Molybdenum-100 Reclamation and Purification for Neutrinoless Double-Beta Decay Experiments

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Outline of the Talk

- Introduction – Physics Motivation
- LMO Crystal Growth
- Molybdenum Recovery
- Purification
- Cryogenic Testing

Acknowledgements

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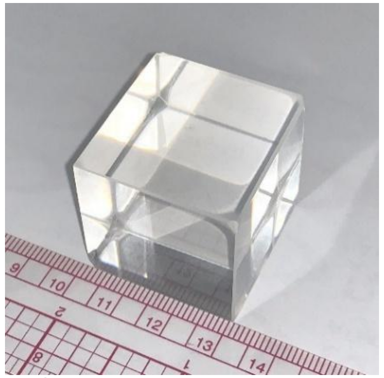
- We thank the CUPID collaboration for discussions and cryogenic characterization.

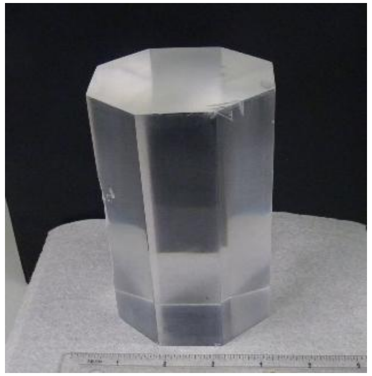
RMD Low-Background Crystals for Nuclear and High-Energy Physics

