy in liquid argon when energy deposition in germanium ± 50 keV at $Q_{\beta\beta}$ (1989–2089 keV). Below 0.1 MeV, approximately 2000 events in liquid argon in 100 keV at $Q_{\beta\beta}$. These 2000 events in the liquid argon are undetected and can not be vetoed by the liquid argon veto system. Such events contribute to the ^{42}K background in the germanium detector.

We have approximately 2000 counts of ^{42}K from 1 mm layer of liquid argon in the 100 keV window (± 50 keV) in germanium detector at $Q_{\beta\beta}$ corresponds to background index ($\mathcal{B}$) for LEGEND-1000 is estimated as $\mathcal{B} \sim 2000 \text{ counts}/(100 \text{ keV} \cdot 1 \text{ ton} \cdot 15 \text{ years}) \sim 1.33 \text{ cts}/(\text{keV} \cdot \text{t} \cdot \text{yr})$. The projected background goal of LEGEND-1000 for ^{42}K is $\mathcal{B} \sim 0.6 \text{ cts}/(\text{keV} \cdot \text{t} \cdot \text{yr})$ [44]. The detector with 0.5 mm dead layer and 1 mm layer of liquid argon produce the larger ^{42}K background than the LEGEND goal.

In conclusion, we need either a detector with a thicker dead layer or reduced liquid argon volume surrounding the detector. Scenario of the thicker dead layer is discussed in the next section.

5.2 ICPC detector with 1 mm dead layer surrounded by 1 mm liquid argon layer; ^{42}K is uniformly distributed in liquid argon

In this part we simulated how a thicker dead layer germanium detector influences the background rate. A 1.0 mm thick dead layer is included on all surfaces of a ICPC germanium detector. Surrounding the detector 1 mm layer of liquid argon is filled, including the bore-hole. The schematic of the setup is shown in Figure 5.8. Radioactive ^{42}K isotopes (10^6 events) are uniformly distributed in liquid argon. We follow a similar procedure as in the previous part and show only final results.

The correlation between total energy in germanium with total energy in liquid argon is shown in Figure 5.9. The total energy in liquid argon when energy deposition in germanium ± 50 keV at $Q_{\beta\beta}$ (1989–2089 keV) is shown in Figure 5.10. The simulation shows below 0.1 MeV, approximately 500 events in liquid argon in 100 keV at $Q_{\beta\beta}$. 0.1 MeV is the estimated threshold of energy deposition detection in liquid argon. These 500 events in the liquid argon are undetected and can not be vetoed by the liquid argon veto system. Such events contribute to the ^{42}K background in the germanium detector.

In this simulation, 1 mm layer of liquid argon surrounding the ICPC detector, including the bore-hole with a volume $V = 43.34 \text{ cm}^3$ and ^{42}K events $N = 10^6$. Using equation 5.1, the lifetime exposure of 10^6 events of ^{42}K in 1 mm liquid argon corresponds to

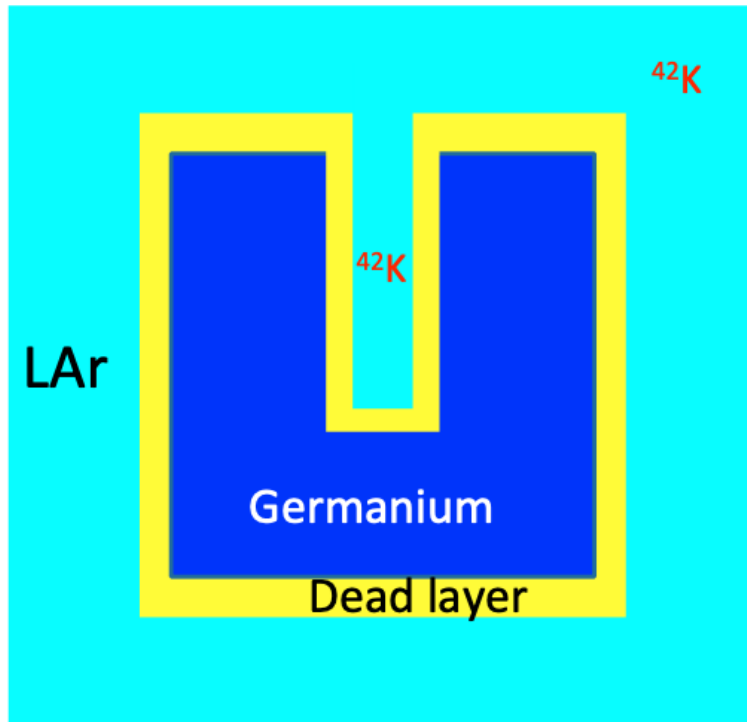


Figure 5.8: Schematics of ICPC detector with 1 mm dead layer surrounded by 1 mm liquid argon layer. The ^{42}K isotope is uniformly distributed in liquid argon.
Not drawn to scale to enhance the visibility for showing dead layer and liquid argon.

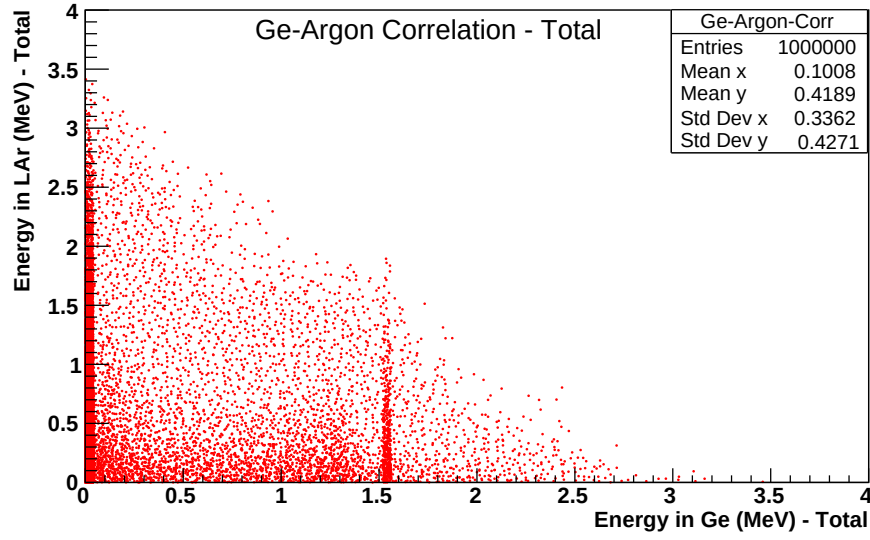


Figure 5.9: 1 mm dead layer, ^{42}K in 1 mm liquid argon: correlation of total energy in germanium with total energy in liquid argon. We have events in the region of interest. By applying a veto cut in argon, we can reduce the ^{42}K background in the germanium detector.

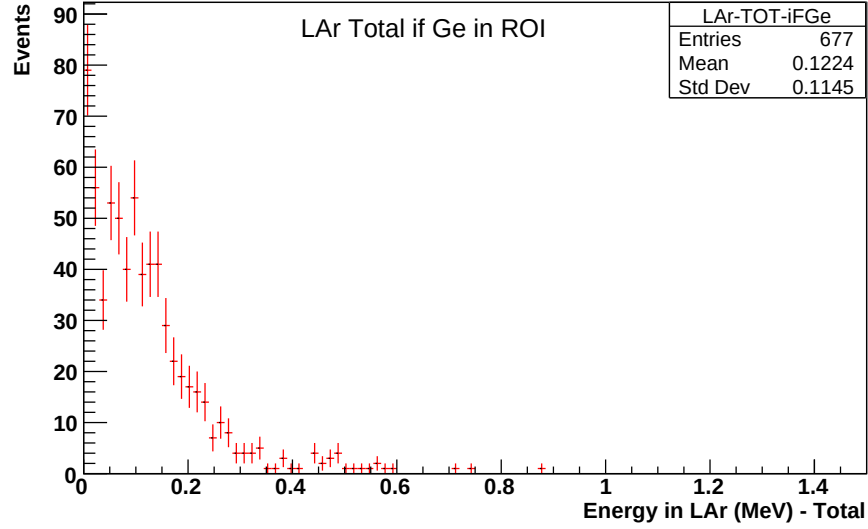


Figure 5.10: 1 mm dead layer, ^{42}K in 1 mm liquid argon: total energy in liquid argon when energy deposition in germanium ± 50 keV at $Q_{\beta\beta}$ (1989–2089 keV). Below 0.1 MeV, approximately 500 events in liquid argon in 100 keV at $Q_{\beta\beta}$. These 500 events in the liquid argon are undetected and can not be vetoed by the liquid argon veto system. Such events contribute to the ^{42}K background in the germanium detector.

$T_{Ar} = 5261$ years/detector. There are 336 detectors in LEGEND-1000 experiment with a mass of 3.02 kg per detector and the total mass of 1 ton. Therefore the simulated lifetime for LEGEND-1000 is $\sim 5261/336$ years ~ 15 years. We have approximately 500 counts of ^{42}K from 1 mm layer of liquid argon in the 100 keV window (± 50 keV) in germanium detector at $Q_{\beta\beta}$ corresponds to background index ($\mathcal{B}$) for LEGEND-1000 is estimated as $\mathcal{B} \sim 500 \text{ counts}/(100 \text{ keV} \cdot 1 \text{ ton} \cdot 15 \text{ years}) \sim 0.33 \text{ cts}/(\text{keV} \cdot \text{t} \cdot \text{yr})$. The projected background goal of LEGEND-1000 for ^{42}K is $\mathcal{B} \sim 0.6 \text{ cts}/(\text{keV} \cdot \text{t} \cdot \text{yr})$ [44]. The detector with 1 mm dead layer and 1 mm layer of liquid argon are within the background goal.

In conclusion, a thicker dead layer meets the ^{42}K background goal. But correspondingly, there is a loss in sensitive germanium mass. Scenario of the reduced liquid argon volume is discussed in the next section.

5.3 ICPC detector with 0.5 mm dead layer surrounded by 0.2 mm liquid argon layer; ^{42}K is uniformly distributed in liquid argon

In this part we simulated how liquid argon thickness influences background rate. We assume that the most tight encapsulation due to the mechanical tolerances could be as small as 0.2 mm. A 0.5 mm thick dead layer is included on all surfaces of a ICPC germanium detector. Surrounding the detector 0.2 mm layer of liquid argon is filled, including the bore-hole. The schematic of the setup is shown in Figure 5.11. Radioactive ^{42}K isotopes (10^6 events) are uniformly distributed in liquid argon. We follow a similar procedure as in previous part and show only final results.

The correlation between total energy in germanium with total energy in liquid argon is shown in Figure 5.12. The total energy in liquid argon when energy deposition in germanium ± 50 keV at $Q_{\beta\beta}$ (1989–2089 keV) is shown in Figure 5.13. The simulation shows below 0.1 MeV, approximately 2000 events in liquid argon in 100 keV at $Q_{\beta\beta}$. 0.1 MeV is the estimated threshold of energy deposition detection in liquid argon. These 2000 events in the liquid argon are undetected and can not be vetoed by the liquid argon veto system. Such events contribute to the ^{42}K background in the germanium detector. In this simulation,

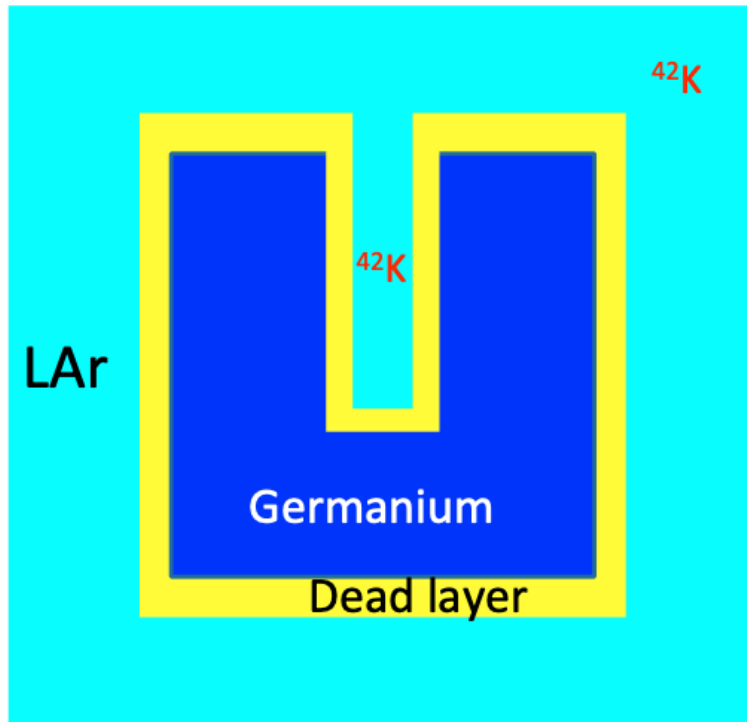


Figure 5.11: Schematics of ICPC detector with 0.5 mm dead layer surrounded by 0.2 mm liquid argon layer. The ^{42}K isotope is uniformly distributed in liquid argon.
Not drawn to scale to enhance the visibility for showing dead layer and liquid argon.

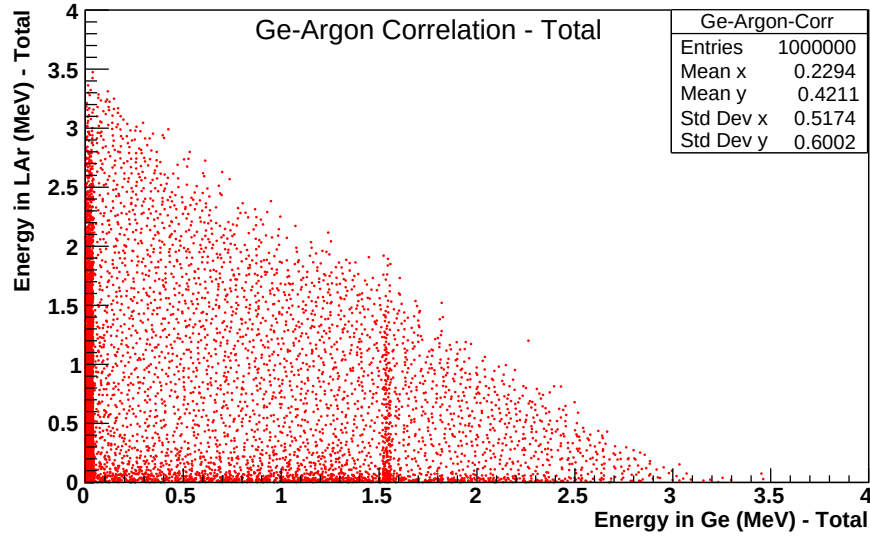


Figure 5.12: 0.5 mm dead layer, ^{42}K in 0.2 mm liquid argon: correlation of total energy in germanium with total energy in liquid argon. We have events in the region of interest. By applying a veto cut in argon, we can reduce the ^{42}K background in the germanium detector.

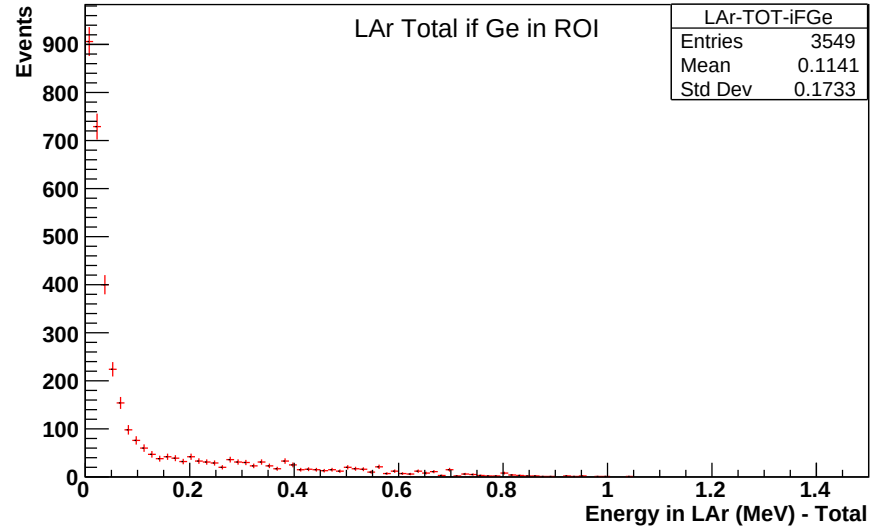


Figure 5.13: 0.5 mm dead layer, ^{42}K in 0.2 mm liquid argon: total energy in liquid argon when energy deposition in germanium ± 50 keV at $Q_{\beta\beta}$ (1989–2089 keV). Below 0.1 MeV, approximately 2000 events in liquid argon in 100 keV at $Q_{\beta\beta}$. These 2000 events in the liquid argon are undetected and can not be vetoed by the liquid argon veto system. Such events contribute to the ^{42}K background in the germanium detector.

0.2 mm layer of liquid argon surrounding the ICPC detector, including the bore-hole with a volume $V = 11.98 \text{ cm}^3$ and ^{42}K events $N = 10^6$. Using equation 5.1, the lifetime exposure of 10^6 events of ^{42}K in 0.2 mm liquid argon corresponds to $T_{Ar} = 19033 \text{ years/detector}$. There are 336 detectors in LEGEND-1000 experiment with a mass of 3.02 kg per detector and the total mass of 1 ton. Therefore the simulated lifetime for LEGEND-1000 is $\sim 19033/336 \text{ years} \sim 57 \text{ years}$.

We have approximately 2000 counts of ^{42}K from 0.2 mm layer of liquid argon in the 100 keV window ($\pm 50 \text{ keV}$) in germanium detector at $Q_{\beta\beta}$ corresponds to background index ($\mathcal{B}$) for LEGEND-1000 is estimated as $\mathcal{B} \sim 2000 \text{ counts}/(100 \text{ keV} \cdot 1 \text{ ton} \cdot 57 \text{ years}) \sim 0.35 \text{ cts}/(\text{keV} \cdot \text{t} \cdot \text{yr})$. The projected background goal of LEGEND-1000 for ^{42}K is $\mathcal{B} \sim 0.6 \text{ cts}/(\text{keV} \cdot \text{t} \cdot \text{yr})$ [44]. The detector with 0.5 mm dead layer and 0.2 mm layer of liquid argon achieve background lower than the goal.

In conclusion, a reduced liquid argon volume surrounding the detector meets the ^{42}K background goal. For this purpose, we need very tight custom build encapsulation to each germanium detector. Simulation results of the above three cases are summarized in Table 5.1.

R&D efforts to build low-background materials for encapsulation and detector components are discussed in the next chapter.

Table 5.1: Summary of ^{42}K background simulation results of the three scenarios. The ^{42}K background index is estimated at ± 50 keV at $Q_{\beta\beta}$. Thicker dead layer detectors meet the LEGEND background goal but significant loss in sensitive germanium mass. Reduced liquid argon volume surrounding the detector also meets the background goal but requires tight encapsulation.

