Investigating the Radiopurity of Lithium Molybdate for Neutrinoless Double Beta Decay Searches

Jay-U Chung[1]

[1] *Middlebury College*

July 28, 2021

Abstract

Detecting neutrinoless double beta decay (DBD), a hypothesized reaction with lifetime above $10^{25} - 10^{26}$ years, remains an important problem for its implications for the Standard Model and the matter-antimatter asymmetry problem. The Cryogenic Underground Observatory for Rare Events (CUORE) is the largest experiment using bolometers to search for neutrinoless DBD. Critical to any search for these rare decays is the use of radiopure materials that will minimize background events. The upcoming CUORE Upgrade with Particle Identification (CUPID) project is therefore investigating Lithium Molybdate (LMO) crystals enriched with molybdenum-100, a new DBD source that can achieve lower backgrounds. We analyze the radioactive spectrum of off-the-shelf LMO powder. Off-the shelf materials, if sufficiently radiopure, have many advantages in terms of cost and convenience. In particular, we aim to characterize high energy events in LMO from cosmogenic activation; such events could be problematic for low background experiments as CUPID. To capture the radioactive spectrum of a 1 kg sample of LMO, we used a high purity germanium detector to perform gamma spectroscopy. We find no statistically significant evidence for excess events above 3 MeV and place an upper limit on the activity at 3034 keV to be 1.0 counts/day. However, we observe significant activity from potassium-40 and estimate that the sample contains 19.8 ± 0.06 μg. Our sample therefore must be purified for use future studies on high energy activity.

1 Background

Our group at Virginia Tech is conducting research and development tests for low background detectors. Specifically, we are studying materials being considered for use at the Cryogenic Underground Observatory for Rare Events (CUORE). The ultimate goal of the CUORE collaboration is to search for neutrinoless double beta decay (DBD). Normal double beta decay is a rare weak reaction in which a (A, Z)

parent nucleus decays to a $(A, Z+2)$ daughter, releasing two electrons and antineutrinos [2].

$$(A, Z) \longrightarrow (A, Z+2) + 2e^- + 2\,\bar{\nu}_e + Q_{\beta\beta} \tag{1}$$

$$(A, Z) \longrightarrow (A, Z+2) + 2e^- + Q_{\beta\beta} \tag{2}$$

If neutrinos are Majorana fermions, they can act as their own antiparticle and self-annihilate. So, it is hypothesized that a neutrinoless mode of DBD can occur, with only the two outgoing electrons being released. Detecting neutrinoless DBD would then prove the Majorana nature of neutrinos and show that lepton number is not a global symmetry.

Searches for neutrinoless DBD therefore aim to find evidence for physics beyond the Standard Model. In particular, neutrinoless DBD supports Grand Unified Theories, which assume lepton number is not a fundamental symmetry and require new neutral fermions [2]. Furthermore, since neutrinoless DBD favors the production of matter - annihilating two antineutrinos and releasing two electrons - it is an important mechanism for theories explaining the dominant presence of matter over antimatter in our universe. [2].

In addition to being theoretically significant, neutrinoless DBD has a clear experimental signature. Since the decay would not release any antineutrinos, the transition energy of the nucleus is carried solely by the outgoing electrons. The electron energy spectrum in neutrinoless DBD will therefore be distinguished by a monoenergetic energy peak at the $Q_{\beta\beta}$ endpoint of normal DBD. However, this peak will only have a very weak signal, with current lifetime limits of neutrinoless DBD standing at $10^{25} - 10^{26}$ years. Distinguishing this peak therefore requires careful materials research; it is critical to ensure low backgrounds and to use materials that can achieve high energy resolutions and efficiencies. For CUORE, the solution is to use bolometers, cryogenic particle detectors highly sensitive to changes in temperature from incoming radiation [8]. Coupling thermal sensors to ton-scale arrays of $^{130}TeO_2$, the DBD source, maximizes their probabilities of observing the reaction.

However, reducing background events remains a persistent challenge for CUORE. While operating temperatures of 10 mK, copper and lead shielding, and detector construction with radiopure materials minimize external events, the alpha decay spectrum remains a prominent source of background above 2.6 MeV [1]. The next iteration of CUORE, the CUORE Upgrade with Particle Identification (CUPID), seeks to address these issues by using ^{100}Mo as a double beta decay source. The high transition energy of ^{100}Mo at 3034 keV is advantageous for DBD detection as it minimizes the gamma background, lying far above the ^{208}Tl 2615 keV energy at which natural radioactivity events drop significantly [8]. Furthermore, ^{100}Mo can be arranged in crystals of scintillating Li_2MoO_4, or lithium molybdate (LMO). Unlike the current $^{130}TeO_2$ crystals, LMO emits coincidental scintillation and thermal signals that can be used discriminate between DBD and α decay events [8].