Li_2MoO_4 (LMO)
Scintillating Bolometer



for neutrinoless double-beta decay

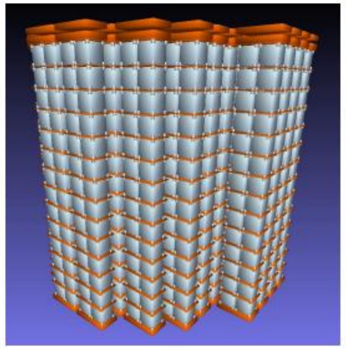
NaI
Room T Scintillator



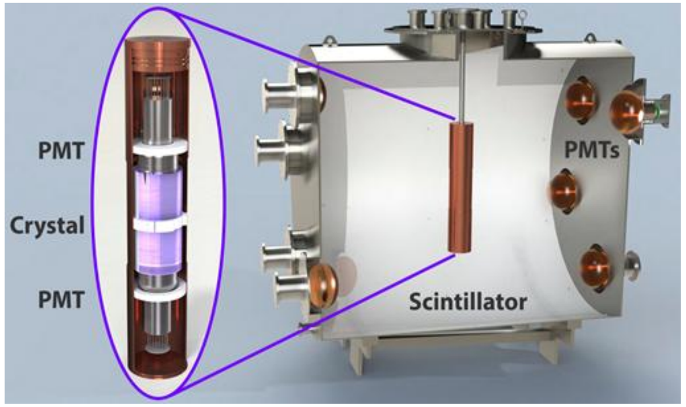
for dark matter search

Zinc
Superconducting Bolometer

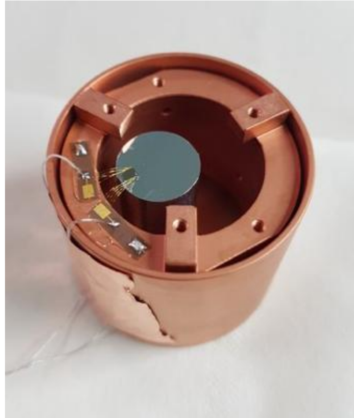
for neutrino scattering



CUPID



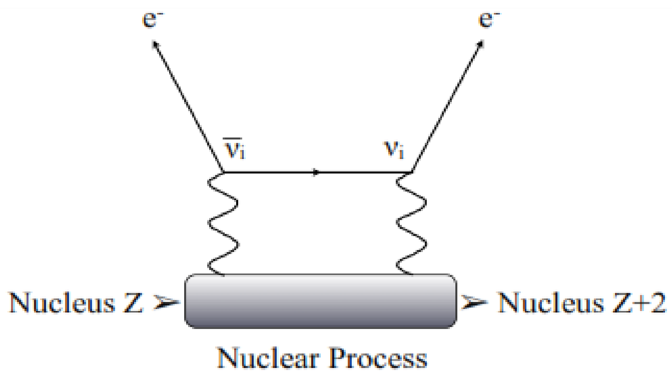
SABRE



RICOCHET

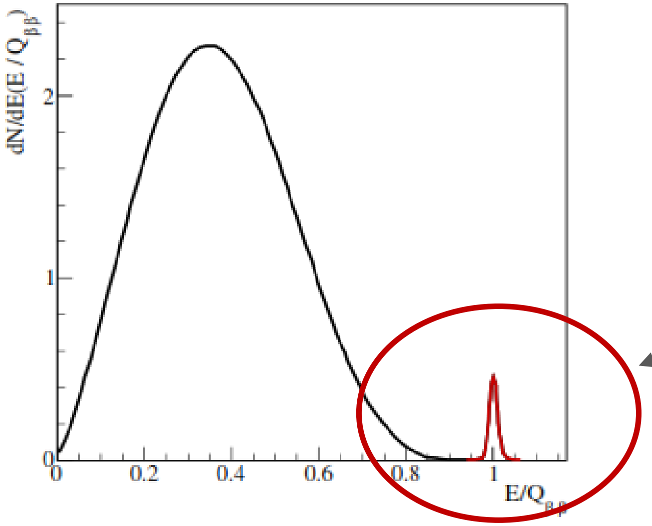
Understanding the Neutrino

- 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?
- The answers to these questions can change the Standard Model of physics.
- Searching for **neutrinoless double beta decay** (0νββ) is one of the highest priority experiments to answer these questions.
- LMO may also be useful in detecting coherent neutrino scattering for reactor monitoring and non-proliferation. (Mo-100 not needed.)



0νββ process

0νββ spectrum



Signature of 0νββ

Selection of Isotopes with Double-beta Decay

Candidate Isotopes for 0νββ Experiments

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

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

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.

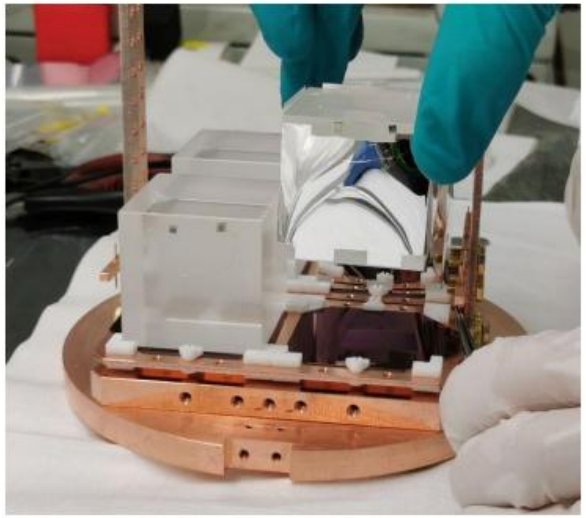
Li_2MoO_4 Scintillating Bolometer

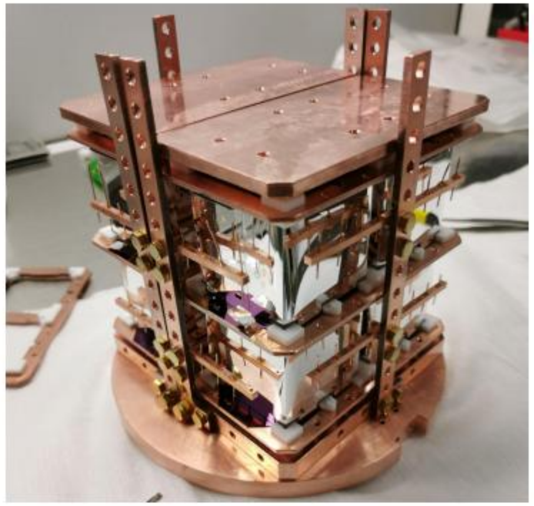
- **Is both the source and detector of 0νββ**
- Detects heat and light signals simultaneously.

***Scintillating Bolometers** are needed for better particle discrimination and background reduction*

CUPID: a next generation neutrinoless double beta decay experiment

- **CUPID**: **C**UORE **U**pgrade with **P**article **Id**entification
  - TeO_2 crystals to be replaced by Li_2MoO_4
- CUPID will consist of about 1600 hybrid heat-light detectors for a total isotope mass of 250 kg (^{100}Mo).
- Operating at ~ 10-20 mK temperature.