Dead layer [mm]	Liquid argon layer [mm]	^{42}K background index [cts/(keV·t·yr)]	Achieve LEGEND goal ~ 0.6 cts/(keV·t·yr)
0.5	1.0	~ 1.33	No
1.0	1.0	~ 0.33	Yes, but with significant loss in sensitive germanium mass
0.5	0.2	~ 0.35	Yes, but tight encapsulation

Chapter 6

R&D of 3D-Printable Low-Background Materials

“The clean plastics program investigates new sources of scintillating plastics where structural integrity is needed or where advances in additive manufacturing techniques would allow the convenience of custom, on-demand printing of transparent, and potentially scintillating, plastic parts to take the place of optically passive materials” – LEGEND Collaboration

Suppression of ^{42}Ar and its decay product ^{42}K is one of the main goals of the LEGEND-1000 experiment. One technique to mitigate ^{42}K background is by enclosing the germanium detector with low-radioactive (radiopure) material, ideally 3D-printable. LEGEND-1000 is aiming to 3D-print the encapsulation because (i) all germanium detectors are different in shape, so we need to customize the encapsulation design for each detector, and (ii) to eliminate the radioactive contamination requires a hands-free environment.

Encapsulation material could potentially reduce the ^{42}Ar background by absorbing the ^{42}K isotope before it can reach the sensitive volume of the germanium detector. The material used to encapsulate the germanium detectors ideally should have the following properties: (i) low-radioactivity, (ii) low-outgassing, (iii) scintillating, (iv) cryogenic stability, (v) 3D-printability, and (vi) mechanical stability.

In collaboration with the LEGEND group of ORNL, my work focuses on the research and development of the additive manufacturing (3D-printing) of low-background materials and

measured the outgassing properties of these materials [49]. This chapter describes the 3D-printing fabrication of low-background materials. Outgassing measurements are discussed in Chapter 7. Radioassay results are discussed in Chapter 8.

6.1 3D Printing of Radiopure Materials

Polyethylene naphthalate (PEN) is a radiopure plastic scintillating material that is currently used in LEGEND-200 as a baseplate to germanium detectors. However, optically active PEN is currently not 3D-printable. We perform R&D to build 3D-printable materials that can be used to build the encapsulation for germanium detectors as well as to build electrical connectors and detector-unit support structures in the LEGEND-1000 experiment.

We use the laser-based stereolithography (SLA) technique of building materials. In SLA 3D-printing, components are fabricated by the interaction of laser light with liquid resin. During fabrication, components are not in contact with the printer parts and or surroundings, which makes for favorable conditions for maintaining the radiopurity requirement.

We adopt the following procedure of building 3D-printable materials: (i) liquid resin, (ii) degas the liquid resin, (iii) cleanroom, (iv) laser-based stereolithographic 3D-printing, and (v) curing.

Liquid resin

Liquid photopolymer resins are thermoset materials that solidify when exposed to ultraviolet radiation. Liquid resins are a primary material for 3D-printing by stereolithographic technique. We use a Foam 2 SLA printer manufactured by Formlabs [57], and the printer can use only the list of liquid resins provided by the manufacturer. For initial test samples, we are selecting two resins: surgical guide resin and dental LT clear resin. These resins have relatively large values of tensile strength. Materials having high tensile strength are recommended for encapsulation and detector components, as these materials will have to be in a cryogenic environment for several years in a LEGEND cryostat. At room temperature both surgical guide and dental LT clear resins are in liquid form; samples are shown in Figure 6.1.



Figure 6.1: Liquid resins after degassing. Left: dental LT clear resin. Right: surgical guide resin. Credit: Thomas Ruland and Chang-Hong Yu.

Degassing

Chemicals, gases, water, volatile organics, and compounds may dissolve during the initial development of liquid resins. Such trapped molecules emanate in vacuum systems for later outgassing measurements, affect the liquid argon purity in the LEGEND cryostat, and may contribute to the background and quenching of scintillation of liquid argon. The presence of such molecules in resin must be extracted prior to 3D- printing components. At ORNL we built a setup to degas the liquid resin as shown in Figure 6.2. Liquid resin is kept in a vacuum chamber connected to a liquid nitrogen cold trap and a vacuum pump. The cold trap holds the molecular compounds released from liquid resin and prevents them from venting back to liquid resin. We upgraded the current degassing chamber so it can degas resin in a much larger quantity.

Cleanroom

After degassing, liquid resins were moved to a cleanroom located in C110 hall of the physics division building 6000. To achieve low-background requirements, we printed in a cleanroom for SLA 3D-printing. We upgraded the existing cleanroom equipped with an SLA 3D printer, sonicator, glycol cold pack, isopropyl alcohol, deionized water, and post-curing setup. The SLA 3D printer in the cleanroom is shown in Figure 6.3. To enter a cleanroom, we adopt full PPE (personal protective equipment) covering: coverall, hairnet, hood, face mask, two pairs of gloves, and boot covers.

SLA 3D-printing

In a cleanroom, we kept a laser-based stereolithographic (SLA) 3D-printer under a laminar flow unit to remove the gases, vapors, and particles surrounding the printer. This is an additional effort to meet the requirement of low-background materials. The main elements of the Foam 2 SLA 3D-printer are an ultraviolet laser, a resin cartridge, a resin tank, and a build platform.

An ultraviolet laser (wavelength 450 nm and power 250 mW) is incident on galvanometers which reflect the photons in the x-y plane. Resin tank filled with liquid photopolymer resin received from a resin cartridge attached at the back of the printer. The build platform is

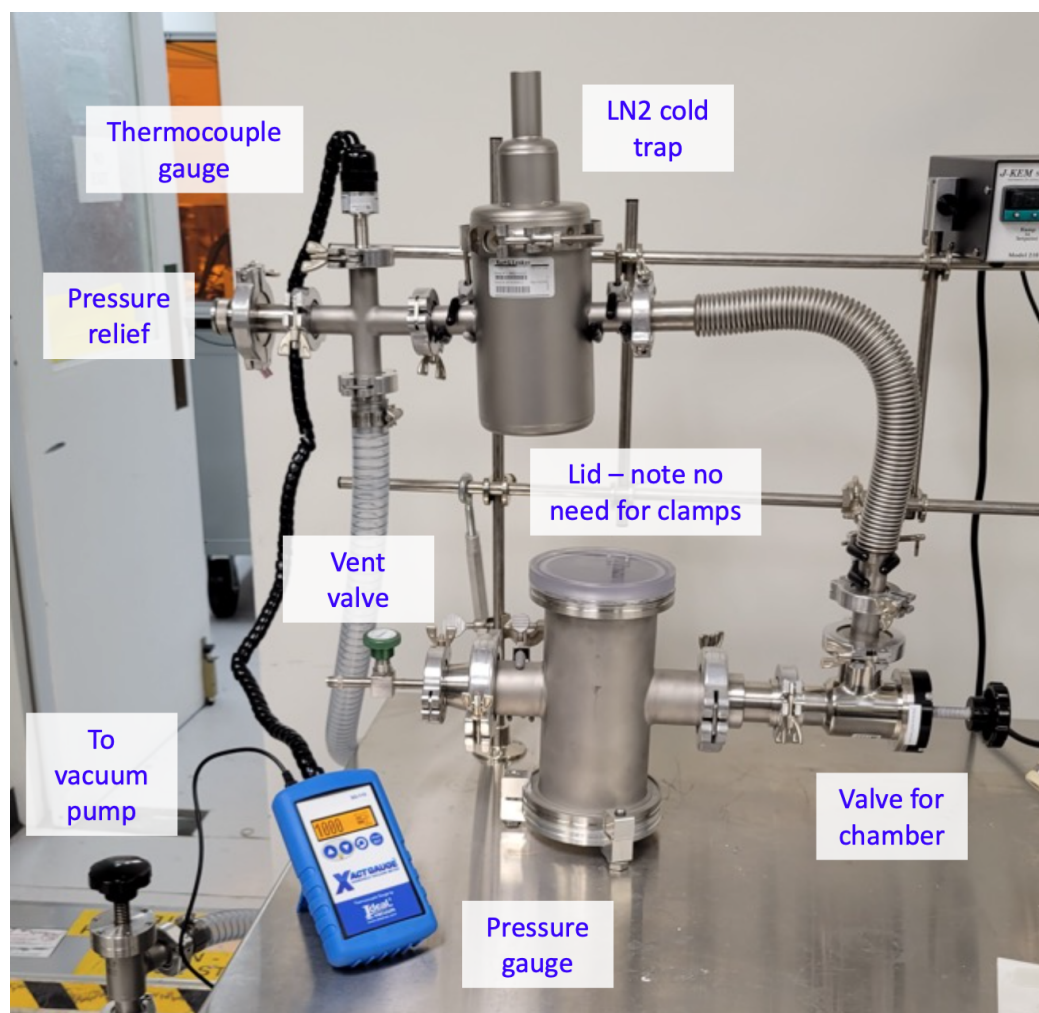


Figure 6.2: Degassing setup at ORNL. Liquid resin is kept in a vacuum chamber connected to a liquid nitrogen cold trap and a vacuum pump. The cold trap holds the molecular compounds released from liquid resin and prevents them from venting back to liquid resin. Credit: Michael Febbraro and Thomas Ruland.

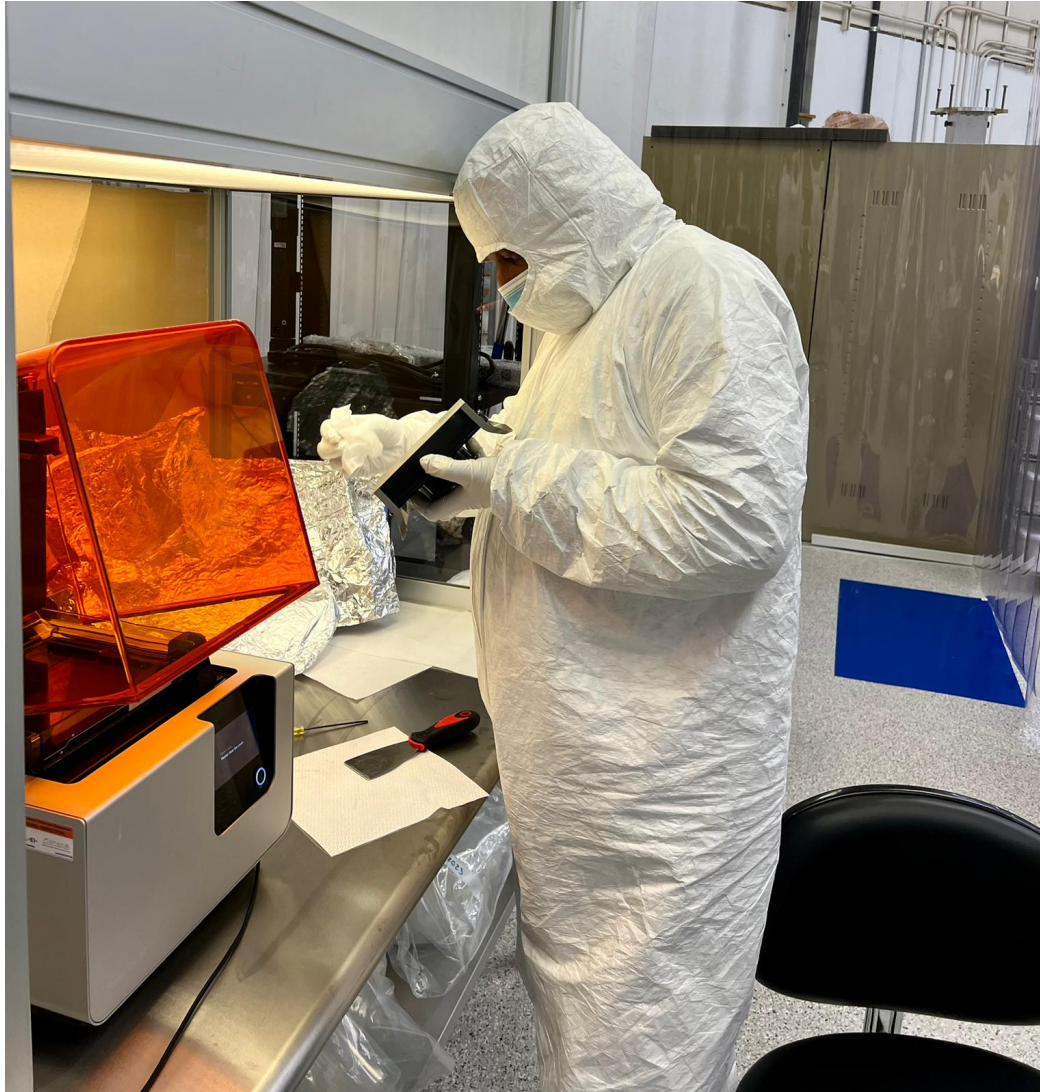


Figure 6.3: Foam 2 SLA 3D-printer kept in a laminar flow unit inside the cleanroom at ORNL. Author (Ibrahim Mirza) is in PPE covering work with the build platform of the 3D- printer. Credit: Alissa Love, Chang-Hong Yu and Cabot-Ann Christofferson.

attached to the elevator which moves in the vertical z-axis. A schematic diagram of the SLA 3D-printer is shown in Figure 6.4. SLA 3D-printing starts with filling the resin tank with liquid resin (previously degassed). The build platform is then immersed in the resin tank. Liquid resin is cured (solidified) on the build platform by the laser irradiance. The printed object is made on the build platform. 3D geometry of the sample is created using “Autodesk Fusion” computer-aided design software. For initial testing, we build a sample of rectangular shape. The geometry file is loaded in a printer by the foam labs dedicated software called “PreForm”.

Curing

After the printing, the object is removed from the build platform. The printed sample can have some deposition of uncured or excessive resin. To remove such residues, we wash the sample by ultrasonic waves produced by a sonicator, an ultrasonic processor. Washing is also performed in the cleanroom.

A stainless steel beaker consists of a triply leached plastic bag filled with isopropyl alcohol (IPA). The printed sample is now immersed in IPA. The entire beaker is placed onto a sonicator. The sonicator is filled with deionized (DI) water. DI water is high-purity water with no mineral ions; this is an additional step to ensure the low-background requirement. DI water heats during the sonication process, and such heat can propagate towards the 3D-printed sample, which could melt the sample. Therefore, two glycol cold packs are placed inside the DI water. A schematic of the ultrasonic washing setup is shown in Figure 6.5. After 20 minutes of sonication, printed parts are removed from IPA and hold it in free space until the IPA evaporates from the sample.

After the ultrasonic washing, a printed sample is placed in a curebox for post-curing. Foam labs provides a curbox called *Form Cure*, a chamber that emits thermal radiation (up to 100 °C) and ultraviolet light (LED wavelength 405 nm). A sample made of surgical guide resin is post-cured at 60 °C for 30 minutes, and a sample made from dental LT clear resin is post-cured at 60 °C for 60 minutes. Post-cured 3D-printed sample (tile) from surgical guide resin (dimension: 3 cm × 3 cm × 0.4 cm) is shown in Figure 6.6 left, and sample (tile) from dental LT clear resin (dimension: 3 cm × 3 cm × 0.4 cm) is shown in

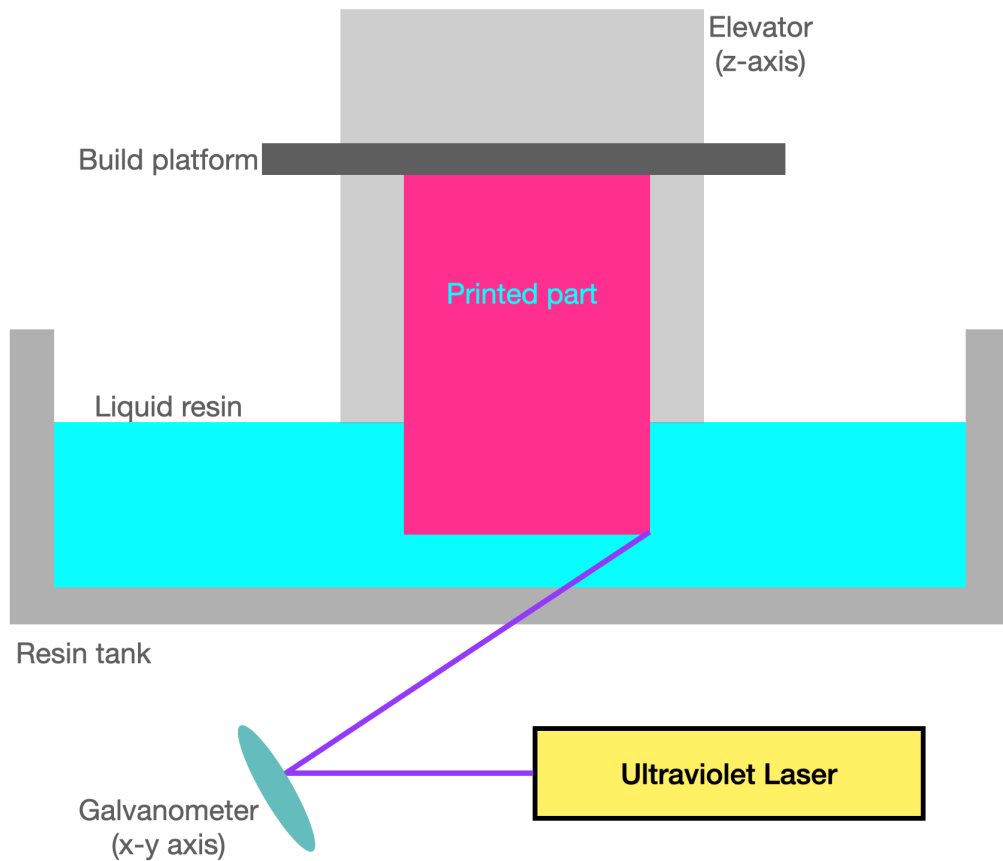


Figure 6.4: Schematic diagram of SLA 3D-printer. Ultraviolet laser incident on galvanometers, which reflects in the x-y plane. The resin tank holds photopolymer liquid resin. The build platform is attached to the elevator which moves in the vertical z-axis. Liquid resin is cured on the build platform by the laser irradiance.

