Scintillating Bolometer Crystal Growth and Purification for Neutrinoless Double Beta Decay Experiments


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RMD Basic and Applied Research and Development

Materials Science

- Scintillators
- Semiconductors
- X-ray Imaging Screens
- Ceramic Lasers and IR windows

Sensors

- APDs SSPMs
- Photosensors
- Wide Band Gap Geiger Photodiodes
- Surgical Beta-Probe

Instruments & Systems

- HiRIS – High Resolution Imaging System
- RadEye Detectors
- Hermes G/n w/ isotope ID
- Robotic nuclear power plant concrete analyzer

RMD Basic and Applied Research and Development

A Dynasil Company
RMD Commercial Products

3” CLYC Crystals  CLYC Pillars

Thermo-Scientific

Scintillation detectors

INL Neutron Imaging System

Zetec ECT power plant probe
Understanding the Neutrino

• A key goal of Nuclear Physics is elucidating the nature of the neutrino
  What are the masses of the neutrino mass eigenstates?
  Is the neutrino its own antiparticle, and thus a Majorana particle?
• The question of the Majorana nature of neutrinos is one of the most important
  questions in physics today
• If the neutrino is a Majorana particle, the neutrino is responsible for the
  matter-antimatter asymmetry we observe in the universe.
• Searching for neutrinoless double beta decay (0νββ) one of the highest priority
  experiments to answer this question
• One such experiment is CUORE: Cryogenic Underground Observatory for Rare
  Events. CUORE uses 1 ton TeO$_2$ bolometers
• The next generation experiment will be CUPID: CUORE with Particle
  Identification. CUPID will use scintillating bolometers
Phase IIA Technical Objectives

The goal is to complete the research and development needed to implement production of Li$_2$MoO$_4$ (LMO) scintillating bolometer crystals suitable for neutrinoless double-beta decay experiments.

- Synthesize Li$_2$MoO$_4$ from the high purity raw materials
- Purify the Li$_2$MoO$_4$ further by zone refining to improve the radioactive background
- Grow single-crystal ingots using Czochralski for fabricating 200 – 250 gm detectors
- Develop processes for shaping and polishing crystals that maintain radio-purity
- Deliver detector crystals to MIT for cryogenic evaluation. Scintillating bolometer testing will include all operational characteristics, such as light output and radioactivity background.
- Demonstrate suitability for the CUPID neutrinoless double-beta decay experiment
- Grow LMO using isotope enriched $^{100}$Mo and produce full-spec detectors to qualify as a supplier for the CUPID experiment.
Selection of Isotopes with Double-beta decay

Candidate Isotopes for 0νββ Experiments

<table>
<thead>
<tr>
<th>element</th>
<th>isotope</th>
<th>end point energy (MeV)</th>
<th>% abundance</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca</td>
<td>48</td>
<td>4.271</td>
<td>.187</td>
</tr>
<tr>
<td>Nd</td>
<td>150</td>
<td>3.367</td>
<td>5.6</td>
</tr>
<tr>
<td>Zr</td>
<td>96</td>
<td>3.35</td>
<td>2.8</td>
</tr>
<tr>
<td>Mo</td>
<td>100</td>
<td>3.034</td>
<td>9.7</td>
</tr>
<tr>
<td>Se</td>
<td>82</td>
<td>2.995</td>
<td>8.8</td>
</tr>
<tr>
<td>Cd</td>
<td>116</td>
<td>2.802</td>
<td>7.5</td>
</tr>
<tr>
<td>Te</td>
<td>130</td>
<td>2.527</td>
<td>24.6</td>
</tr>
<tr>
<td>Xe</td>
<td>136</td>
<td>2.457</td>
<td>8.9</td>
</tr>
<tr>
<td>Ge</td>
<td>76</td>
<td>2.039</td>
<td>7.8</td>
</tr>
</tbody>
</table>

Requirements for isotope
1. Must decay by double beta process.
2. Good natural abundance and ability to enrich.
3. High endpoint energy (above 2.6 MeV $^{232}$Th gamma ray).
4. Major constituent in a scintillating crystal.

$^{100}$Mo half-life = 7.8$\times 10^{18}$ y
$^{82}$Se half-life = 0.97$\times 10^{20}$ y

Scintillating Bolometers are needed for better particle discrimination and background reduction in next generation experiments.
Li₂MoO₄ (LMO) Synthesis

1. MoO₃ 99.9995% + Li₂CO₃ 99.99% high purity powders
   \[ \text{MoO}_3 + \text{Li}_2\text{CO}_3 \rightarrow \text{Li}_2\text{MoO}_4 + \text{CO}_2 \]

2. Mix powders in a plastic mixing bottle overnight on a roller

3. Press the mixture in a Teflon piston jig with a cold press to form a compact and dense mixture puck

4. Place and melt the puck inside a platinum crucible at 650°C

5. Repeat steps 1-4 until crucible is sufficiently full
LMO Purification

• Start with good purity raw materials
  – Good sources identified in previous phase
  – Decent purity achieved without further purification

• Evaluating zone refining
  – Trying different crucible materials (Pt, carbonized quartz, etc.)