CUPID Cryostat

** GiovanniBenato_TIPP2021(2).pdf*

RMD Previous Work Developing LMO

- Synthesis and crystal growth processes developed.
- Fabrication of 45 mm cubes of LMO for cryogenic testing.
- Cryogenic testing done by MIT and CUPID Collaboration.
- Reclamation of MoO_3 from LMO scraps was demonstrated on a small scale (Phase I).
- Purification of MoO_3 was demonstrated on a small scale (Phase I)

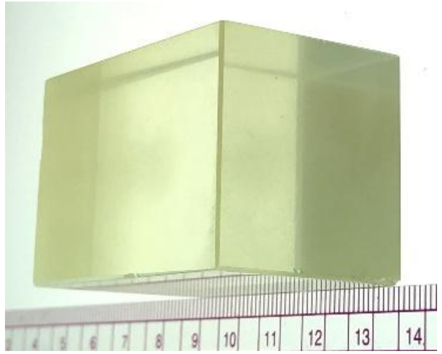
LMO Synthesis

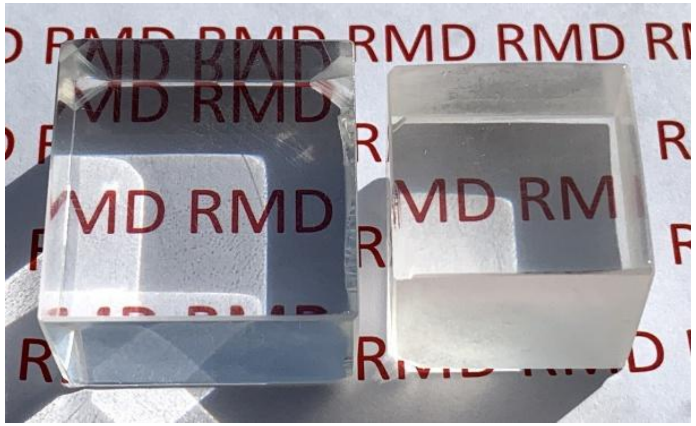
Start with best purity raw materials

- MoO_3 (99.9995%) + Li_2CO_3 (99.99%) High Purity Powders

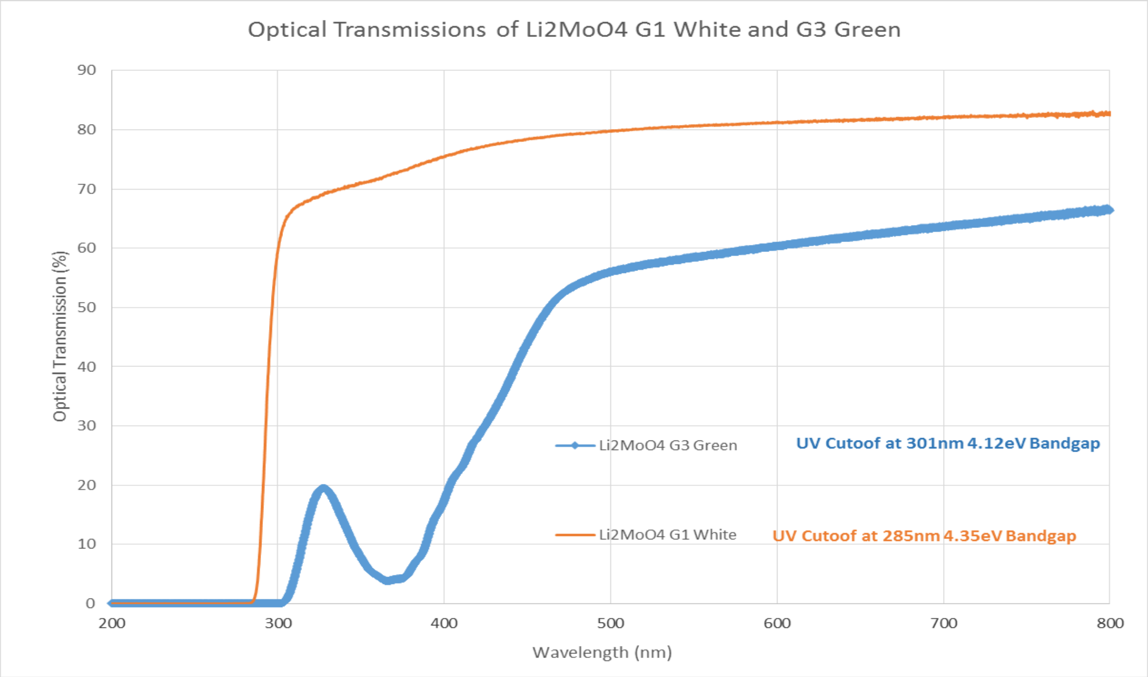
- $MoO_3 + Li_2CO_3 \longrightarrow Li_2MoO_4 + CO_2$
- Synthesis and growth in platinum crucible

- Greenish or brownish crystals can result if best purity materials are not used.
- Must be colorless for best scintillation.