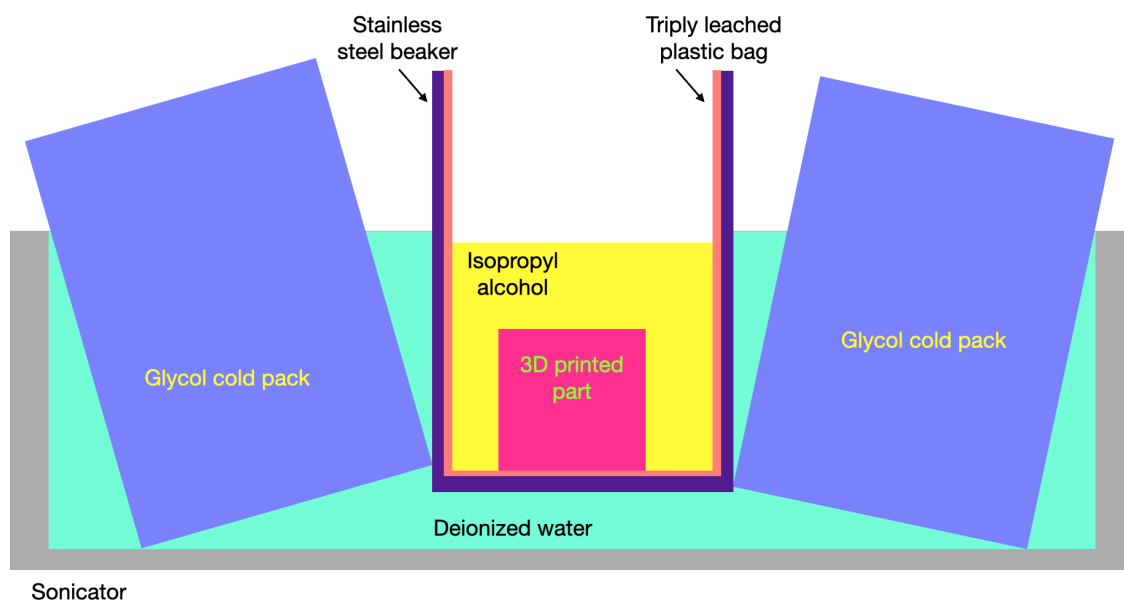


Figure 6.5: Schematic of the ultrasonic washing setup. The printed sample is immersed in isopropyl alcohol. The sonicator is filled with deionized water. *Not drawn to scale to enhance the visibility of components.* Credit: Thomas Ruland and Chang-Hong Yu.

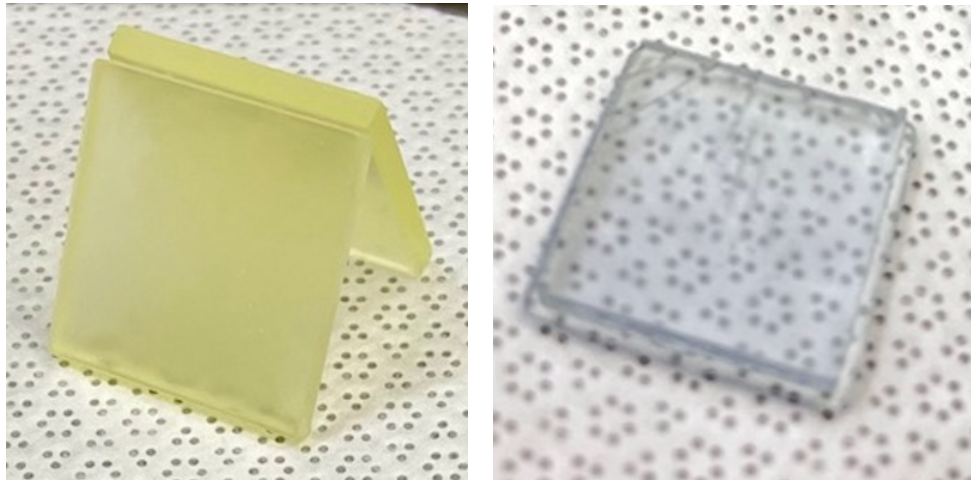


Figure 6.6: Post-cured 3D-printed samples. Left: tile of surgical guide resin. Right: tile of dental LT clear resin, each dimension $3\text{ cm} \times 3\text{ cm} \times 0.4\text{ cm}$. Credit: Thomas Ruland and Chang-Hong Yu.

Figure 6.6 right. We observe that a sample from dental LT clear resin is relatively more optically transparent than the sample from surgical guide resin. Samples are handled with nitrile gloves and kept in an airtight triply leached bag. Printed samples are tested for outgassing measurement, discussed in a next chapter. ICP-MS and gamma counting measurements of samples are discussed in Chapter 8.

3D-Printing of LEGEND-1000 Parts

CAD geometry of the LEGEND-1000 experiment is developed by the collaboration. Small detector components include fiber-holding arches and PEN insulators. To test the functionality of the 3D-printing with LEGEND-1000 parts, we start to print small components. Fiber-holding arches and PEN insulators are printed with surgical guide resin in a cleanroom. Parts are printed with a surgical guide resin. The full procedure of degassing, 3D printing, and post-curing discussed in the previous section is followed. The 3D-printed fiber-holding arch is shown in Figure 6.7 left, and the PEN insulator is shown in Figure 6.7 right.

We developed procedures and equipment to build ultra-clean 3D-printable materials. To further reduce the background, encapsulation and detector components can be 3D-printed in a glove box and handled by robotic equipment directly at the underground detector location. Printed components will be immediately transferred to the LEGEND-1000 cryostat without exposure to external background.

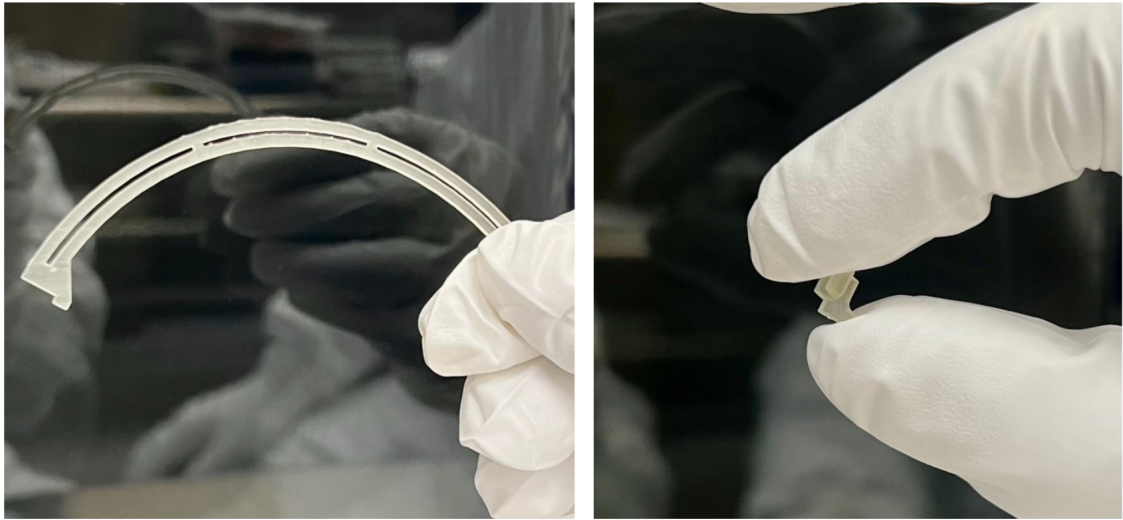


Figure 6.7: 3D-printed LEGEND-1000 prototype parts made of surgical guide resin. Left: fiber-holding arch. Right: PEN insulator. Credit: Matthew Busch and Chang-Hong Yu.

Chapter 7

Outgassing of 3D-Printed Low-Background Materials

LEGEND-1000 is an ultra-low background experiment. The presence of ^{42}Ar in liquid argon and its decay progeny ^{42}K could hide the $0\nu\beta\beta$ decay signal. Encapsulation of germanium detectors is one of the techniques under consideration to mitigate the ^{42}K background. Material used to build encapsulation may have impurities. This could become a source of background. Based on radioactivity, impurities can be grouped into two categories: (a) radioactive impurities and (b) non-radioactive impurities. 3D-printed encapsulation material or detector components must have a lower level of both kinds of contamination.

Radioactive impurities—all materials may contain some level of radioimpurities; ^{238}U , ^{232}Th , and ^{40}K . Decay chains of such isotopes could overlap the signal region and contribute to the background. For this purpose, the radiopurity level of materials are measured. Radioassay results of our 3D-printed materials are discussed in the next chapter.

Non-radioactive impurities—during fabrication of liquid resin or in general solid material, may trap some foreign molecules. Such molecules could come off (outgas) during LEGEND operations and diffuse in a surrounding liquid argon. Such molecules may affect (i) performance of the liquid argon veto system, (ii) quenching of scintillation of liquid argon, (iii) purity of liquid argon in the LEGEND cryostat. Therefore, it is also essential to minimize the non-radioactive impurities in the material used for encapsulation and detector

components. For this purpose, we design the outgassing equipment and measure the outgassing rate of our 3D-printed samples, these are discussed in the next sections.

7.1 Outgassing rate

At atmospheric pressure, external gas molecules are normally trapped between interstitial sites in the lattice structure of liquid resin fabrication. When a 3D-printed sample is kept in high vacuum, such gas molecules are ejected from the surfaces of the bulk material; this phenomenon is termed *outgassing*. Evolution of trapped gas molecules occurs due to the large pressure difference in a high vacuum chamber. However, in practice several kinds of processes in a vacuum system can contribute towards the outgassing: (i) diffusion, (ii) permeation, (iii) backstreaming, (iv) real leaks, (v) internal leaks, (vi) vaporization, and (vii) desorption [58]. The amount of outgassing from a material is expressed in terms of outgassing rate in units of torr-liter/(cm²-sec). Various methods have been developed to measure the outgassing rates of samples, which are described in [59]. We adopt the *pressure-rate-of-rise method* to measure the outgassing rate of our 3D-printed samples. In this method, the 3D-printed sample is kept in a high vacuum chamber. As a result of outgassing, trapped gas molecules evolved and accumulated in the chamber, leading to the rise in pressure, which is measured by the mass spectrometer (residual gas analyzer) attached to the chamber. Figure 7.1 represents the typical behavior of outgassing rate as a function of time. The outgassing rate by the pressure-rate-of-rise method is defined as,

$$\text{Outgassing rate} = \frac{\text{rate of pressure rise} \cdot \text{volume of vessel}}{\text{area of sample}} \quad (7.1)$$

Outgassing rate is first measured with an empty chamber to estimate the vacuum vessel contribution to outgassing, which is subtracted from the sample value. The outgassing rate of the 3D-printed sample (Q_{sample}) is computed as,

$$Q_{\text{sample}} = \left. \frac{dP}{dt} \right|_{\text{sample in chamber}} \cdot \frac{V_{\text{chamber}}}{A_{\text{sample}}} - \left. \frac{dP}{dt} \right|_{\text{empty chamber}} \cdot \frac{V_{\text{chamber}}}{A_{\text{chamber}}} \quad (7.2)$$

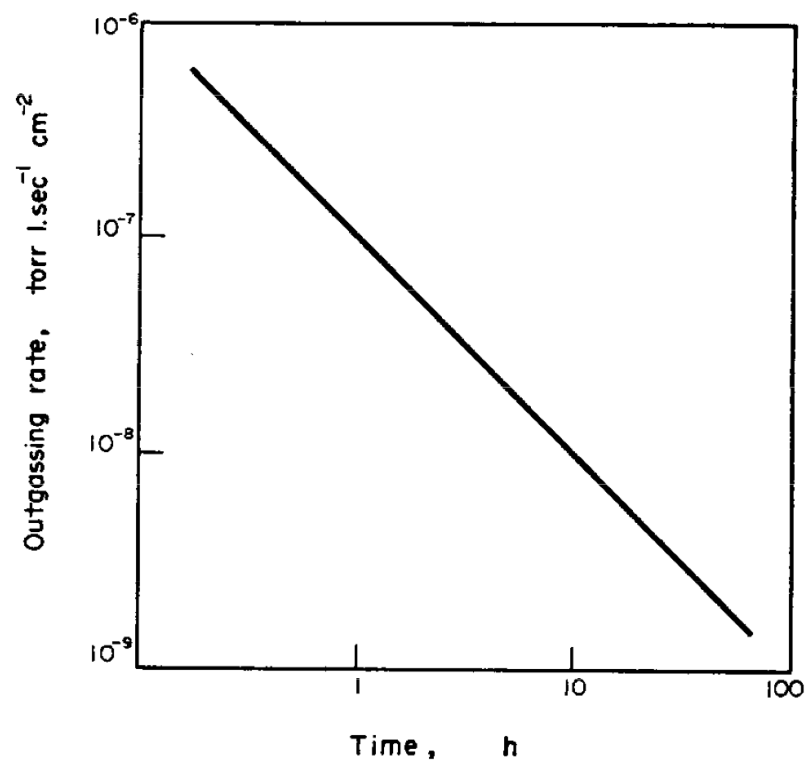


Figure 7.1: Typical log-log plot of the outgassing rate as a function of time. Credit [59].

Where P represents the pressure in units of Torr, dP/dt is pressure rise as a function of time t in units of sec, V is the volume of the chamber in units of cm^3 , and A is the surface area of the sample or chamber in units of cm^2 . Equipment used to measure outgassing rate is described in the next section.

7.2 Apparatus for outgassing measurement

The next step after 3D-printing the sample is to measure its outgassing rate. This testing does not need a cleanroom, but the samples are carefully handled using nitrile gloves while transferring to a vacuum chamber. Outgassing data reflects the cleanliness of the sample primarily with respect to non-radioactive contaminants. The 3D-samples having a lower outgassing rate qualify for radiopurity measurements. For this purpose, UTK and ORNL jointly commission the apparatus to measure the outgassing rate of 3D-printed samples.

I was involved with the team for commissioning the apparatus to measure the outgassing rate of low-background materials. We developed this facility in the physics division at ORNL. Initially we utilized the existing vacuum system built for different purposes. We prepared this vacuum system for outgassing measurements as shown in Figure 7.2. We measured outgassing rates of various samples from this setup. This vacuum assembly was not optimized primarily for outgassing measurements, as vacuum chambers are too big and cumbersome to operate, it does not have good controls, and take a long time to achieve high-vacuum. Therefore, we required a dedicated facility for outgassing measurements of our 3D-printed samples.

We have gained experience in vacuum system designing and understand the requirements specific for outgassing measurements of 3D-printed materials. For this purpose, we modified the previous vacuum assembly and optimized it primarily for outgassing rate measurements; the upgraded facility is shown in Figure 7.3. The main components of our outgassing setup are the following:

- (i) Stainless steel vacuum chamber (*vessel to hold high-vacuum*)
- (ii) Roughing pump (*for low-vacuum*)
- (iii) Turbomolecular pump (*for high-vacuum*)
- (iv) Thermocouple gauge (*pressure measurement in low-vacuum*)

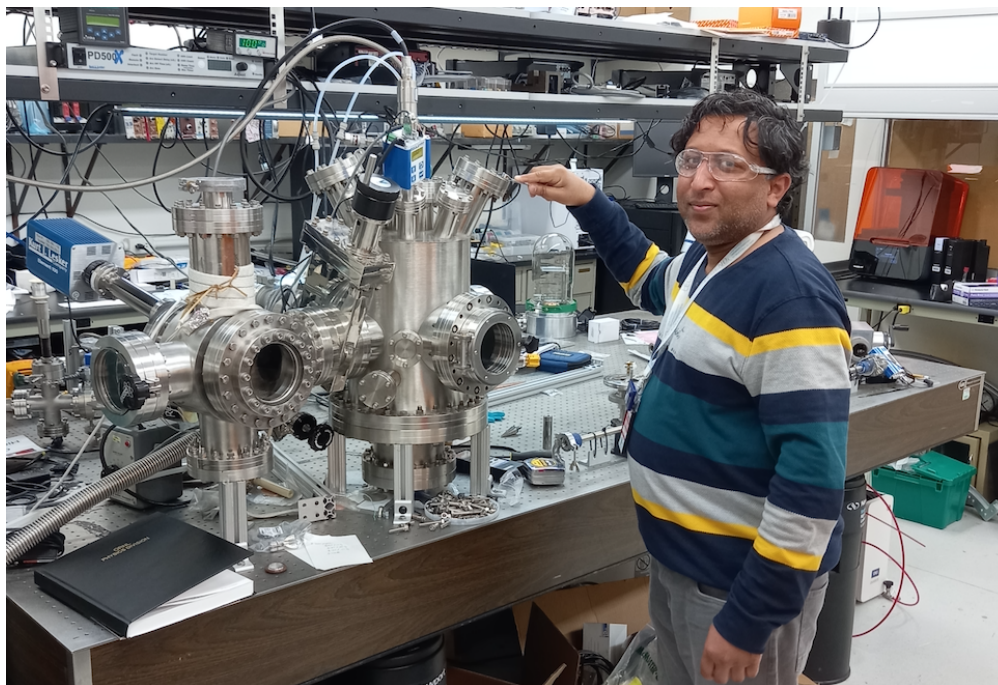


Figure 7.2: First vacuum chamber for outgassing measurements.
Credit: Michael Febbraro, Toby King, Chang-Hong Yu and Yuri Efremenko.



Figure 7.3: Left: front view of the upgraded vacuum chamber optimized for outgassing measurement. Right: side view of chamber; residual gas analyzer is connected at the back of the vessel. Credit: Sean Hollander and Chang-Hong Yu.

- (v) Ionization gauge (*pressure measurement in high-vacuum*)
- (vi) RGA (residual gas analyzer) for *partial-pressure measurement & characterization of outgassed molecular species*
- (vii) Gate valve (*isolate the chamber from pumping*)
- (viii) Pressure relief valve (*prevent from overpressure*)
- (ix) Quick-access door with viewport (*inserting the 3D-printed sample in chamber*)
- (x) Stainless steel mesh (*sample holder in a vessel*)
- (xi) Data acquisition software (*spectral analysis and outgassing data collection*)

Supplementary equipment used in our vacuum setup are:

- (xii) Argon gas (*back-filling to prevent atmospheric gas entry while opening the chamber*)
- (xiii) Heat tape (*baking the vacuum chamber, especially to evaporate water*)
- (xiv) Helium gas (*leak checking the vacuum chamber*)
- (xv) Helium leak detector (*leak checking the vacuum chamber*)

The vacuum chamber is assembled with components including flanges clamped with copper gaskets and a quick door with an elastomer O-ring. The chamber is connected with two vacuum pumps (roughing and turbomolecular), two pressure gauges (thermocouple and ionization), and a residual gas analyzer. Using the quick-access door, the sample is placed in the vacuum chamber. To open this door, we first backfill the chamber with pure argon gas, which prevents the entry of external atmospheric gas and moisture into the chamber, and then the sample is immediately placed in the chamber. In order to remove the traces of water vapor hidden in the interior of the chamber, we baked the vacuum vessel by attaching it to the heat tape. For checking the leakages in the vacuum chamber, we sprayed the helium gas, and the same residual gas analyzer is used for leak detection.

The 3D-printed sample is placed on stainless steel mesh (a sample holder) inside a vacuum chamber. The roughing pump started at atmospheric pressure (760 torr) and created a vacuum of 10^{-3} Torr; pressure in the chamber as directly measured by a thermocouple gauge. Then the turbomolecular pump started from 10^{-3} Torr to make a high-vacuum of 10^{-8} Torr; pressure inside the chamber was directly measured by the ionization gauge. Once the high-vacuum was achieved, the vacuum chamber was ready for outgassing data collection from the sample or background (empty chamber) by the attached residual gas analyzer, which is discussed in the next section.