We aim to characterize the radioactive spectrum of LMO. Specifically, we present our radioassay of a 99.9% purity sample of off-the-shelf LMO powder before activation with high-energy proton and neutron beams at an accelerator facility. Since the sample is not guaranteed to be radiopure, establishing its radioactive content is necessary before further work is pursued. We also examine the high energy spectrum to identify events from muon or cosmic ray activation; the associated Compton scattering background can interfere with sensitive studies as CUPID.

2 Data Collection

2.1 Experimental Setup

Our experiments were conducted in the Virginia Tech Center for Neutrino Physics lab, a surface-level lab on the Virginia Tech campus. We used the "MELISSA" Canberra Low Background coaxial detector, a high purity germanium detector, to perform gamma spectroscopy on our sample.

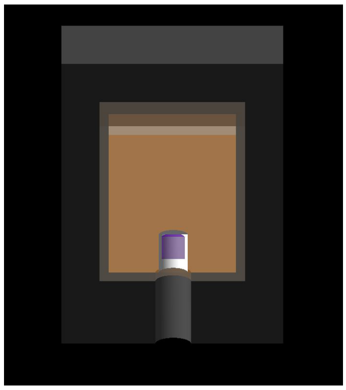

Figure 1: Left: The "MELISSA" Canberra LB coaxial HPGe detector in a copper and lead shield. Right: A Monte Carlo simulation of the empty detector in Geant4.

Germanium is commonly used to detect gamma radiation since it, a semiconductor, forms a depletion region under a bias voltage. Ionizing radiation incident on this depletion region liberates electron and hole pairs. Under the bias voltage, these pairs produce a current corresponding to the energy of the radiation [6]. With germanium, these signals can be produced with high energy resolutions; the MELISSA detector has an energy resolution of 1.70 keV at the full-width half maximum (FHWM) of the 1.33 MeV energy and a manufacturer's relative efficiency of 50% [3].

To process the signals from MELISSA, a preamplification step is done to boost the event signals above the noise in the cables. Using the analog-to-digital converter (ADC) channel, the ORTEC EASY-MCA-K multichannel analyzer converts the voltage pulses from MELISSA into counts in 8192 bins. However, a variable amplifier must be set to the appropriate gain; high gains will restrict the data range and low gains will pileup events to the same bins. Our setup uses the MAESTRO software to plot histograms of the event counts in each analog bin. For our data runs, we used sealed sources of ^{60}Co and KCl (a natural source of ^{40}K) to calibrate the bins of ADC counts to corresponding energies.

The germanium detector's high signal to noise ratio also necessitates shielding from external radiation. The MELISSA detector is housed within a copper box, comprised of 6 copper plates measuring approximately 16 inch $\times$ 16 inch $\times$ 1 inch. To shield from environmental sources of gamma radiation, a lead brick shielding further surrounds this copper housing.

2.2 Data Runs

We began by taking data on the background of the empty detector. Since there is significant activity from natural radioactive impurities in the copper and lead shielding, cosmic muons, products of cosmogenic activation, and the radon gas in the environment, it is important to establish this baseline spectrum before comparing the LMO sample data. After setting the variable amplifier to a coarse gain of five times, the initial run of the background was taken from June 1 to June 16, over a total live time of 15.6 days.

The gain was unmodified for the LMO sample run. A Marinelli beaker with the 1011.3 g of LMO was then placed over the detector. This data run was taken from June 17 to July 6, for a live time of 18 days.

Later runs with higher gains of twenty times were done to characterize the efficiency of the detector. The detector background was recorded for a live time of 11.8 days. A Marinelli beaker with 102.2 g of a radioactive sand calibration source [5] was then placed over the detector. Data on this calibration source was taken for a live time of 5 days.

3 Analysis

We used the tools in C++ ROOT program to analyze the MELISSA data. After converting the ADC histogram bins to energies with the calibration from sealed sources, we fitted a Gaussian model with a linear background to the significant peaks. These primarily consist of radioisotopes in the ^{238}U, ^{232}Th, ^{60}Co, and ^{40}K decay chains, whose long half-lives make them a persistent source of background. The initial calibration, done by eye, caused significant discrepancies in the means of fitted peaks and literature values of the transition energies. Therefore, we plotted the difference in the measured and theoretical energies against the fitted mean values and created a secondary energy calibration by fitting a second-order polynomial.