Optical transmission is good indication of crystal purity



Czochralski Growth of Li_2MoO_4

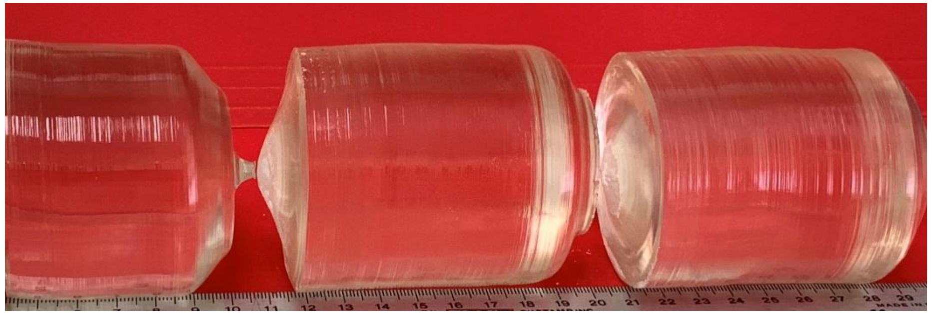
~4 cm OD
~200 grams

65mm OD x 65mm
813 grams

[010] growth

trigonal crystal structure, scheelite (CaWO4) , space group C2/c

Melting temp ~ 705C



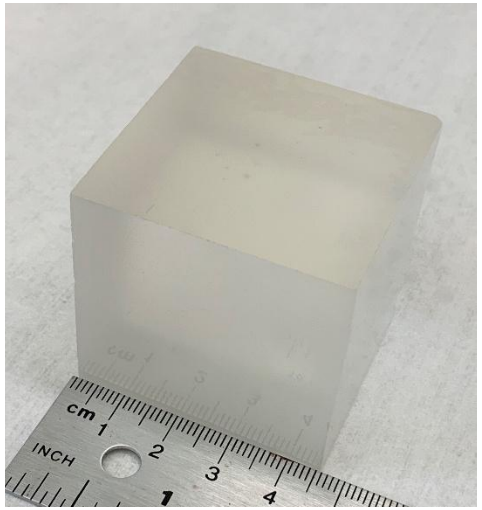
Repeatable growth process

Czochralski Crystal Growth Furnace

Polishing of Li_2MoO_4 to 45 mm Cubes for CUPID

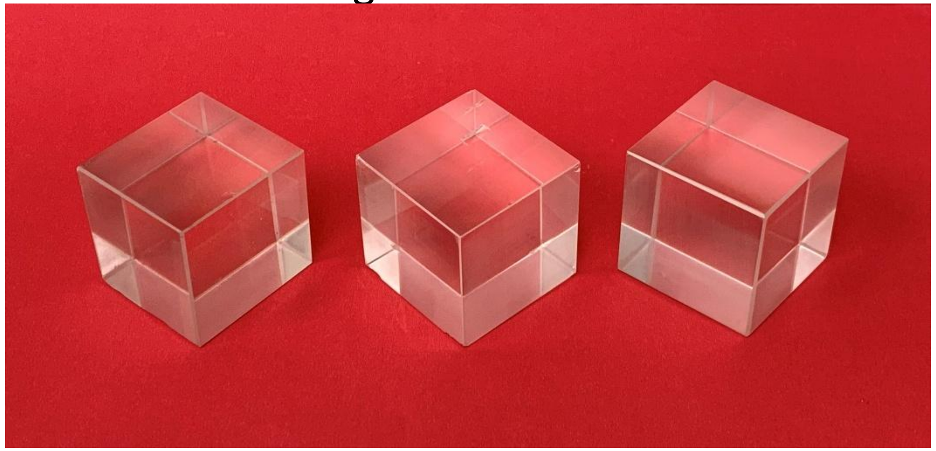
Polishing machine with customized fixture for 45 mm cubes.

Before Polishing



Initial polishing done in open air.