7.3 Residual Gas Analyzer

To measure the outgassing of 3D-printed objects from the pressure-rate-of-rise method, we first maintained the chamber at a high-vacuum of 8×10^{-8} Torr connected with a residual gas analyzer (RGA). This instrument provides the partial-pressure contributed by the outgassed and residual gases present in the vacuum chamber and simultaneously identifies the type of molecular species [60]. The RGA consists of three main components:

(i) Ionizer: the first stage of the RGA is the ionization of outgas molecules. Ionizer consists of a filament (cathode) and a grid cage (anode). Electrons are thermally emitted from the hot filament and accelerated towards the anode grid, gaining high kinetic energy. When the molecules arrive between the filament-grid cage region (where high-energy electrons are crossing), gas molecules receive a strong impact from passing high-energy electrons, and molecules become ionized (positive ions of charge $+e$). Ions are then extracted from the ionization regions by the focusing plate. Ion production is proportional to the gas density or partial-pressure.

(ii) Mass spectrometer: the second stage of the RGA is the mass separation of ions. By electric fields, ions are injected into the quadrupole mass analyzer. A mass filter consists of two pairs of rods that are connected to a combination of direct and alternating voltage. For a constant radio-frequency and quadrupole radius, the mass of the ions is proportional to radio-frequency voltage. This allows quadrupoles to select specific charge-to-mass ratios of ions to pass through them.

(iii) Ion detector: the third stage of the RGA is the detection of ions. After exiting the mass spectrometer, ions arrive in the Faraday cup detector. Incident ions are then neutralized after striking the Faraday cup. As a result, current flows in the connected electrode, which is equal to the incident current produced by the ion (I_{ion}).

The main mechanism of the RGA is represented in Figure 7.4. Partial-pressure (P_{ion}) of the molecules (ion) is proportional to the I_{ion} . The RGA measures P_{ion} by the equation,

$$I_{\text{ion}} = S \cdot P_{\text{ion}} \quad (7.3)$$

where S is the sensitivity of the ionizer, $S = 3 \times 10^{-4}$ Amps/Torr.

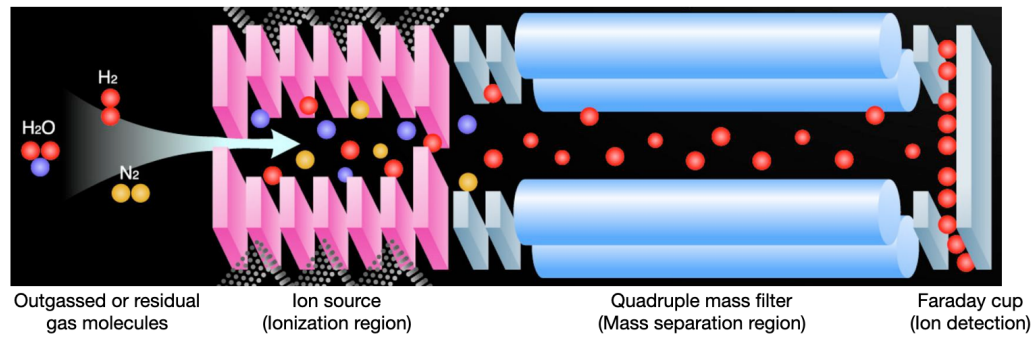


Figure 7.4: Main functionality of residual gas analyzer (RGA). Outgassed molecules are coming from the left side, which are ionized by the collision with high-energy electrons emitted by the filament; ions are then separated in mass-to-charge ratio by a quadrupole mass spectrometer and then detected by a Faraday cup. RGA converts ion current into partial-pressure for each mass. Credit [61].

The ion current or partial-pressure signal for a given mass range (1-50 amu) is then readout by the attached electronics box. We use software provided by the manufacturer (Kurt J. Lesker) for data acquisition. The RGA monitor provides a mass spectrum as a function of partial pressure (produced by the ionization current) in a real-time. Partial-pressure peaks represent the molecule corresponding to the atomic mass unit (u), as the peaks at masses 2 u, 18 u, 28 u, and 44 u are identified as hydrogen gas, water vapor, nitrogen gas, and carbon dioxide, respectively, as shown in Figure 7.5. In one scan, the RGA sweeps the entire mass range (1-50 amu) and records the ion current or partial-pressure for each mass. Data of all scans is stored. RGA specifications and parameters used in our outgassing data acquisition are listed in Table 7.1. Analysis of outgassing rate is discussed in the next section.

7.4 Outgassing rate of 3D-printed materials

We perform outgassing rate analysis as follows: The stored data file recorded by the RGA consists of partial-pressure values of each mass species as a function of time of measurement for all scans. To compute the outgassing rate of the bulk 3D-printed sample, we require total pressure. Using Dalton’s law of partial pressure, total pressure (P) is the sum of partial-pressure of all molecules (ions),

$$P = \sum_{\text{ion}=1}^{50} P_{\text{ion}} \quad (7.4)$$

Rise in pressure for surgical guide resin sample, dental LT clear sample, and background are shown in Figure 7.6. During the first two minutes, the dominant contribution to outgassing is from the 3D-printed sample, the stainless steel vessel (large outgassing of hydrogen gas), and the residual gases (mainly nitrogen gas and traces of water vapor, etc.) that remain in the chamber. Slope (dP/dt) is determined for the first two minutes, where outgassing from the sample is prominent. A similar slope is determined for the empty chamber. The volume of the chamber is estimated as 12.52 Liter with a surface area of 5738.17 cm². The 3D-printed samples have dimensions is 3 cm × 3 cm × 0.4 cm with a surface area of 24 cm². The outgassing rate of the sample is calculated by substituting all values in Equation 7.2. Results of the outgassing rate of 3D-printed samples are listed in Table 7.2.

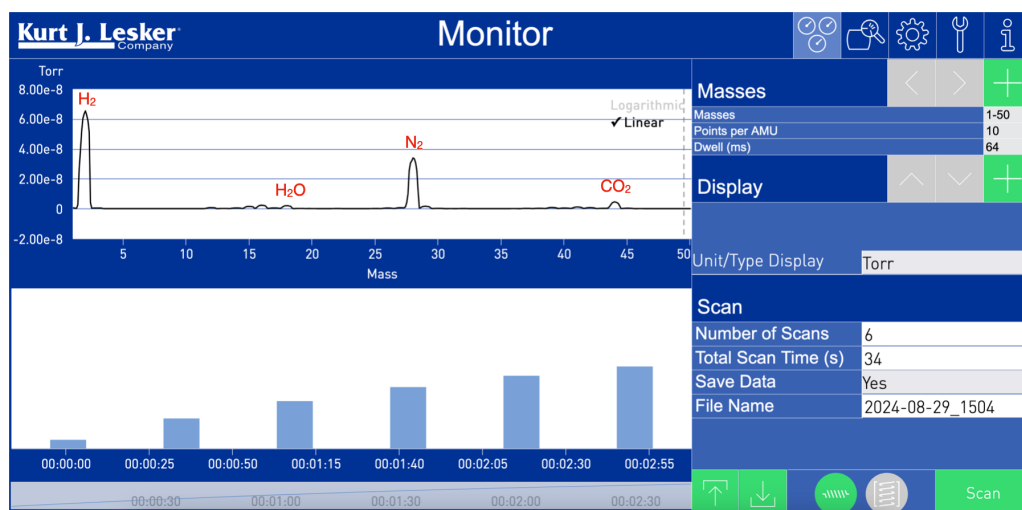


Figure 7.5: Residual gas analyzer monitor. Mass spectrum obtained from the 3D-printed tile (surgical guide resin) in the vacuum chamber, including the contributions from vessel and residual gases present in the chamber. Mass scale (horizontal) is in atomic mass units (u). Peaks at the masses 2 u, 18 u, 28 u, and 44 u are identified as hydrogen gas, water vapor, nitrogen gas, and carbon dioxide, respectively. For all measurements with an upgraded chamber, in our measurements RGA input parameters: masses = 1-50 amu, points per amu = 10, dwell time = 64 milliseconds, numbers of scans = 6, and total scan time (for one scan) = 34 seconds.

Table 7.1: Residual gas analyzer specifications and parameters used in our outgassing data measurements. Prior pressure in our vacuum chamber 8×10^{-8} Torr.

Parameter	Value
Detector type	Faraday cup
Mass range	1 - 50 amu
Points per amu	10
Dwell time	64 milliseconds
Number of scans	6
Scan time	34 seconds
Maximum operating pressure	5×10^{-4} Torr
Minimum detectable partial-pressure	2.6×10^{-12} Torr
Sensitivity	3×10^{-4} Amps/Torr

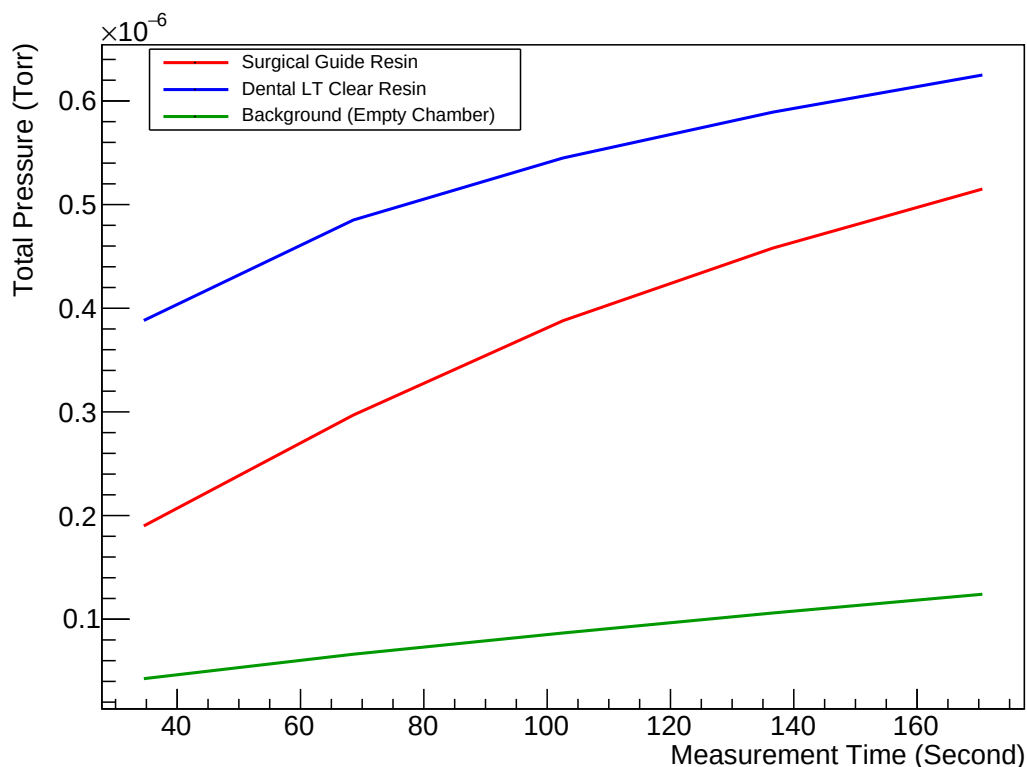


Figure 7.6: Total pressure as a function of time during outgassing. Pressure contribution to outgassing of the surgical guide resin sample is shown in red, the dental LT clear resin sample is shown in blue, and the background (empty chamber) is shown in green. During the measurement, the chamber was not pumped, and the partial-pressure in the chamber was collected by the residual gas analyzer. Total pressure is calculated as the sum of all the partial-pressure contributions by the molecular species present in the chamber. During the first two minutes, the dominant contribution to outgassing is from the 3D-printed sample, the stainless steel vessel (large outgassing of hydrogen gas), and the residual gases (mainly nitrogen gas and traces of water vapor, etc.) that remain in the chamber. Prior to each measurement, the chamber is maintained at a high-vacuum of 8×10^{-8} Torr.

Table 7.2: Outgassing rate of 3D-printed materials. Sample values are background (empty chamber) subtracted. Both samples were printed in a cleanroom, and liquid resin was degassed prior to printing. We made multiple measurements of the samples. Both 3D-printed samples show low outgassing rates. The difference in outgassing rates of dental LT clear and surgical guide resin is small and within measurement uncertainty. To reduce systematics, we show comparisons in outgassing rates, and we select samples with the same surface area, measured in the same vacuum chamber, and same prior pressure. Precise measurement uncertainties could not be deduced due to various reasons. The outgassing rate of surgical guide resin is the average of the two measurements on the same day in identical conditions. Knowing that the two measurements on the same day with identical conditions can yield different results, this just demonstrates that the difference we measured between the dental clear and surgical guide is within measurement uncertainty. The outgassing rate of the samples is for comparison only, because we only estimated the volume of our vacuum chamber due to its irregular shape.

3D-printed sample	Surface area [cm ²]	Outgassing rate [Torr-Liter/(cm ² -sec)]
Surgical guide resin	24	1.397×10^{-9}
Dental LT clear resin	24	1.207×10^{-9}
Background	5738.17	1.853×10^{-12}

Data shows the difference in outgassing rates of dental LT clear resin and surgical guide resin is small and within measurement uncertainties. To reduce systematics, we show comparisons in outgassing rates, and we select samples with the same surface area, measured in the same vacuum chamber, and same prior pressure. Precise measurement uncertainties could not be deduced due to various reasons. The RGA has $\sim 2\%$ accuracy for outgassing rate measurement.

To estimate the error in the outgassing rate of the 3D-printed sample or background, we ran the RGA for longer scans. The partial-pressure as a function of time of each molecular species between 1–50 amu was collected by the RGA. The total pressure is calculated by using the equation 7.4 and plotted against total RGA scan time.

As a result of outgassing, the total pressure rises linearly as a function of time for initial RGA scans and reaches a plateau as the scans progress. The total pressure is predicted by fitting a sixth-order polynomial to the data corresponding to each scan. The corrected pressure (measured pressure – predicted pressure) is calculated. The standard deviation in the corrected pressure is estimated for the plateau region. This error is used in computing the slope (dP/dt) during the initial RGA scans (a dominant outgassing period for the first two minutes after stopping pumping). This slope is substituted in equation 7.2 to obtain the outgassing rate of the sample.

By request of the LEGEND collaboration, we also measured the outgassing rates of various extra samples: PEN, ELJEN-108, Rogers, Cirlex, and G-10. Among them, PEN is known as a good plastic scintillator currently used in the LEGEND-200 experiment. Rogers, Cirlex, and G-10 were measured by request of collaboration because those materials can be used inside liquid argon cryostat. To show comparison in outgassing rates and reduce the systematics, we selected samples with nearly the same surface area. These materials are shown in Figure 7.7 and Figure 7.8. Outgassing rates of these materials are listed in Table 7.3. We did not have exact specs, but from previous experiments we know that the PEN is good enough (no degradation in liquid argon), so we used it as a reference point for 3D-printed samples.

After the outgassing measurements, the 3D-printed samples were sent for radioassay measurements, which are discussed in the next chapter.

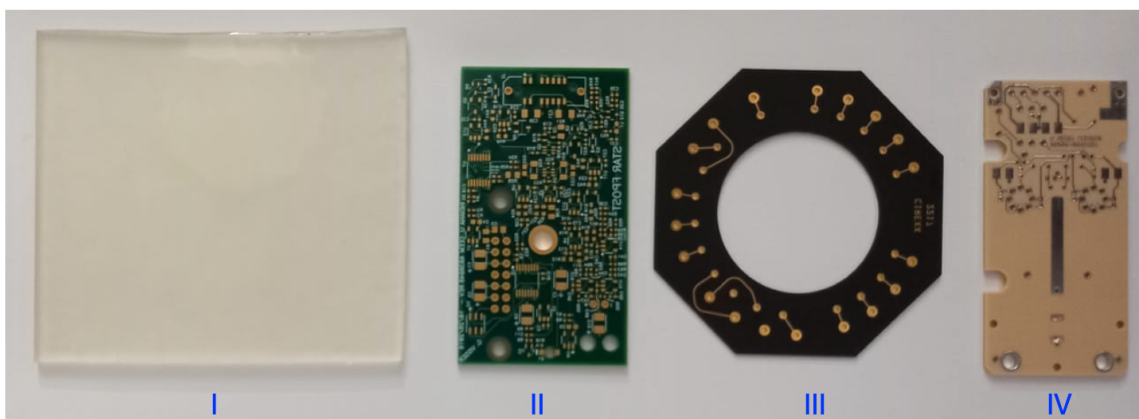


Figure 7.7: Miscellaneous vacuum materials. Figure I is our first 3D-printed sample (surgical guide resin). This resin was not degassed and not printed in a cleanroom. Figure II is Rogers printed circuit board. Figure III is Cirlex material. Figure IV is a G-10 printed circuit board. Outgassing rates of these materials are listed in Table 7.3.

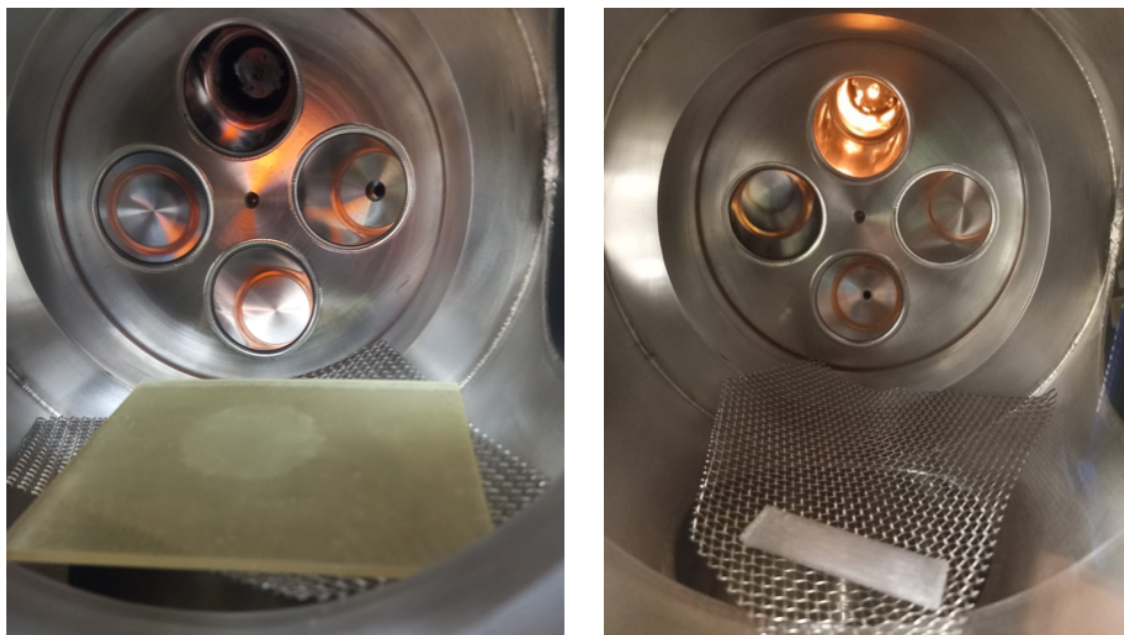


Figure 7.8: Miscellaneous materials in a vacuum chamber. Left: our first 3D-printed sample (surgical guide resin). Right: ELJEN-108. Outgassing rate of ELJEN sample is listed in Table 7.3.