Where significant peaks exist, we obtain the counts by integrating the area under the Gaussian fits. We note that histogram counts are divided by bin widths: in our original runs, these bin widths are not constant, but peaks are narrow enough that differences are negligible to four significant digits.

Where no distinguishable peaks exist, we use the counts of specific bins. If it is determined that there is no significant activity above the background, equation 3

$$signal\ counts \leq 1.64 \times \sqrt{background\ counts} \tag{3}$$

sets an upper limit on the sample's signal counts at the 90% confidence level [3].

To characterize the detector efficiency, we used Geant4, a simulation toolkit modeling physics processes of particles passing in matter. This toolkit allows us to encode the geometry of the detector and radioactive sample. We then run Monte Carlo simulations (Figure 1) with a set number of decays, producing an expected number of counts recorded by the detector. With experimental data on a calibration source with known activity, the accuracy of these simulated counts can be assessed. The simulation data, however, does not account the Gaussian spreading caused by the finite resolution of the detector. This is corrected by applying the spread of the detector's energy resolution: an energy resolution function is obtained by fitting the standard deviations of peaks in the ^{238}U, ^{232}Th, and ^{40}K decay chains.

We calculate the detector efficiency by taking the ratio of peak counts in the calibration source data to peak counts in the corrected simulation data (with simulation counts scaled to match the activity of the calibration source). This efficiency is then used to derive the total activity and quantity of the radioactive impurity in our sample. We note our calculations do not account for systematic uncertainties in our measurements of the detector. Our efficiency can also be improved with more exact information on the size of the germanium crystal, its position inside the aluminum endcap, and the thickness of the dead layer.

4 Results

4.1 Background Spectrum Radioassay

To compare with the LMO data, we examined the background activity at the characteristic energies of ^{40}K and ^{100}Mo. At the 1461 and the 3034 keV energies, we find respective activities of 1.355 ± 0.772 mBq and 71.2 ± 7.3 μBq.

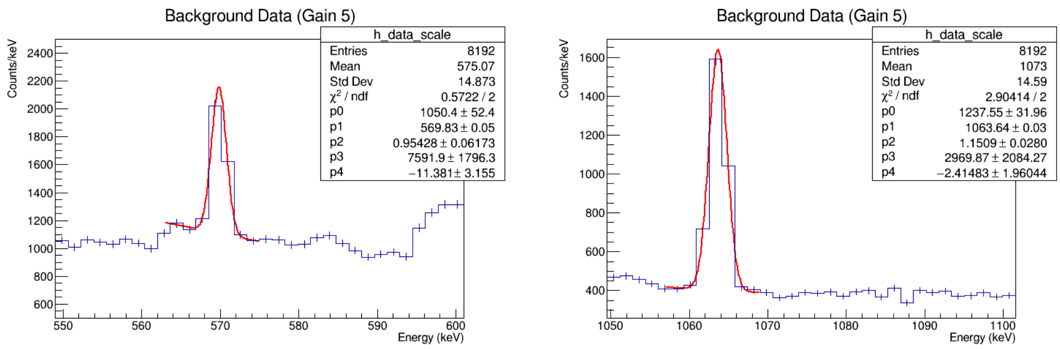

Figure 2: Left: The background spectrum fitted peak at 569 keV. Right: The background spectrum fitted peak at 1063 kV. Parameters in order are the Gaussian norm, mean, standard deviation, linear intercept, and slope.

We also note two peaks at 569 and 1063 keV with activities of 1.861 ± 0.120 mBq and 2.646 ± 0.094 mBq. These peak energies are not consistent with the decay radiation of isotopes in the ^{238}U and ^{232}Th decay chains nor that of any other common radioisotopes. The primary candidate we identified is ^{207}Bi: an isotope of bismuth with gamma radiation peaks at 569.968 and 1063.656 keV, occurring with relative intensities of 97.75% and 74.5% [7].

Given the lack of cosmic ray shielding in our experiment and the lack of natural radioactive decays with this transition energy, we suspect the ^{207}Bi is produced by the cosmogenic activation of lead. A study by Guiseppe et al. on the activation of lead in neutron beams at the Los Alamos Neutron Science Center sets the sea level production rate of ^{207}Bi at 0.17 atom/kg·day. However, Guiseppe et. al also note that simulations of the production of ^{207}Bi in 100 ppm of bismuth impurity in the lead contributed to 10% of the measured production rate [4]. Since ^{207}Bi is a nuclear fallout isotope, it remains a strong possibility that our lead additionally contains Bi impurities. Our initial simulations with ACTIVIA on the neutron activation in lead also do not produce ^{207}Bi as a spallation product. This may be a limitation of the program - ACTIVIA only simulates primary reactions while ^{207}Bi is produced by protons released in secondary reactions with incident cosmic neutrons [4]. Further work will therefore need to be done to conclude if the production rate alone is consistent with the observed activity.