• MoO$_3$ (99.9995%) + Li$_2$CO$_3$ (99.99%) High Purity Powders

Greenish or brownish crystals can result if best purity materials are not used.
Czochralski Growth of Li$_2$MoO$_4$

Congruent growth for Li$_2$MoO$_4$

No structural phase change for Li$_2$MoO$_4$


30 x 30 x 20 mm sample used for cryogenic testing

Optical Transmission of Li$_2$MoO$_4$ G5-Top Crystal
Cryogenic Testing of Scintillating Bolometers

Above ground cryogenic testing by MIT at CSNSM

- Samples held at ~ 20 mK for multi-day testing.
- Light and heat pulses measured separately.
Mean light and heat pulses from LMO

Light pulse is \( \sim 100\times \) faster than heat.
Light versus Heat Chart for LMO

- 5 days background measurement
- Temperature stable 20 mK +/- 0.1 mK

Good separation of alphas!

Energy spectrum
Calibrated Heat Spectrum for LMO

- Baseline FWHM: 10.4 keV
- Sensitivity: 11 nv/keV

Calibrated Spectrum

Gamma peaks from U-238 chain
LMO Light Channel Spectrum

Light Channel Spectrum

- FWHM Baseline Resolution: 0.81 keV
- Sensitivity: 1.1 uV/keV
- Light Yield ~ 0.5 keV/MeV

Calibrated with muon background
Alpha Particle Discrimination

Alpha Particle Discrimination Power = 3.0

\[ DP = \frac{|\mu_1 - \mu_2|}{\sqrt{\sigma_1^2 + \sigma_2^2}} \]

\[ DP = \frac{0.520 - 0.145}{\sqrt{0.682^2 + 0.104^2}} \]

\[ DP = 3.0 \]
LMO Internal Alpha Background Limits

Alpha Contamination: Limits are 0.08 to 0.3 mBq/kg
– Comparable to the CLYMENE crystal

### Alpha Contamination Limits

<table>
<thead>
<tr>
<th>Chain/Contamination</th>
<th>Nuclide</th>
<th>Q-Value (keV)</th>
<th>Counts</th>
<th>Limit on Activity (mBq/kg)</th>
<th>CLYMENE LMO-Small (mBq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Th-232</td>
<td>Th-232</td>
<td>$4081.6 \pm 1.4$</td>
<td>5</td>
<td>$&lt;0.24$</td>
<td>$&lt;0.5$</td>
</tr>
<tr>
<td>Th-228</td>
<td>Th-228</td>
<td>$5520.08 \pm 0.22$</td>
<td>8</td>
<td>$&lt;0.10$</td>
<td>$&lt;0.55$</td>
</tr>
<tr>
<td>U-238</td>
<td>U-238</td>
<td>$4269.7 \pm 2.9$</td>
<td>9</td>
<td>$&lt;0.12$</td>
<td>$&lt;0.72$</td>
</tr>
<tr>
<td>Ra-226</td>
<td>Ra-226</td>
<td>$4870.62 \pm 0.25$</td>
<td>-</td>
<td>$&lt;0.21$</td>
<td>$&lt;0.50$</td>
</tr>
<tr>
<td>Rn-222</td>
<td>Rn-222</td>
<td>$5590.4$</td>
<td>13</td>
<td>$&lt;0.21$</td>
<td>-</td>
</tr>
<tr>
<td>Po-218</td>
<td>Po-218</td>
<td>$6002.4$</td>
<td>7</td>
<td>$&lt;0.08$</td>
<td>-</td>
</tr>
<tr>
<td>Po-210</td>
<td>Po-210</td>
<td>$5407.45 \pm 0.07$</td>
<td>8</td>
<td>$&lt;0.10$</td>
<td>$&lt;1.7$</td>
</tr>
<tr>
<td>Pt-190</td>
<td>Pt-190</td>
<td>$3252 \pm 6$</td>
<td>15</td>
<td>$&lt;0.25$</td>
<td>-</td>
</tr>
</tbody>
</table>

- Feldman-Cousins tables are used to set 90% limits
- Count limits are converted to activity limits with the exposure of $0.22 \text{ kg*days}$
- Ra-226 limit is set by assuming secular equilibrium with Rn-222
- Comparison is to CLYMENE (Exposure 0.039 kg*days)
- Accounting for different exposures, the two sets of limits are comparable (arXiv:1801.07909 [physics.ins-det])
Manufacturing Plan

Supply of $^{100}$Mo

- The molybdenum, supplied as $^{100}$MoO$_3$ powder, will be purchased from ISOFlex in a quantity sufficient for the prototype objective of the Phase IIA project.
- The enriched $^{100}$MoO$_3$ is by far the most expensive component of the LMO detectors planned for CUPID, at approximately $69,000 per kg for the $^{100}$Mo.
- There is a choice of 1 or 2 stages of centrifugation.
- The 2-stage centrifugation cost more but has significantly better purity and will be utilized for this project.