Table 7.3: Outgassing rate of materials. All outgassing values are background (empty chamber) subtracted. Background outgassing rate typically varies from $(1.7-3.623) \times 10^{-12}$ torr-Liter/(cm²-sec). Except ELJEN-108 sample, all materials were measured in an old vacuum chamber.

Material	Surface area [cm ²]	Outgassing rate [Torr-Liter/(cm ² -sec)]
ELJEN-108	20.98	1.077×10^{-9}
PEN	31.21	7.529×10^{-10}
Rogers RO-4003B	43.18	2.744×10^{-10}
Circlex	48.45	4.923×10^{-10}
G-10	53.02	3.267×10^{-10}

Chapter 8

Radioassay of 3D-Printed Low-Background Materials

“The material screening program ensures the radiopurity of all relevant candidate and deployed construction materials required by the experiment’s background goal through direct measurements of their radioactivity using established assay instruments and techniques.”

– LEGEND Collaboration

LEGEND-1000 is an ultra-low background experiment. To achieve the LEGEND goal of low-background, all materials used in the cryostat should have a low level of radioactive contamination. Germanium detectors are under consideration to be encapsulated with 3D-printing material to mitigate the background produced by the ^{42}K isotope present in the liquid argon. The material selected to build encapsulation should have a low concentration of radioactive impurities. To measure the radiopurity of our 3D-printing samples, we perform mass spectrometry and gamma-ray spectrometry. These radioassay techniques and results are discussed in this chapter.

All materials in general consist of some level of internal and external radioactivity, including ^{238}U , ^{232}Th , and ^{40}K . Those three elements have a very long lifetime of billion years; and are widely distributed within the Earth. In a construction material, such decay chains could become a source of background. Hence the traces of radioactive isotopes ^{238}U , ^{232}Th , and ^{40}K and their decay progeny are to be measured. The decay chains of ^{238}U and ^{232}Th

are shown in Figure 8.1. The presence of external radioisotopes could break the secular equilibrium (parent/daughter separations) that occur in both ^{238}U and ^{232}Th decay chains. The isolated presence of ^{226}Rn or ^{222}Rn in the environment would break the equilibrium of the ^{238}U decay chain, and the isolated presence of ^{210}Pb would again break the equilibrium of the ^{238}U decay chain. Similarly, the isolated presence of ^{220}Rn would break the equilibrium of the ^{232}Th decay chain. Radon progenies are deposited on surfaces on materials, called the *plate-out effect*. Radon emanation techniques are used to measure the radon contamination. The interaction of cosmic-rays with material could activate some radioisotopes, which could become a source of background or break the equilibrium of the uranium and thorium decay chains. Therefore, for the final product of encapsulation, LEGEND aims to 3D-print the encapsulation and some detector components in a glove box and handle them by robotic equipment directly at the underground detector location at LNGS. Printed components will be immediately transferred to the LEGEND-1000 cryostat. This additional practice would further reduce the background induced by the cosmic-ray interactions.

Encapsulation material, as with all detector components in the LEGEND-1000 cryostat, will undergo material screening. Depending on the parent nuclide and the broken equilibrium, these assay techniques are classified as follows:

- (i) Mass spectrometry (for parent nuclide ^{238}U , ^{232}Th , and ^{40}K)
- (ii) Gamma-ray spectrometry (γ -line from decay chain of ^{238}U and ^{232}Th nuclides)
- (iii) Neutron activation analysis (for parent nuclide ^{238}U and ^{232}Th)
- (iv) Radon emanation (for ^{220}Rn and ^{222}Rn nuclides produced in broken equilibrium in both ^{238}U and ^{232}Th decay chains)
- (v) Surface assay (for ^{210}Pb nuclide produced in broken equilibrium of ^{238}U decay chain)
- (vi) α spectrometry (α decaying nuclides in both ^{238}U and ^{232}Th decay chains)

Due to the presence of background radioisotopes, secular equilibrium can be broken. Therefore, two different radioassay techniques are required for the radiopurity measurements of the full decay chain of ^{238}U and ^{232}Th . We assayed our 3D-printed samples using mass spectrometry analysis for parent nuclide (^{238}U and ^{232}Th) identification and gamma-ray spectrometry for γ -emission from the entire decay chain of daughter nuclides.

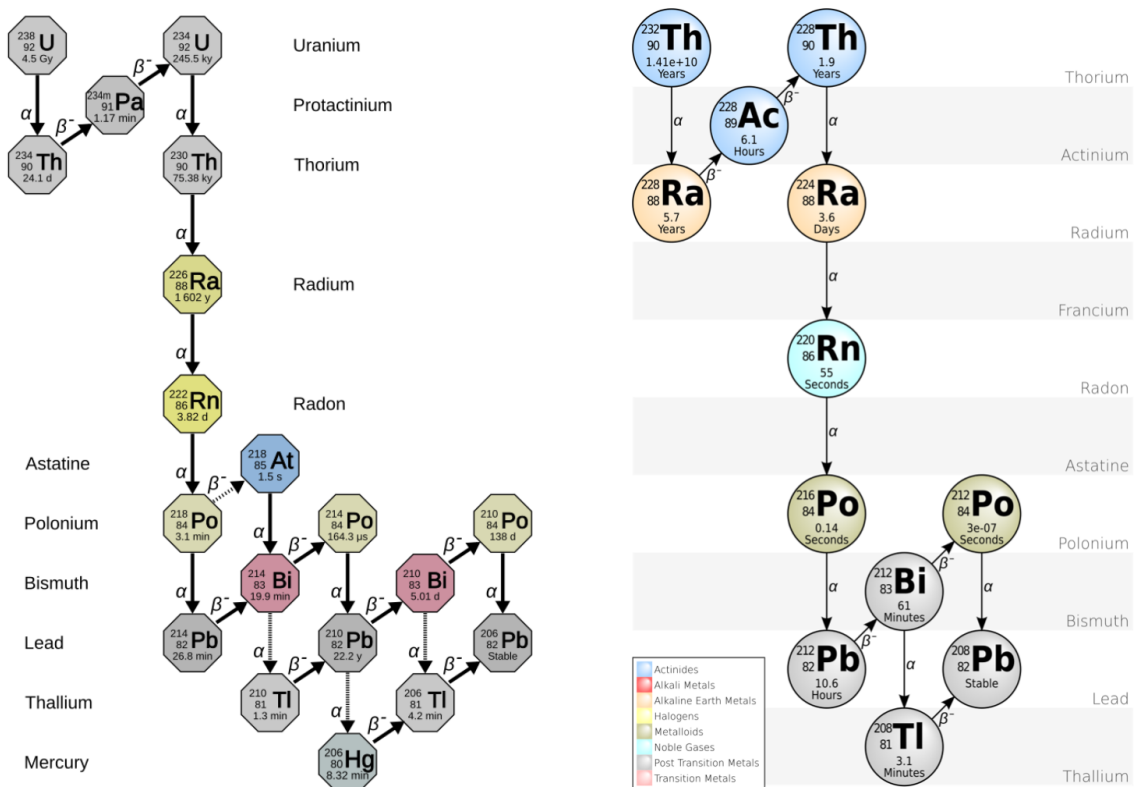


Figure 8.1: Left: ^{238}U decay chain. Right: ^{232}Th decay chain. Secular equilibrium can be broken due to the presence of external background radioisotopes. Therefore, a single radioassay technique is not sufficient to identify ^{238}U and ^{232}Th isotopes as well as their decay progeny. We are using two radioassay methods: mass spectrometry for parent nuclide measurement and gamma-ray spectrometry for the entire decay chain of daughter nuclides identification. Credit [62].

To extract the actual concentration of isotopes in a sample, careful simulation of the gamma-counting facility and efficiency are required. Therefore, we performed Monte Carlo simulations of gamma-ray spectroscopy. The radioassay results and simulations are discussed in the sections below.

8.1 Mass Spectrometry

To measure the radioactivity level mainly due to the presence of parent nuclides, ^{238}U , ^{232}Th and ^{40}K in our 3D-printed sample (surgical guide resin) we used the Radioactive Materials Analytical Lab (RMAL) at ORNL for mass spectrometry for Inductively Coupled Plasma Mass Spectrometry (ICP-MS) of our sample. ICP-MS is a destructive method of measuring radioisotopes, in which sample preparation is required; our 3D-printed sample (solid) is dissolved in liquid. ICP-MS measures the very low concentration (parts per trillion) of radioactive elements. There are several advantages of ICP-MS over gamma-ray spectrometry, including:

- (i) Measurement of ultra traces of intrinsic radioisotopes (parent nuclide ^{238}U , ^{232}Th , and ^{40}K) present in a sample simultaneously.
- (ii) High sensitivity (low detection limits) and accurate measurements.
- (iii) Measurement of low-level long-lived intrinsic radioisotopes in a specimen.
- (iv) Only a small amount of sample is required.
- (v) Liquid resin can also be directly tested prior to 3D-printing, and sample preparation work can be minimized.
- (vi) Measurements are completed within a short-period of time.
- (vii) Peak analysis of individual nuclide is not required.
- (vii) No special shape of sample, e.g., Marinelli (a specialized sample design used in gamma-ray spectrometry to maximize detection efficiency), is required.
- (ix) Measurement can be performed at the surface, and there is no need for an underground location.

However, there are several disadvantages of ICP-MS over gamma-ray spectrometry, such as:

- (i) Sensitivity only to the long lived elements (^{238}U , ^{232}Th , and ^{40}K) within the decay chain.
- (ii) It is a destructive method of assay, detector components cannot be directly tested, so only witness samples (samples developed in the same conditions) can be used.

A solid sample is dissolved in liquid and introduced in a nebulizer by the peristaltic pump (for fluid pumping). The liquid sample is sprayed into an inductively coupled plasma torch consisting of high-temperature plasma. As a result of interaction with the plasma, liquid molecules are converted into ions. After exiting, the plasma ions are guided by electric fields toward the quadrupole mass spectrometer. A mass filter, consisting of two pairs of rods that are connected to a combination of direct and alternating voltage, allows the selection of specific charge-to-mass ratios of ions to pass through them. After exiting, ions arrived in a detector consisting of an electron multiplier.

The preliminary ICP-MS results obtained from the surgical guide resin are shown in Table 8.1, and limits from the blank (background or without sample) are shown in Table 8.2. The ICP- MS data shows, for the surgical guide sample, ^{40}K presence is higher than both ^{238}U and ^{232}Th . The ^{238}U concentration is $\sim 7\%$ above the background (blank, without sample), and the ^{232}Th concentration is $\sim 21\%$ above the background (blank, without sample). A complete report of ICP-MS analysis is provided by RMAL, ORNL, and is attached in Appendix A.

An analysis report indicating “not completely broken down components” as they seen on the Figure 1 in Appendix A. This means that the sample was not completely broken down during the preparation process. Therefore, the preliminary measurements obtained from this incomplete process may differ from the final, validated results.

8.2 Gamma-ray Spectrometry

The ICP-MS technique identifies only the presence of parent nuclides (^{238}U and ^{232}Th) in the sample. External contamination could break the secular equilibrium. Therefore, we need to perform additional radioassay techniques. To get the information from the full decay chain, we require gamma-ray spectrometry or gamma-counting. This radioassay technique

Table 8.1: Preliminary ICP-MS results for surgical guide resin obtained at Radioactive Materials Analytical Lab (RMAL) at ORNL. Credit: Tamara Keever, Chang-Hong Yu, Yuri Efremenko.

Unit conversion used ^{40}K : 1 mBq/kg = 32.3 $\mu\text{g/g}$, ^{232}Th : 1 mBq/kg = 24.6×10^{-5} $\mu\text{g/g}$, ^{238}U : 1 mBq/kg = 8.1×10^{-5} $\mu\text{g/g}$.

Element/isotope	$\mu\text{g/g}$	Uncertainty	mBq/kg
^{40}K	3.88	3.88×10^{-1}	0.12 ± 0.01
^{232}Th	1.07×10^{-5}	2.15×10^{-6}	0.04 ± 0.01
^{238}U	1.70×10^{-5}	3.41×10^{-6}	0.21 ± 0.04

Table 8.2: Preliminary ICP-MS results for method blank, trace metals ashing obtained at Radioactive Materials Analytical Lab (RMAL) at ORNL. Credit: Tamara Keever, Chang-Hong Yu, Yuri Efremenko.

Element/isotope	$\mu\text{g/ml}$
^{40}K	$< 2.91 \times 10^{-2}$
^{232}Th	$< 8.37 \times 10^{-6}$
^{238}U	$< 1.58 \times 10^{-5}$

uses high-purity germanium detectors, and because of the good energy resolution of germanium, it provides detailed information of nuclides emitting γ -lines from the entire decay chain. To perform gamma-ray spectrometry, we sent our 3D-printed sample (dental LT clear resin) to the Canfranc Underground Laboratory [63].

In addition, we performed GEANT4 simulations for gamma-ray spectroscopy, which is discussed in the next section. There are several advantages of gamma-ray spectrometry over ICP-MS, including:

- (i) Energy spectra directly provide information about all the ^{238}U and ^{232}Th decay progenies.
- (ii) Good energy resolution of germanium detectors easily distinguishes the γ -lines produced by the radioactive nuclei present in the sample.
- (iii) This technique is non-destructive, so no additional sample preparation is required (detector components can be directly tested without destroying them), and after the assay, the sample can be used in the experiment.
- (iv) Large samples or detector components can also be tested.
- (v) Obtained energy spectrum could directly provide information of the broken equilibrium.

However, there are several disadvantages of gamma-ray spectrometry over ICP-MS, as:

- (i) Longer measurement time.
- (ii) Requires a large mass of sample.
- (iii) Requires an underground location and specially designed multi-layer shielding for low-background.

To measure gamma-counting in low-background conditions requires an underground location. The Canfranc Underground Laboratory is equipped with a gamma-ray counting facility specially built for ultrasensitive radioassay. This laboratory is located 780 meters deep underground in Spain, as shown in Figure 8.2. The gamma-ray spectroscopy instrument consists of a high-purity germanium detector surrounded by a thin copper wall, which is kept in a pure copper chamber filled with nitrogen gas, as shown in Figure 8.3.

To measure γ -counting from this setup, we 3D-print the sample from Dental LT clear resin in the shape of Marinelli. An inside view of this assay instrument is shown in Figure 8.4,

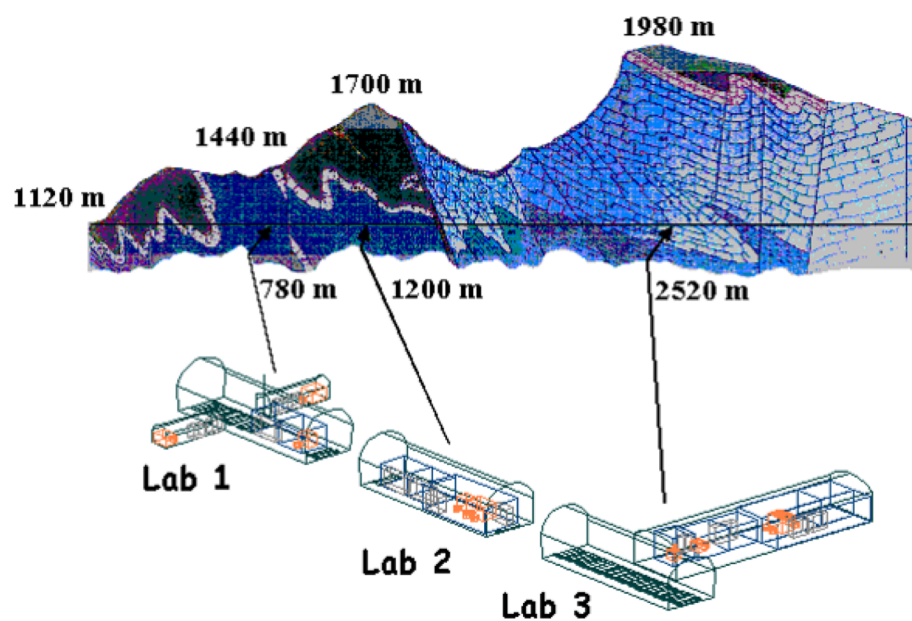


Figure 8.2: Canfranc Underground Laboratory in Spain. Credit [64].



Figure 8.3: Gamma-ray spectrometer inside the Canfranc Underground Laboratory. Credit [63].

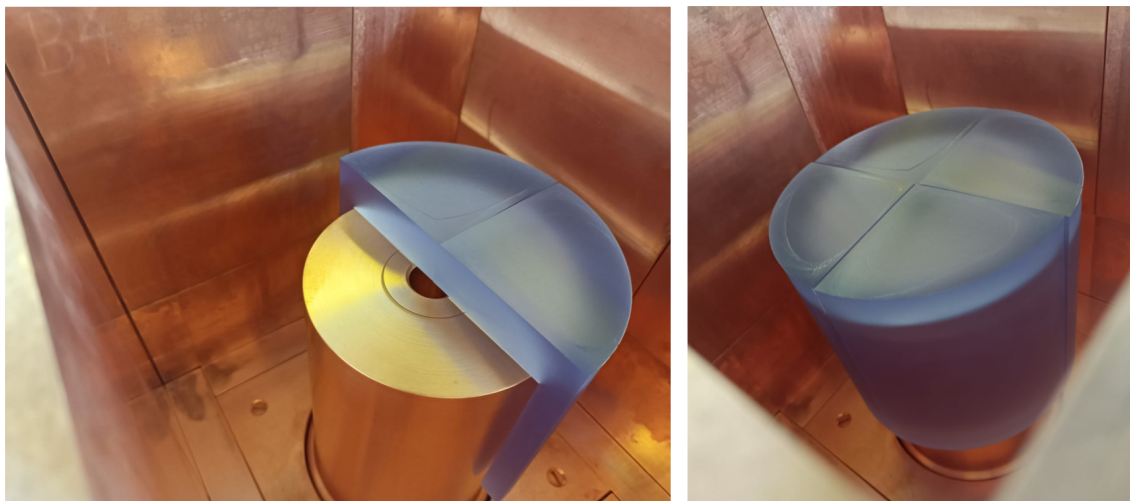


Figure 8.4: Left: 3D-printed sample (light blue) portion in gamma-ray spectrometer. Right: Fully assembled sample. This low-background, high-purity germanium gamma-ray spectrometer is located at the Canfranc Underground Laboratory in Spain. The germanium detector is located at the central region of this apparatus, surrounded by a thin copper wall. A Marinelli-shaped sample is placed around this copper wall. Sample is 3D-printed from dental LT clear resin in a cleanroom at ORNL. Marinelli shape parameters—chemical formula: $C_{23}H_{24}O_4$, molecular weight: 364.43 g/mol, density: 1.15 g/cm³ of solidified, outer radius = 74.93 mm, inner radius = 62.23 mm, outer height = 133.35 mm, inner height = 107.95 mm, base thickness = 25.4 mm. High-purity germanium (natural ^{72}Ge) detector dimension: diameter = 85 mm, height = 80 mm, mass = 1.170 kg, live time of measurement = 3.04×10^6 seconds. During assay measurements the copper chamber is filled with nitrogen gas to push out radon. Uranium and thorium present in the sample undergo radioactive decay and emit a gamma-line from each decay progeny. If broken equilibrium occurs, then the decay of the corresponding radionuclide also emits a gamma-line. Both of these gamma photons arrive at the high-purity germanium diode (central region) and are detected via Compton scattering, photoelectric absorption, or pair production depending on their incident energy. Credit: Thomas Ruland, Chang-Hong Yu, Grzegorz Zuzel, Yuri Efremenko, and Canfranc Underground Laboratory.

and the Marinelli-shaped 3D-printed sample is shown in light blue. The energy spectrum obtained from gamma-ray spectrometry of 1.170 kg of a dental LT clear sample with a measurement live time of 3.04×10^6 seconds (~ 45 days) is shown in Figure 8.5. The few gamma-lines originate from ^{232}Th and ^{238}U decay progeny are marked. Assuming secular equilibrium is not broken, the activity of ^{238}U and ^{232}Th is calculated from the daughter isotopes using the equation,

$$\mathcal{A} = \frac{S}{D \cdot t \cdot M \cdot (\text{B.R.})} \pm \frac{\sqrt{N}}{D \cdot t \cdot M \cdot (\text{B.R.})} \quad (8.1)$$

where $\mathcal{A}$ is activity in Bq/kg, S is signal counts at region of interest (ROI), N is total counts at ROI, D is detection efficiency of the gamma-line, t is measurement time in seconds, M is mass of the sample in kg, and B.R. is the branching ratio of the gamma-line. The gamma-ray spectrometry results for dental LT clear resin are listed in Table 8.3.