4.2 LMO Spectrum Radioassay

The LMO sample displays a significant signal at 1461 keV. The activity at the peak (Figure 3) is 7.870 ± 0.124 mBq, an increase of 6.516 ± 0.146 mBq from the background. This indicates that the LMO sample contains additional potassium impurities; though our sample is purified, potassium, also a group 1A alkali metal, bonds readily to molybdate.

Our comparison of the 1461 keV peaks in the simulated and recorded data of the radioactive sand sample (see Figure 3) yields a detector efficiency of $77.7 \pm 1.2\%$. With this efficiency and the counts from a simulation of the LMO sample, we find a total activity of 5.26 ± 0.15 Bq, corresponding to 19.8 ± 0.06 μg of ^{40}K. Assuming natural abundances, this places 0.172 ± 0.005 g of total potassium in the LMO sample. We therefore conclude that our LMO sample is not pure enough to meet the standards set by CUPID; the observed ^{40}K activity of 5.20 ± 0.14 Bq/kg far exceeds the 5 mBq/kg limit. Though occurring at low energies, any excessive activity is important to note since pile-up events can add background to higher energies, notably at 3034 keV. For future studies, either a higher purity sample will need to be acquired or the potassium chemically removed.

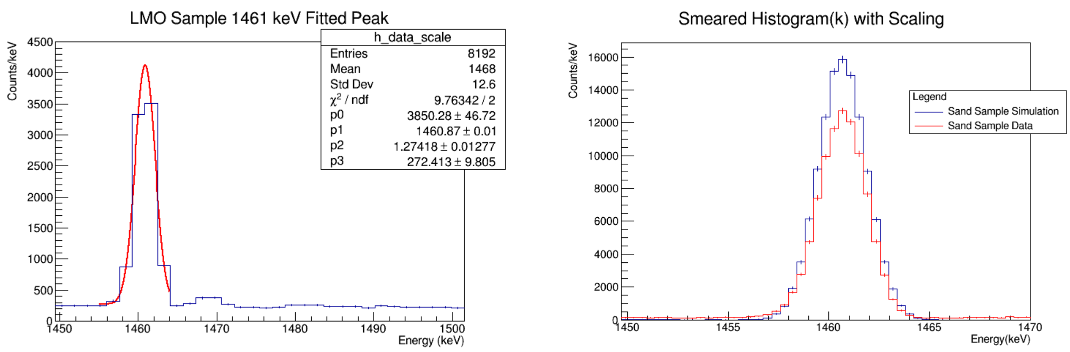

Figure 3: Left: LMO spectrum fitted 1461 keV peak. Parameters in order are Gaussian norm, mean, standard deviation, linear intercept. Right: The 1461 keV peak in the recorded radioactive sand data and the simulation (26,975,895 decays).

In the high energy part of the LMO spectrum (> 3 MeV), we observe no significant peaks (see Figure 4). At the ^{100}Mo transition energy of 3034 keV, we observe an activity of 62.3 ± 6.3 μBq. This is a decrease from the background activity of 8.9 ± 9.8 μBq, and is therefore not statistically significant. We then place an upper limit of 11.9 μBq on the activity from the LMO (see equation 3).

This analysis is repeated for the bins listed in Table 4.1. None were found to exceed the activity upper limits. Of the bins (without rebinning) above 3034 keV, 8.9% exceeded the activity upper limit, a figure consistent with the expected 90% confidence level.

However, we note that the LMO activity at high energies trends low, being below the background in 58.9% of bins. Simulations with Poisson distributions centered on expected counts (background counts scaled to match live time of LMO) never contain

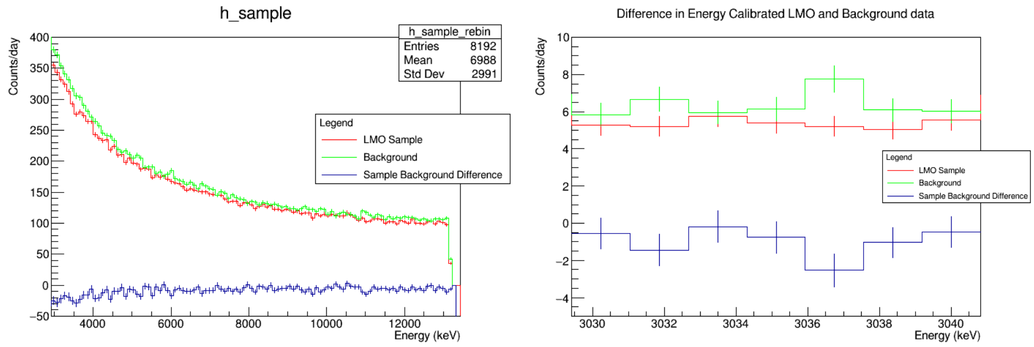

Figure 4: Left: High energy spectrum (> 3MeV) of LMO sample, background, and difference, rebinned by 60 times. Right: LMO spectrum at 3034 keV, no rebinning.