Production Schedule

- The required shape size and delivery schedule for LMO crystals to meet the US contribution to CUPID.
- 600 crystal cubes 4.5 cm on a side by 2025. We plan to begin delivering crystals to CUPID in 2022.
- One Czochralski crystal puller can produce up to 100 crystals per year.
- Three pullers will be needed to complete the delivery on time.
# Purity of Enriched $^{100}$Mo from ISOFlex

## 1 stage of centrifugation

<table>
<thead>
<tr>
<th>Element</th>
<th>Permissible Abundance</th>
<th>Element</th>
<th>Permissible Abundance</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>&lt;0.01 ppm</td>
<td>Co</td>
<td>&lt;30 ppm</td>
</tr>
<tr>
<td>Th-232</td>
<td>&lt;0.01 ppm</td>
<td>Cu</td>
<td>&lt;30 ppm</td>
</tr>
<tr>
<td>W</td>
<td>&gt;1000 ppm*</td>
<td>Zr</td>
<td>&lt;30 ppm</td>
</tr>
<tr>
<td>Sr</td>
<td>&lt;30 ppm</td>
<td>Zr</td>
<td>&lt;30 ppm</td>
</tr>
<tr>
<td>Ba</td>
<td>&lt;30 ppm</td>
<td>Nb</td>
<td>&lt;30 ppm</td>
</tr>
<tr>
<td>Si</td>
<td>&lt;50 ppm</td>
<td>Cd</td>
<td>&lt;30 ppm</td>
</tr>
<tr>
<td>Al</td>
<td>&lt;30 ppm</td>
<td>Sn</td>
<td>&lt;30 ppm</td>
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<tr>
<td>Sc</td>
<td>&lt;30 ppm</td>
<td>Sb</td>
<td>&lt;30 ppm</td>
</tr>
<tr>
<td>Ti</td>
<td>&lt;30 ppm</td>
<td>Hf</td>
<td>&lt;30 ppm</td>
</tr>
<tr>
<td>Cr</td>
<td>&lt;30 ppm</td>
<td>Ta</td>
<td>&lt;30 ppm</td>
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<tr>
<td>Mn</td>
<td>&lt;30 ppm</td>
<td>Pb</td>
<td>&lt;30 ppm</td>
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<tr>
<td>Fe</td>
<td>&lt;30 ppm</td>
<td>Bi</td>
<td>&lt;30 ppm</td>
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<tr>
<td>Ni</td>
<td>&lt;30 ppm</td>
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</table>

## 2 stages of centrifugation

<table>
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<th>Element</th>
<th>Permissible Abundance</th>
<th>Element</th>
<th>Permissible Abundance</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>&lt;0.01 ppm</td>
<td>Ni</td>
<td>&lt;5 ppm</td>
</tr>
<tr>
<td>Th-232</td>
<td>&lt;0.001 ppm**</td>
<td>Co</td>
<td>&lt;5 ppm</td>
</tr>
<tr>
<td>Ra-226</td>
<td>&lt;10 mBq/kg *</td>
<td>Cu</td>
<td>&lt;5 ppm</td>
</tr>
<tr>
<td>W</td>
<td>&lt;50 ppm</td>
<td>Zn</td>
<td>&lt;5 ppm</td>
</tr>
<tr>
<td>Sr</td>
<td>&lt;5 ppm</td>
<td>Zr</td>
<td>&lt;5 ppm</td>
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<tr>
<td>Ba</td>
<td>&lt;5 ppm</td>
<td>Nb</td>
<td>&lt;5 ppm</td>
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<tr>
<td>Si</td>
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<tr>
<td>Mn</td>
<td>&lt;5 ppm</td>
<td>Pb</td>
<td>&lt;5 ppm</td>
</tr>
<tr>
<td>Fe</td>
<td>&lt;5 ppm</td>
<td>Bi</td>
<td>&lt;5 ppm</td>
</tr>
</tbody>
</table>
Summary and Plans for the Remainder of Phase IIA

We will continue follow the original plan described in the proposal.

- Scale up the crystal growth to 3” diameter by 3” long
- Optimize purification methods, especially for MoO$_3$
- Continue to provide crystal samples to MIT for cryogenic evaluation.
- Incorporate enriched $^{100}$Mo and produce a prototype detector crystal fully suitable for CUPID.
- Finalize and document the production process
- Produce full size crystals and transition to Phase III production