Comparison of radiopurity results of dental LT clear resin and surgical guide resin by two independent measurements, preliminary ICP-MS and gamma-ray spectrometry, are listed in Table 8.4. Radiopurity results of both resins are in agreement with each other and within the radiopurity of the best materials available for consideration for use in LEGEND-1000. A complete report of gamma-ray spectrometry is provided by the Canfranc Underground Laboratory and is attached in Appendix B.

GEANT4 simulation of this radioassay technique and results are discussed in the next section.

8.3 Gamma-ray Spectrometer Simulation

We performed Monte Carlo simulations of the full gamma-ray spectrometry instrument located at the Canfranc Underground Laboratory. The GEANT4 schematic of the complete original setup is shown in Figure 8.6, and the corresponding block diagram is shown in Figure 8.7.

This simulation was performed in two steps, one for each ^{238}U and ^{232}Th decay chain. As a result of nuclear decay in the 3D-printed sample (in the shape of Marinelli), gamma-lines are emitted from each decay daughter (or progeny). These gamma photons arrive at the

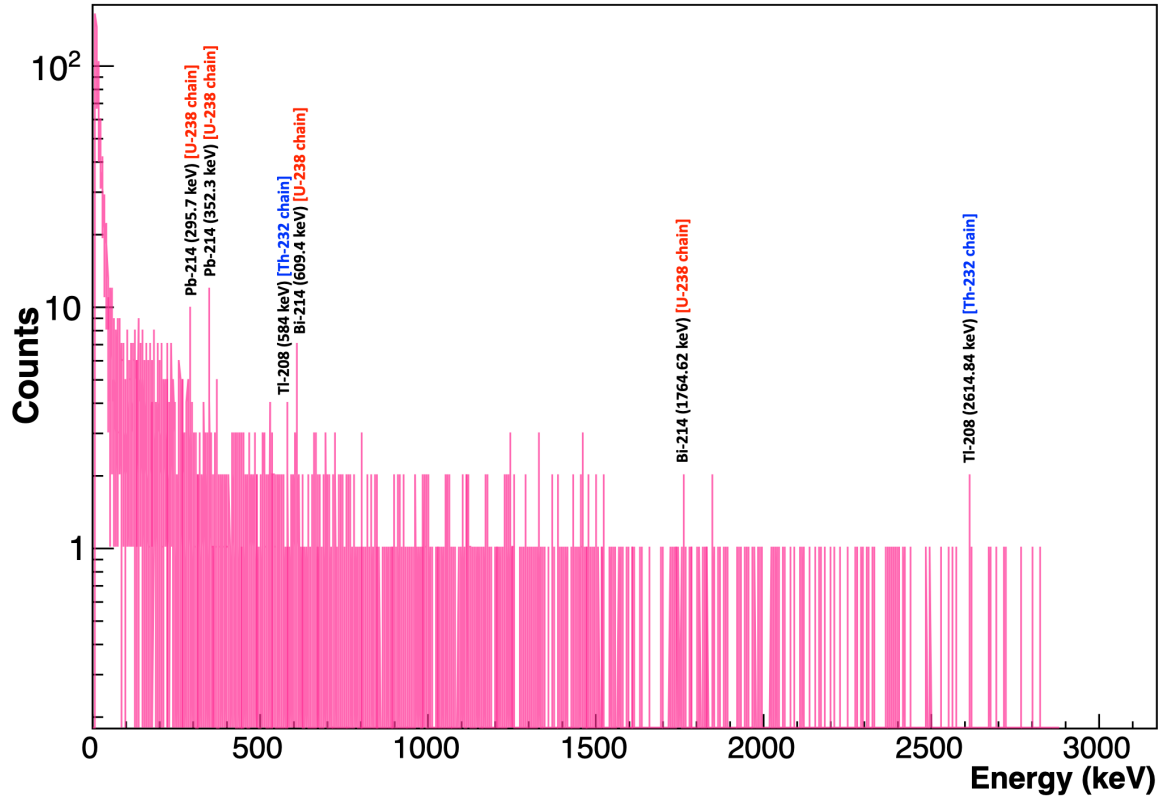


Figure 8.5: Energy spectrum of a dental LT clear resin sample from gamma-ray spectrometry. Sample mass 1.170 kg and live time of measurement 3.04×10^6 seconds. Few gamma-lines originate from ^{232}Th and ^{238}U decay progeny are marked. Credit: Grzegorz Zuzel, Canfranc Underground Laboratory.

Table 8.3: Gamma-ray spectrometry results for dental LT clear resin.

Chain	Isotope	Energy [keV]	Signal counts	Detection efficiency	Branching ratio	Activity [mBq/kg]	Activity [$10^{-5} \mu\text{g/g}$]
^{232}Th	^{208}Tl	584.1	5	0.025	0.31	0.18 ± 0.11	4.46 ± 2.67
^{232}Th	^{208}Tl	2614.8	2	0.007	0.36	0.22 ± 0.16	5.50 ± 3.89
^{238}U	^{214}Pb	352.3	27	0.034	0.37	0.60 ± 0.14	4.87 ± 1.13
^{238}U	^{214}Pb	295.7	26	0.038	0.19	1.00 ± 0.24	8.11 ± 1.97
^{238}U	^{214}Bi	609.4	19	0.026	0.46	0.45 ± 0.12	3.61 ± 0.99

Table 8.4: Comparison of radiopurity results of dental LT clear resin and surgical guide resin by two independent measurements: preliminary ICP-MS and gamma-ray spectrometry. The radiopurity results of both resins are within the order of magnitude with each other and within the radiopurity of the best materials available for consideration for use in LEGEND-1000.

	Dental LT clear (Gamma-counting) [mBq/kg]	Surgical guide (ICP-MS) [mBq/kg]
^{232}Th	0.18 ± 0.11	0.04 ± 0.01
^{238}U	0.60 ± 0.14 (< 7.4)	0.21 ± 0.04

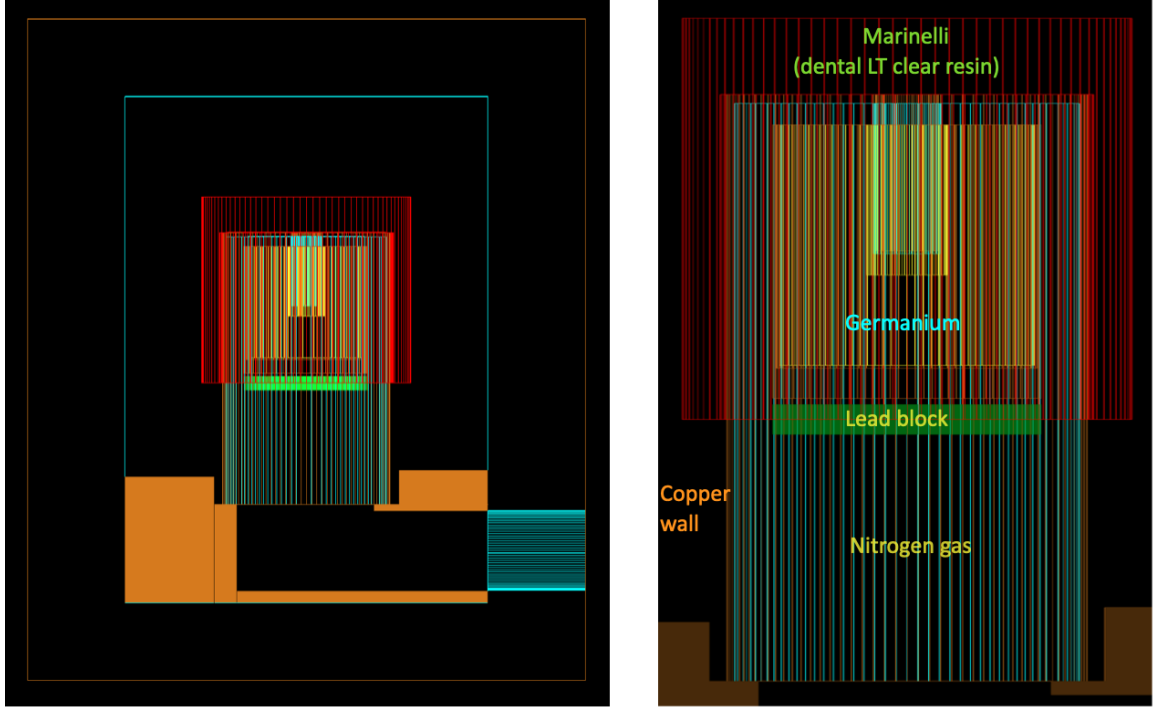


Figure 8.6: Left: GEANT4 simulation of the full radioassay instrument—gamma-ray spectrometer at the Canfranc Underground Laboratory. Right: zoom-in view of the central region of the instrument. 3D-printed Marinelli shaped sample is represented by the outer red structure, and the high-purity germanium detector is represented by the inner yellow structure. Radionuclides ^{238}U and ^{232}Th are uniformly distributed (separately) in the sample, and events deposited in the germanium detector are recorded for each decay.

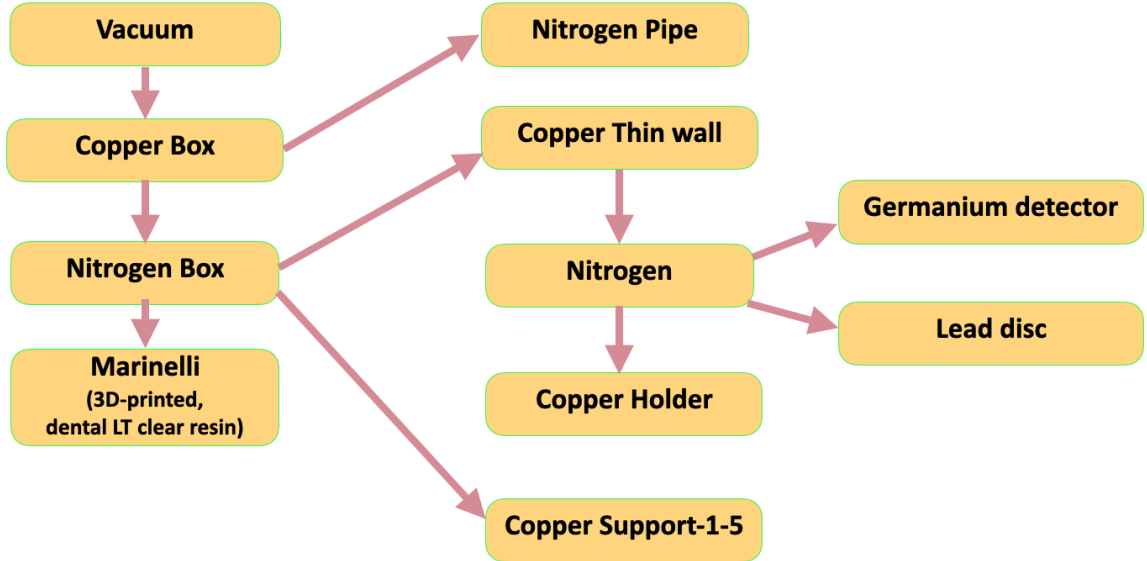


Figure 8.7: Block diagram of the complete radioassay instrument—gamma-ray spectrometer located at the Canfranc Underground Laboratory.

high-purity germanium diode (central region) and are detected via Compton scattering, photoelectric absorption, or pair production depending on their incident energy. Events are collected in the germanium detector for each decay chain separately.

Simulation of ^{238}U decay chain: For this part, 10^6 events of ^{238}U radionuclide were uniformly distributed in the sample. The complete decay chain of ^{238}U occurs, and generated events are deposited in the germanium detector. The simulated energy spectrum of the ^{238}U decay chain is shown in Figure 8.8. Peaks above the continuum are due to the gamma-lines emitted from all the decay progeny of ^{238}U decays in the dental sample. Only visible (above the background) gamma-lines of radionuclides having a branching ratio $> 10\%$ are marked in the spectrum at their respective gamma energies with their corresponding nuclide. Seven nuclides from decay progeny are clearly identified: ^{214}Pb (77.1 keV), ^{234}Th (92.5 keV), ^{214}Pb (295.1 keV), ^{214}Pb (351.9 keV), ^{214}Bi (609.3 keV), ^{214}Bi (1120.3 keV), and ^{214}Bi (1764.5 keV). Efficiency is calculated for these isotopes using the equation below.

$$\text{Efficiency} = \frac{S-B}{10^6 \times (\text{B.R.})} \pm \frac{S-B}{10^6 \times (\text{B.R.})} \times \frac{\sqrt{S+B}}{S-B} \quad (8.2)$$

where S is maximum events at given gamma energy, B is background events (average event adjacent to corresponding maximum event), and B.R. is the branching ratio of the given gamma-line. The characteristic nuclide and the branching ratio are identified from the nuclear database at their respective γ -energy [65]. Radionuclides are selected with a branching ratio $> 10\%$. Efficiency values of these nuclides are listed in Table 8.5.

Simulation of ^{232}Th decay chain: For this part, 10^6 events of ^{232}Th radionuclide were uniformly distributed in the Marinelli-shaped sample. The complete decay chain of ^{232}Th occurs, and generated events are deposited in the germanium detector. The simulated energy spectrum of the ^{232}Th decay chain is shown in Figure 8.9. Peaks above the continuum are due to the gamma-lines emitted from all the decay daughter of ^{232}Th decays in the sample. Only visible (above the background) are γ -lines of radionuclides having a branching ratio $> 10\%$, which are marked in the spectrum at their respective gamma-energy with their corresponding nuclide. The characteristic nuclide and the branching ratio are identified from the nuclear database at their respective γ -energy. Radionuclides are selected with a branching ratio $> 10\%$. Efficiency values of these nuclides are listed in Table 8.6.

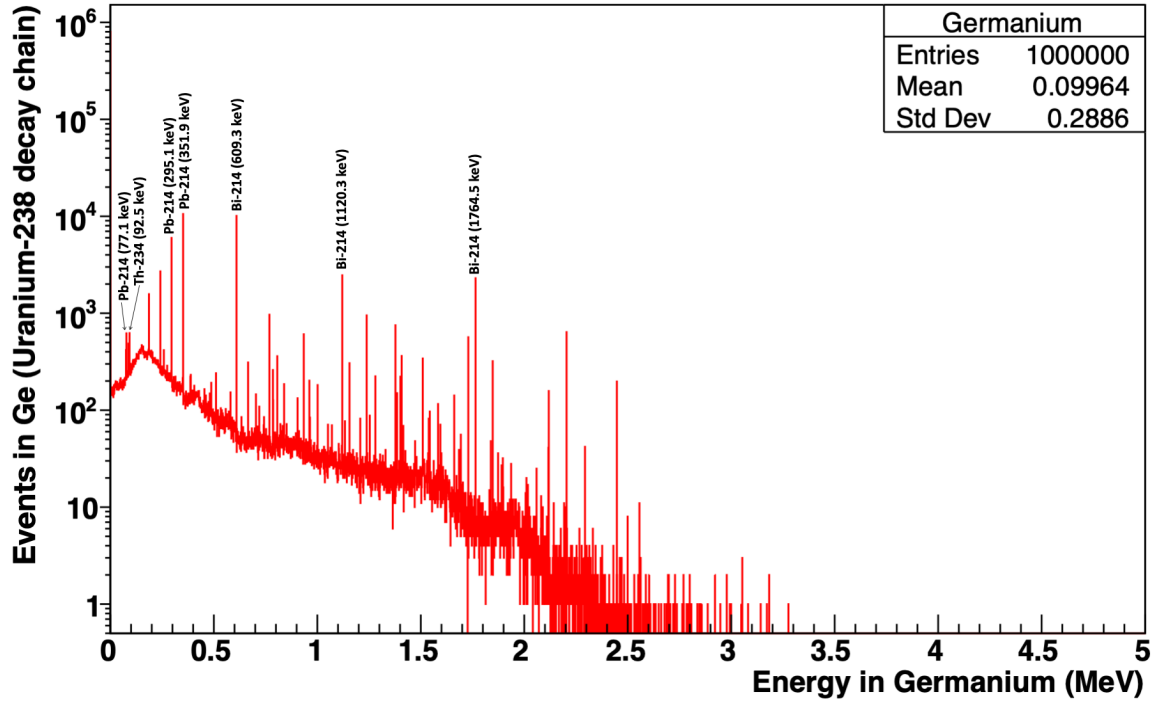


Figure 8.8: Simulated energy spectrum of the ^{238}U decay chain. ^{238}U radionuclides are uniformly distributed in the entire Marinelli-shaped sample. 10^6 nuclear decay events of ^{238}U are simulated in GEANT4. Resulting gamma-lines from the entire decay progeny are collected in the high-purity germanium detector. Peaks above the continuum appear due to the gamma-emission from all decaying radionuclides. Only visible (above the background) gamma-lines having a branching ratio $> 10\%$ are marked at their respective gamma-energy with their corresponding nuclide. Fine binning is selected with a bin size (energy/bin) of 1 keV/bin. Simulation is performed with a complete gamma-ray spectrometer located at the Canfranc Underground Laboratory.