Bin	Energy (keV)	Background (mBq)	LMO (mBq)	LMO-Background Difference (mBq)	Upper Limit (mBq)
40	3809.6 - 3907.4	3.24 ± 5	3.06 ± 4	-0.19 ± 0.07	0.0807
71	6844.5 - 6942.5	1.85 ± 0.04	1.74 ± 0.03	-0.11 ± 0.05	0.0609
102	9887.4 - 9985.7	1.41 ± 0.03	1.33 ± 0.03	-0.09 ± 0.04	0.0532

Table 4.1: Activity of LMO, background, difference, and upper limit in randomly selected bins, 60 times rebinning.

greater than 58.9% bins below the background. Further work will need to be done to analyze if this trend is consistent with statistical fluctuations. Though unlikely to be significant at high energies, it is possible that the sample has a shielding effect on the detector. Simulations of the cosmic ray flux in Geant4 can ascertain if this accounts for the lower activity.

We also emphasize that our findings cannot rule out small signals that may be present at high energies. On the surface, the detector's sensitivity is limited by the high background from cosmic rays. This can be improved by repeating these measurements in an underground lab, where the background level above 3 MeV is expected to be lower due to the reduced cosmic ray flux. Furthermore, by activating the sample under high intensity protons and neutron beams, we hope to amplify any potential signals and more conclusively demonstrate if significant activity in the high energy region of LMO exists.

5 Conclusion

The CUORE and CUPID projects are searching for neutrinoless double beta decay to uncover new insights on the nature of neutrinos and on physics beyond the Standard Model. Achieving the energy resolution and low background necessary to detect neutrinoless double beta decay requires careful and rigorous analysis of the materials in the detector. Our work therefore examined the radioactive background of an unenriched LMO sample. Within the sensitivity of our study, we find no evidence for background events associated with LMO above energies of 3 MeV. Monte Carlo simulations of the detector also indicate that our sample is not sufficiently radiopure for future experiments.

We hope that future work, in addition to characterizing the high energy spectrum of LMO after activation at an accelerator, will improve our characterization of the detector geometry, address the source of bismuth-207, and investigate simulations of the high energy activity at the 3034 keV energy.

6 Acknowledgments

I would like to thank my program mentor, Dr. Thomas O'Donnell, and his graduate students Vivek Sharma and Joe Camilleri for their comments and advice on this project. I would also like to thank the National Science Foundation for funding this project as part of the Virginia Tech Center for Neutrino Physics REU program.

References

[1] A. Armatol et. al, The CUPID Collaboration, Characterization of Cubic $Li^{100}_2MoO_4$ Crystals for the CUPID Experiment, Eur. Phys. J. C 81, 104 (2021).

[2] M. J. Dolinski, A. W. P. Poon, and W. Rodejohann, Neutrinoless Double-Beta Decay: Status and Prospects, Annu. Rev. Nucl. Part. Sci. 69, 219 (2019).

[3] P. Finnerty, S. MacMullin, H. O. Back, R. Henning, A. Long, K. T. Macon, J. Strain, R. M. Lindstrom, and R. B. Vogelaar, Low-Background Gamma Counting at the Kimballton Underground Research Facility, Nucl. Instrum. Methods Phys. Res. Sect. Accel. Spectrometers Detect. Assoc. Equip. 642, 65 (2011).

[4] V. E. Guiseppe, S. R. Elliott, N. E. Fields, and D. Hixon, Fast-Neutron Activation of Long-Lived Nuclides in Natural Pb, Astropart. Phys. 64, 34 (2015).

[5] IAEA Reference Products For Environment And Trade.

[6] W.R. Leo, Techniques for Nuclear and Particle Physics Experiments (Springer Berlin Heidelberg, Berlin, Heidelberg, 1994).

[7] National Nuclear Data Center, Brookhaven National Laboratory.

[8] The CUPID Interest Group, CUPID Pre-CDR, ArXiv Preprint ArXiv: 1907.09376 (2019).