Table 8.5: Simulated data of the efficiency of gamma for ^{238}U decay chain. Efficiency is calculated using the equation 8.2 and the data of gamma-lines obtained from GEANT4 simulation. Only nuclides with a branching ratio $> 10\%$ are selected for efficiency calculation. Statistics of 10^6 events are generated. ^{238}U radionuclides are uniformly distributed in the entire Marinelli-shaped sample. Resulting nuclear decay emits gamma-lines from the entire decay progeny. Energy in the germanium detector is deposited. Each nuclide has a characteristic branching ratio corresponding to its γ -energy, which is obtained from the nuclear database. Peak visibility in the simulated energy spectrum is marked. Highest efficiency is predicted for two isotopes: 3% for ^{214}Pb (295.1 keV), and 2.8% for ^{214}Pb (351.9 keV). Data is obtained using the GEANT4 simulation of a complete geometry of gamma-ray spectrometer located at the Canfranc Underground Laboratory.

Isotope	Max. events	Bkg. events	γ -energy (keV)	Branching ratio	Efficiency %	Peak visibility
^{214}Pb	622	208.5	77.107	0.11	0.376 ± 0.026	visible
^{234}Th	625	235.5	92.5	0.0213	1.829 ± 0.138	visible
^{214}Pb	5993	227	295.091	0.192	3.003 ± 0.041	visible
^{214}Pb	10571	142	351.87	0.371	2.811 ± 0.028	visible
^{214}Bi	10106	53	609.311	0.461	2.181 ± 0.022	visible
^{214}Bi	2468	32	1120.273	0.15	1.624 ± 0.033	visible
^{214}Bi	2302	7.5	1764.490	0.159	1.443 ± 0.030	visible

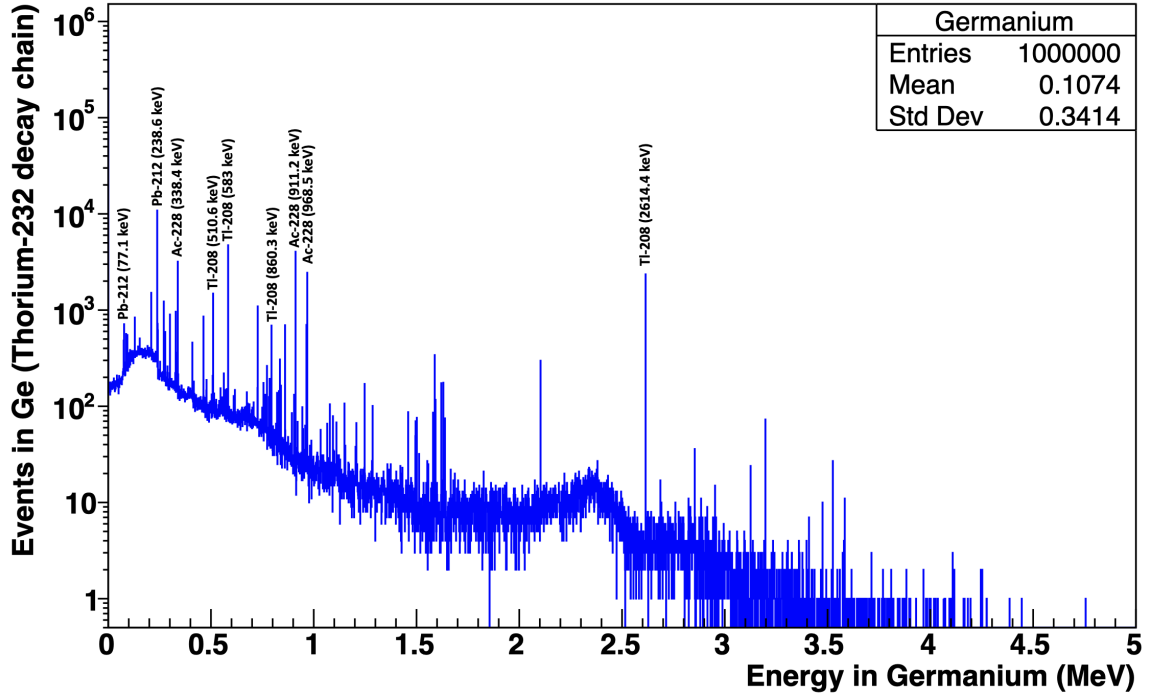


Figure 8.9: Simulated energy spectrum of the ^{232}Th decay chain. ^{232}Th radionuclides are uniformly distributed in the entire sample. 10^6 nuclear decay events of ^{232}Th are simulated in GEANT4. Resulting gamma-lines from the entire decay progeny are collected in the high-purity germanium detector. Peaks above the continuum appear due to the gamma-emission from all decaying radionuclides. Only visible (above the background) gamma-lines having a branching ratio $> 10\%$ are marked at their respective gamma-energy with their corresponding nuclide. Fine binning is selected with a bin size (energy/bin) of 1 keV/bin. Simulation is performed with a complete gamma-ray spectrometer located at the Canfranc Underground Laboratory.

Table 8.6: Simulated data of the efficiency of gamma for ^{232}Th decay chain. Efficiency is calculated using the equation 8.2 and the data of gamma-lines obtained from GEANT4 simulation. Only nuclides with a branching ratio $> 10\%$ are selected for efficiency calculation. Statistics of 10^6 events are generated. ^{232}Th radionuclides are uniformly distributed in the entire sample. Resulting nuclear decay emits gamma-lines from the entire decay progeny. Energy in the germanium detector is deposited. Each nuclide has a characteristic branching ratio corresponding to its γ -energy, which is obtained from the nuclear database. Peak visibility in the simulated energy spectrum is marked. The highest efficiency predicted for two isotopes: 2.46 % for ^{228}Ac (338.4 keV) and 2.42% for ^{212}Pb (238.6 keV). Data is obtained using the GEANT4 simulation of a complete geometry of gamma-ray spectrometer located at the Canfranc Underground Laboratory.

Isotope	Max. events	Bkg. events	γ -energy (keV)	Branching ratio	Efficiency %	Peak visibility
^{212}Pb	714	221	77.107	0.177	0.279 ± 0.017	visible
^{212}Pb	10847	276.5	238.578	0.436	2.424 ± 0.024	visible
^{228}Ac	3191	136.5	338.42	0.124	2.463 ± 0.047	visible
^{208}Tl	1486	98.5	510.066	0.078	1.779 ± 0.051	visible
^{208}Tl	4727	77	583.022	0.31	1.5 ± 0.022	visible
^{208}Tl	697	39.5	860.30	0.043	1.529 ± 0.063	visible
^{228}Ac	4048	39	911.16	0.29	1.382 ± 0.022	visible
^{228}Ac	2447	32.5	968.97	0.174	1.388 ± 0.029	visible
^{208}Tl	2356	3	2614.35	0.359	0.655 ± 0.014	visible

Nine nuclides from decay progeny are clearly identified: ^{212}Pb (77.1 keV), ^{212}Pb (238.6 keV), ^{228}Ac (338.4 keV), ^{208}Tl (510.6 keV), ^{208}Tl (583 keV), ^{208}Tl (860.3 keV), ^{228}Ac (911.2 keV), ^{228}Ac (968.97 keV), and ^{208}Tl (2614.4 keV).

Dependency of detection efficiency on gamma energy: To see how the apparatus is sensitive to the various gamma energies, the detection efficiency of single gamma-rays was calculated for the spectrometer. To simulate, 10^6 events of gamma photons of various energies are uniformly distributed in the Marinelli-shaped sample. Events produced after the interaction of gamma photons are recorded in the germanium detector. Detection efficiency of the spectrometer as a function of gamma energy is shown in Figure 8.10. Detection efficiency is at maximum around 200 keV. The simulation shows the detection efficiency decreases with increasing gamma energy, due to the lesser interaction of gamma photons with the germanium detector. As an example of energy deposition in a germanium detector for a 1 MeV gamma photon with origin uniformly in the sample is shown in Figure 8.11.

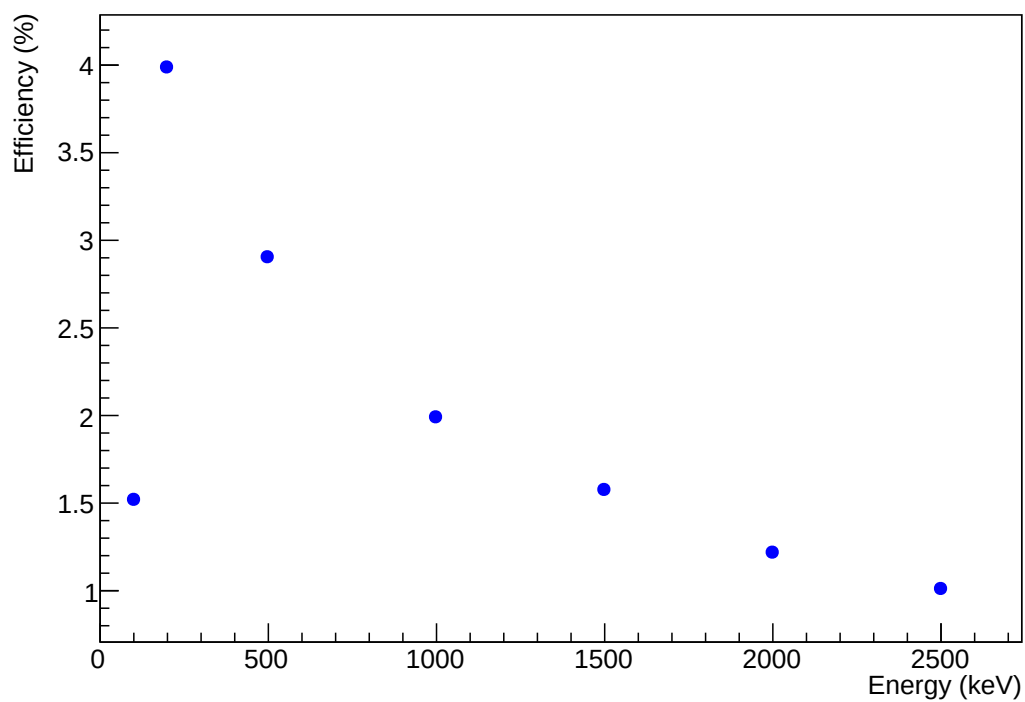


Figure 8.10: Detection efficiency of the gamma-ray spectrometer as a function of gamma energy. To simulate, 10^6 events of gamma photons of various energies are uniformly distributed in the Marinelli-shaped sample. Events produced after the interaction of gamma photons are recorded in the germanium detector. Detection efficiency is at maximum around 200 keV. Simulation shows detection efficiency decreases with increasing gamma energy, due to the lesser interaction of gamma photons with the germanium detector.

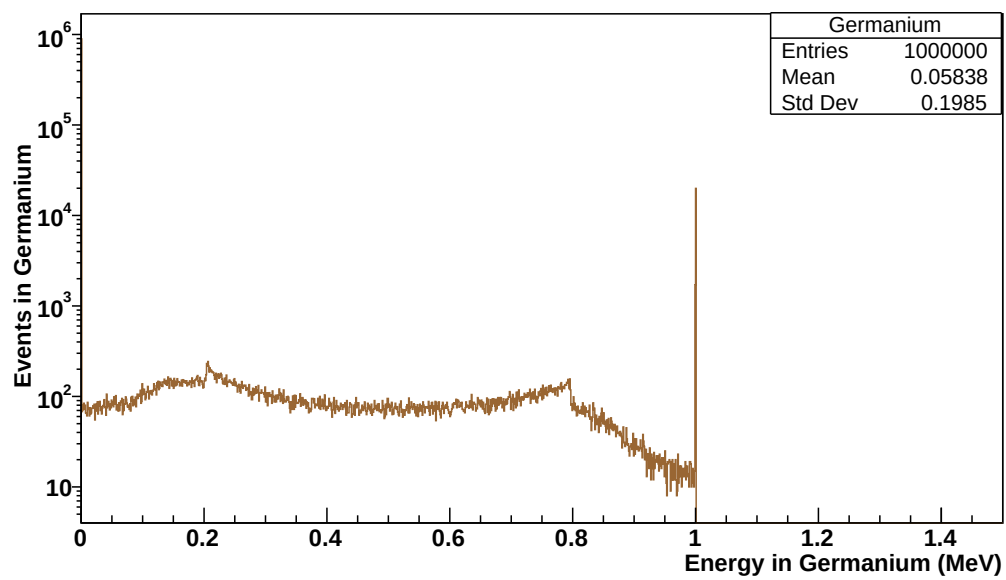


Figure 8.11: Simulated energy spectrum in a germanium detector for a 1 MeV gamma photon with origin uniformly in the sample.

Chapter 9

Conclusion

The search for an extremely rare nuclear transition, neutrinoless double-beta ($0\nu\beta\beta$) decay will confirm if neutrinos are Majorana fermions and provide constraints or measure effective Majorana neutrino mass ($m_{\beta\beta}$). As the decay violates the total lepton number conservation, it provides an avenue to interpret the matter-antimatter asymmetry in the Universe. In addition, $0\nu\beta\beta$ decay also benefits nuclear physics by providing the experimental value of the nuclear matrix element and paves the way for physics beyond the Standard Model. Current experimental limits on $T_{1/2}^{0\nu}$ of $0\nu\beta\beta$ decay and corresponding limits on $m_{\beta\beta}$ are listed in Table 2.2. The sensitivity of current and next-generation experiments are shown in Figure 2.3.

The LEGEND (Large Enriched Germanium Experiment for Neutrinoless $\beta\beta$ Decay) experiment started collecting physics data for its first phase with 200 kg ^{76}Ge (LEGEND-200). Recently released first results of LEGEND-200 are shown in Figure 3.5. LEGEND-200 set the lower limit on half-life $T_{1/2}^{0\nu} > 1.9 \times 10^{26}$ yr for ^{76}Ge , and that corresponds to the upper limits on $m_{\beta\beta} < 75\text{--}200$ meV [3].

LEGEND-1000 is a next-generation $0\nu\beta\beta$ decay experiment with 1000 kg of ^{76}Ge and is currently in the R&D phase. LEGEND-1000 aims for a “quasi-background-free” experiment with a background goal $< 1 \times 10^{-5}$ cts/(keV·kg·yr) around 2.039 MeV [44].

Design and review of LEGEND-1000 is currently ongoing. Critical Decision 1 is scheduled in 2025, and after the approval from the Department of Energy, LEGEND-1000 construction will begin at LNGS. The first physics data of LEGEND-1000 is expected to start in 2030.

The expected total energy spectrum, including all backgrounds, is shown in Figure 3.7, and on the same plot, the expected signal of $0\nu\beta\beta$ decay with $T_{1/2}^{0\nu}$ of 10^{28} years is shown. The detector parameters of LEGEND-1000 are listed in Table 3.1. Ultra-low background is extremely important for the observation of $0\nu\beta\beta$ decay. ^{42}K (decay daughter of ^{42}Ar) is one of the main backgrounds in LEGEND-1000. My work is focused on the mitigation of ^{42}K background. We performed extensive studies on how to mitigate the ^{42}Ar background. Since underground-sourced liquid argon will not be available, the R&D discussed in this dissertation was performed.

1. We performed Monte Carlo simulations of ^{42}K background in a germanium detector. The effect of germanium detector dead layer thickness on ^{42}K background events as a function of argon volume are summarized in Table 4.2. In conclusion, there will be a large mass loss, and this approach is very expensive, since the germanium detector is very costly and the production of enriched germanium ^{76}Ge detector is a long process, so we do not intend to reduce the sensitive volume of the germanium. Due to the germanium mass loss, this method reduces the exposure of the $0\nu\beta\beta$ decay half-life sensitivity measurements since $\text{half-life} \propto \text{exposure}^{1/2}$, in the presence of background.
2. We performed Monte Carlo simulations to study the impact of the ^{42}K background on a germanium detector with a dead layer surrounded by liquid argon. We simulated three scenarios: (a) The detector with 0.5 mm dead layer and 1 mm layer of liquid argon produced a larger ^{42}K background than the LEGEND goal. (b) The detector with 1 mm dead layer and 1 mm layer of liquid argon results are within the background goal but with a loss in sensitive germanium mass. (c) The detector with 0.5 mm dead layer and 0.2 mm layer of liquid argon achieved background lower than the goal. In conclusion, thicker dead layer detectors meet the LEGEND background goal but have a loss in sensitive germanium mass. Reduced liquid argon volume surrounding the detector also meets the background goal but requires tight encapsulation. Simulation results of the above three cases are summarized in Table 5.1. The simulation of PEN encapsulation of a germanium detector with energy deposition in germanium at various PEN thicknesses for the incident electrons of 3.525 MeV (Q -value of ^{42}K) is shown in Figure 4.10. The simulation shows 6 to 8 mm PEN encapsulation for the

germanium would be sufficient to lower the energy of electrons below the 2039 keV ($Q_{\beta\beta}$ of ^{76}Ge). However, encapsulation of detectors with 6 to 8 mm PEN would not be viable because of radio-purity considerations. Encapsulation with 2 mm PEN along with a liquid argon veto would be a viable option for the reduction of the background caused by ^{42}K at the region of interest. Encapsulation of detectors by PEN is a promising approach. However, ultra-clean PEN is difficult to build, and this is not 3D-printable material. Because all detectors have different sizes, customization of PEN encapsulation could be a long and expensive approach. As a result, we focused on the materials that are 3D printable and can encapsulate in a clean, hands-free environment.

3. In collaboration with the LEGEND group of ORNL, my work focuses on the research and development of the additive manufacturing (3D-printing) of low-background materials. We developed procedures and equipment to build ultra-clean 3D-printable materials. Laser-based stereolithography technique is used to build materials in a cleanroom. We observe that a sample from dental LT clear resin is relatively more optically transparent than the sample from surgical guide resin. 3D-printed samples with surgical guide resin and dental LT clear resin are shown in Figure 6.6, and LEGEND-1000 parts built with surgical guide resin are shown in Figure 6.7.
4. To measure non-radioactive impurities, we developed a vacuum system to measure outgassing rates of the 3D-printed samples. Both 3D-printed samples show lowering outgassing rates. The outgassing rate of dental LT clear resin and surgical guide resin is small and within measurement uncertainties. Outgassing rates are listed in Table 7.2.
5. To measure radioactive impurities, we sent 3D-printed samples to two different radioassays. A surgical guide resin sample was sent to the Radioactive Materials Analytical Lab (RMAL) at ORNL for Inductively Coupled Plasma Mass Spectrometry (ICP-MS); preliminary results are listed in Table 8.1. Data shows ^{40}K presence is higher than both ^{238}U and ^{232}Th . A dental LT clear resin sample sent to Canfranc Underground Laboratory, Spain, for gamma-ray spectrometry, obtained

energy spectrum is shown in Figure 8.5 and results are listed in Table 8.3. The radiopurity results of both resins are within the order of magnitude with each other and within the radiopurity of the best materials available for consideration for use in LEGEND-1000. In addition, we performed Monte Carlo simulations of the full gamma-ray spectrometry instrument located at the Canfranc Underground Laboratory. The simulated energy spectrum of the ^{238}U decay chain is shown in Figure 8.8, and for the ^{232}Th decay chain is shown in Figure 8.9. Detection efficiency is calculated from the simulated data. Efficiency of the ^{238}U decay chain is listed in Table 8.5, and for the ^{232}Th decay chain is listed in Table 8.6.

We conclude that ^{42}K background mitigation is possible for the LEGEND-1000 experiment with the atmospheric liquid argon and encapsulation for germanium detectors. It requires very tight custom-built encapsulation of every individual detector. Monte Carlo simulations show for passive encapsulation even a 1 mm gap between passive encapsulation and the detector is too much. Using scintillation material for encapsulation can mitigate such a requirement. We prove that 3D-printing can produce ultra-radiopure components. During reported work, a lineup of equipment and processes able to produce and test ultra-pure materials has been established.

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Appendix

A ICP-MS Data

To measure the radioactivity level mainly due to the presence of parent nuclides, ^{238}U and ^{232}Th and ^{40}K in our 3D-printed sample (surgical guide resin), we sent it to Radioactive Materials Analytical Lab (RMAL) at ORNL for mass spectrometry. They perform Inductively Coupled Plasma Mass Spectrometry (ICP-MS) with our sample. The original report of the ICP-MS analysis provided by RMAL, ORNL, is attached on the next pages.

RMAL250237 Surgical Guide #1 sample results at ORNL

Tamara Keever

5/5/25

3D printed small pieces of Surgical Guide resin, printed by Chang-Hong in C110 cleanroom in February 2025.



These samples were printed on Feb.25, 2025. They are support for possible fiber holders for LEGEND-1000 proposed and designed by Matthew Busch. The printed fiber holders were given to Busch at the March Collaboration meeting by Brennan.

The surgical guide resin was printed in the Physics Division Cleanroom, no de-gassing was performed before printing. After printing, the products were taken outside of the cleanroom, washed in isopropanol for 25 min. within the ultrasound cleaner, air dried, then placed in the curing box and cured according to specifications: 30 minutes at 60 degree C.

On Feb. 28, 2025, these samples were then given to Tammy Keever of the Radioactive Materials Analytical Lab in CSD at ORNL. These are what Tammy referred to as the “second sample”, and are the samples used for analysis.

Surgical Guide # 1 sample (our standard 1” tile) was received at Radioactive Materials Analytical Lab (RMAL) on 2/21/25 for method development of the assay of trace U238, Th232 and K. A second sample was received 2/28/25. The second sample was submitted as a “hardened” sample that would perhaps be easier to work with than the larger tile for this method development. The sample is associated with the LabWare RMAL ID 250237. The initial sample has not been opened and is in custody at RMAL.

The hardened sample material was observed to have a printed lattice orientation and was much more attainable to sizing pieces for sample preparation. This sample was sent to the Chemical and Isotopic Mass Spectrometry (CIMS) group where it was received at the ISO 7 cleanroom for sample preparation. A mass of 4.1g hardened material was weighed and ashed in a tube furnace at 600C for 12 hours using a pre-leached quartz crucible. A blank quartz crucible was included in the tube furnace as a blank control. The residual sample was quantitatively transferred to a clean Savillex bottle, with the blank crucible rinsed into a second Savillex bottle as a control. Four rinses using 4N nitric acid (Optima grade, Fisher Scientific) was used to move the sample residue from the crucible to the Savillex bottle.

The ash was leached in the Savillex bottle using 10ml of 4N nitric, with addition of 1ml concentrated hydrofluoric acid (Optima grade, Fisher Scientific). The ash residue was heated with acid leaching using a hotplate at 110C for ~18 hours. The residual leach and method blank acids were reduced in volume and returned to the RMAL for analysis. The RMAL received the leach solution and method blank on 3/19/25. At the RMAL, the solutions were further reduced in volume to near dryness on a hotplate. A matrix of 2% HNO₃ was added to the Savillex by weight, capped and allowed to warm at 70C on a hotplate for 2 hours. The ash did not go fully into solution, figure 1.



Figure 1. Surgical Guide Resin #1, hardened final acid leach of residual ash from muffle furnace preparation.

Mass spectrometry analysis

Both the sample preparation and method blank were diluted in 2% nitric acid matrix and analyzed using inductively coupled plasma mass spectrometry (ICP-MS), iCAP-RQ (Thermo Scientific) and ISDS Qtegra software. Elemental responses were established using NIST-traceable standards ²³⁸U and ²³²Th obtained from High Purity Standards (Charlston, SC) and Inorganic Ventures (Christiansburg, VA) and processed with least squares linear regression calibration fits. The total Th, U, and K data was reported as ug/g solid and ug/ml method blank. The data for Surgical Guide #1 and method blank are listed in Table 1 and Table 2.

Table 1. ICP-MS results for Surgical Guide #1.

Element/isotope	250237-001 Surgical guide #1		RL, ug/g
	ug/g	uncert	
K	3.88E+00	3.88E-01	3.92E-01
Th	1.07E-05	2.15E-06	2.26E-06
U	1.70E-05	3.41E-06	4.27E-06

Table 2. ICP-MS results for method blank, trace metals ashing.

Element/isotope	250237-BLK method blank		RL, ug/ml
	ug/ml	uncert	
K	<	2.91E-02	2.91E-02
Th	<	8.37E-06	8.37E-06
U	<	1.58E-05	1.58E-05

A second portion of the hardened material was hot acid leached using aqua regia and HF at 200C using hotplate over 4 hours. This portion of the material was also analyzed by ICP-MS, but detection limits could not be achieved for U and Th because no signal can be seen. The screen data results of this leach sample indicated elevated potassium levels. The hardened material was consumed in these method development tests.

Mechanical properties of surgical guide resin. Credit: FoamLab.

Material Properties	Post-Cured ^{1, 2}	Method
Elongation at Break	12%	ASTM D638
Flexural Strength	> 102 MPa	ASTM D790
Flexural Modulus	> 2400 MPa	ASTM D790

Sterilization Compatibility

E-beam	35 kGy E-beam radiation
Ethylene Oxide	100% Ethylene oxide at 55 °C for 180 minutes
Gamma	29.4 - 31.2 kGy gamma radiation
Steam Sterilization	Autoclave at 134 °C for 20 minutes Autoclave at 121 °C for 30 minutes

For more details on sterilization compatibilities, visit formlabs.com

Disinfection Compatibility

Chemical Disinfection	70% Isopropyl Alcohol for 5 minutes
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Surgical Guide Resin is a Class I Medical Device as defined in Article 2 of the Medical Device Regulation 2017/74 (MDR) in the EU and in Section 201(h) of the Federal Food Drug & Cosmetic (FD&C) Act.

Surgical Guide Resin has been evaluated in accordance with ISO 10993-1, Biological evaluation of medical devices - Part 1: Evaluation and testing within a risk management process, and ISO 7405, Dentistry - Evaluation of biocompatibility of medical devices used in dentistry, and passed the requirements for the following biocompatibility risks:

ISO Standard	Description ³
EN ISO 10993-5	Not cytotoxic
EN ISO 10993-10	Not an irritant
EN ISO 10993-10	Not a sensitizer

The product was developed and is in compliance with the following ISO Standards:

ISO Standard	Description
EN ISO 13485	Medical Devices – Quality Management Systems – Requirements for Regulatory Purposes
EN ISO 14971	Medical Devices – Application of Risk Management to Medical Devices

¹ Material properties may vary based on part geometry, print orientation, print settings, temperature, and disinfection or sterilization methods used.

² Data for post-cured samples were measured on Type IV tensile bars printed on a Form 2 printer with 100 µm Surgical Guide Resin settings, washed in a Form Wash for 20 minutes in ≥99% Isopropyl Alcohol, and post-cured at 60°C for 30 minutes in a Form Cure.

³ Surgical Guide Resin was tested at NAMSA World Headquarters, OH, USA.

B Gamma-Ray Spectrometry Data

To measure the radioactivity level mainly due to the presence of decay progenies of ^{238}U and ^{232}Th decay chains in our 3D-printed sample (dental LT clear resin), we sent it to Canfranc Underground Laboratory in Spain for gamma-ray spectrometry. They perform gamma-counting with our sample. The original report of the gamma-ray screening provided by the Canfranc Underground Laboratory is attached on the next pages.

Dental Clear material

Sample Preparation

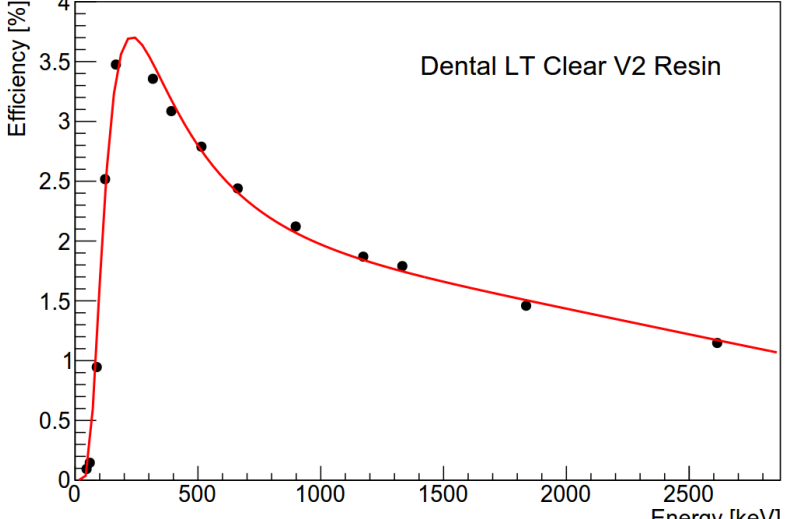
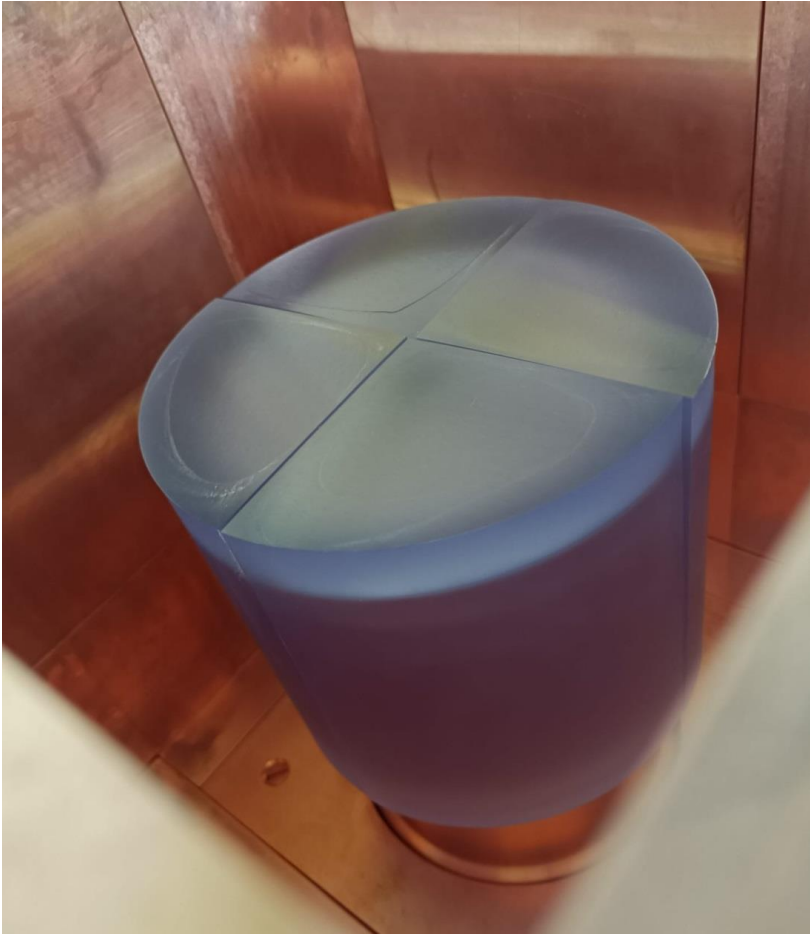
The Dental Clear Resin was degassed in Lab 107 Building 6000 ORNL, first (pumped by a cold trap to extract moisture and other unwanted elements), then immediately moved into cleanroom for printing. Each 1/4 of the Marinelli was printed using the Form 2 printer, then washed in isopropanol solution within an ultrasonic cleaner for 30 minutes. After air dry, the samples were cured in the curing box according to specifications (60 minutes at 60 degrees C). The cured samples were then double bagged using cleanroom bags, taken out of the cleanroom, packaged and shipped to Spain.

Sample has Marinelli shape with following parameters: outer radius = 74.93 mm, inner radius = 62.23 mm, outer height = 133.35 mm, inner height = 107.95 mm, base thickness = 25.4 mm.

Chemical formula: $C_{23}H_{24}O_4$, molecular weight: 364.43 g/mol, density: 1.15 g/cm³

At Canfranc underground lab, the pieces have been wiped with ethanol (using clean room tissues and HP ethanol) just before placing in the detector.

Report γ-ray screening				
Detector:	GeRysy / LSC			
Report No.:	04/2025			
Operator:	E. Ruiz, G. Zuzel			
Analysis:	G. Zuzel			
Sample:	3D Printed Resin ORNL (LEGEND-1000 Experiment)			
	Mass:	1170	g	
	Amount:	4	pieces	
Measurement:	From:	29.05.2025 10:10		
	To:	15.07.2025 15:28		
	Live Time:	3 043 008	sec	
Results:				
	Activity [mBq]	Activity concentration [mBq/kg] [mBq/piece]		Remarks
²³⁸ U:				
²¹⁰ Pb				
²²⁶ Ra	0.64 ± 0.17	0.54 ± 0.14		
²³⁸ U	< 8.8	< 7.4		90% C.L.
²³² Th:				
²²⁸ Ra				
²²⁸ Th	< 0.4	< 0.35		90% C.L.
²³⁵ U:				
⁴⁰ K:	< 2.3	< 1.9		90% C.L.
¹³⁷ Cs:				
⁶⁰ Co:	< 16	< 14		90% C.L.
Comments:	Sample in four pieces.			

Applied Efficiency Curve:	Dental LT Clear V2 Resin  <table border="1"><caption>Approximate data points from the Applied Efficiency Curve</caption><thead><tr><th>Energy [keV]</th><th>Efficiency [%]</th></tr></thead><tbody><tr><td>0</td><td>0.0</td></tr><tr><td>50</td><td>0.2</td></tr><tr><td>100</td><td>0.9</td></tr><tr><td>150</td><td>2.5</td></tr><tr><td>200</td><td>3.5</td></tr><tr><td>250</td><td>3.7</td></tr><tr><td>300</td><td>3.4</td></tr><tr><td>400</td><td>3.1</td></tr><tr><td>500</td><td>2.8</td></tr><tr><td>600</td><td>2.5</td></tr><tr><td>700</td><td>2.2</td></tr><tr><td>800</td><td>2.0</td></tr><tr><td>900</td><td>1.8</td></tr><tr><td>1000</td><td>1.7</td></tr><tr><td>1200</td><td>1.6</td></tr><tr><td>1400</td><td>1.5</td></tr><tr><td>1600</td><td>1.4</td></tr><tr><td>1800</td><td>1.3</td></tr><tr><td>2000</td><td>1.2</td></tr><tr><td>2200</td><td>1.1</td></tr><tr><td>2400</td><td>1.0</td></tr><tr><td>2600</td><td>0.9</td></tr></tbody></table>	Energy [keV]	Efficiency [%]	0	0.0	50	0.2	100	0.9	150	2.5	200	3.5	250	3.7	300	3.4	400	3.1	500	2.8	600	2.5	700	2.2	800	2.0	900	1.8	1000	1.7	1200	1.6	1400	1.5	1600	1.4	1800	1.3	2000	1.2	2200	1.1	2400	1.0	2600	0.9
Energy [keV]	Efficiency [%]																																														
0	0.0																																														
50	0.2																																														
100	0.9																																														
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200	3.5																																														
250	3.7																																														
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Sample Photo:																																															

Mechanical properties of dental LT clear resin V2. Credit: FoamLab.

Material Properties	METRIC ¹	METHOD
	Post-Cured ²	
Tensile Properties	METRIC ¹	METHOD
Ultimate Tensile Strength	52 MPa	ASTM D638-10 (Type IV)
Young's Modulus	2080 MPa	ASTM D638-10 (Type IV)
Elongation at Break	12%	ASTM D638-10 (Type IV)
Flexural Properties	METRIC ¹	METHOD
Flexural Strength	84 MPa	ASTM D790-15 (Method B)
Flexural Modulus	2300 MPa	ASTM D790-15 (Method B)
Hardness Properties	METRIC ¹	METHOD
Hardness Shore D	78D	ASTM D2240-15 (Type D)
Impact Properties	METRIC ¹	METHOD
Unnotched Izod	449 J/m	ASTM D4812-11 (Unnotched)
Other Properties	METRIC ¹	METHOD
Water Absorption	0.54%	ASTM D570-98 (2018)

Dental LT Clear Resin (V2) has been evaluated in accordance with ISO 10993-1:2018, Biological evaluation of medical devices - Part 1: Evaluation and testing within a risk management process, and ISO 7405:2018, Dentistry - Evaluation of biocompatibility of medical devices used in dentistry, and passed the requirements for the following biocompatibility risks:

ISO Standard	Description ³
ISO 10993-5:2009	Not cytotoxic
ISO 10993-10:2010/(R)2014	Not an irritant
ISO 10993-10:2010/(R)2014	Not a sensitizer
ISO 10993-3:2014	Not mutagenic
ISO 10993-17:2002, ISO 10993-18:2005	Not toxic (subacute / subchronic)

The product was developed and is in compliance with the following ISO Standards:

ISO Standard	Description
EN ISO 13485:2016	Medical Devices – Quality Management Systems – Requirements for Regulatory Purposes
EN ISO 14971:2012	Medical Devices – Application of Risk Management to Medical Devices

¹ Material properties may vary based on part geometry, print orientation, print settings, temperature, and disinfection or sterilization methods used.

² Data were measured on post-cured samples printed on a Form 3B printer with 100 µm Dental LT Clear Resin (V2) settings, washed in a Form Wash for 20 minutes in 99% isopropyl alcohol, and post-cured at 60 °C for 60 minutes in a Form Cure.

³ Dental LT Clear Resin (V2) was tested at NAMSA World Headquarters, OH, USA.

Vita

Ibrahim Mirza earned an undergraduate degree in physics and mathematics from Shia P.G. College, University of Lucknow, in 2011 and a master's degree in physics with a specialization in condensed matter physics from Christ Church P.G. College, Chhatrapati Shahu Ji Maharaj University, Kanpur, in 2015. During 2016–2018, he worked on neutrino oscillations and light dark matter at the University of Lucknow. In 2019, Ibrahim started his graduate studies in physics at The University of Tennessee Knoxville. In 2021, he earned a concurrent master's degree in physics with a concentration on elementary particle physics from The University of Tennessee Knoxville. He worked on the search for neutrinoless double-beta decay at LEGEND-1000 experiment and conducted his research at Oak Ridge National Laboratory. Ibrahim defended his PhD in physics on August 25, 2025.