After Polishing



SBIR Phase II Goals

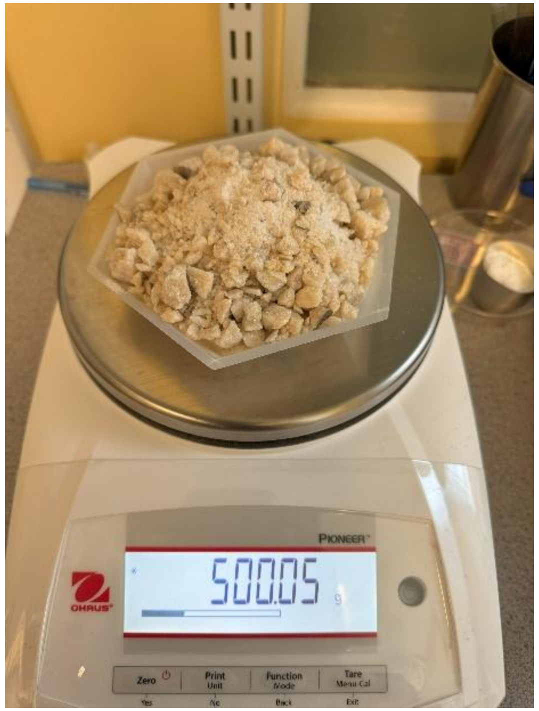
- Scale-up processes for recovery of MoO_3 from LMO crystal scraps and growth residuals
- Scale-up and optimize the processes to purify the MoO_3 by sublimation and by wet chemistry.
- Grow single-crystal ingots of LMO by Czochralski using the reclaimed and purified MoO_3 powder.
- Evaluate the chemical purity of the purified MoO_3 powder and grown crystals.
- Evaluate crystal samples for radio-purity and scintillation performance at cryogenic temperatures, by MIT and the CUPID collaboration.
- Demonstrate reclamation and recovery processes using enriched Mo-100*.

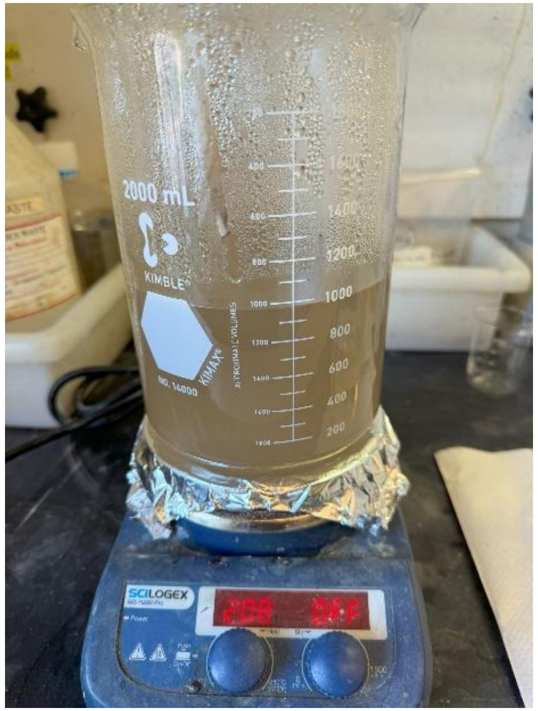
* if available

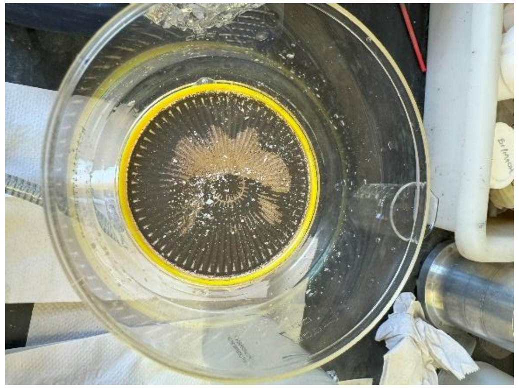
Reclamation of MoO_3 from LMO Scraps

- CUPID requires LMO to be enriched with Mo-100 isotope (>90%)
  - Mo-100 very expensive (~$100/gm)
  - Mo-100 hard to get due to geopolitical situations
  - Recovery of precious Mo-100 from LMO scraps is crucial
- Start with scraps of LMO from cutting and crucible residuals
- Convert from LMO to MoO_3 using wet chemistry
- Purify by sublimation and wet chemistry
- Grow LMO using the reclaimed MoO_3

Dissolving of LMO scraps in DI Water and Filtering Out Undissolved Residuals

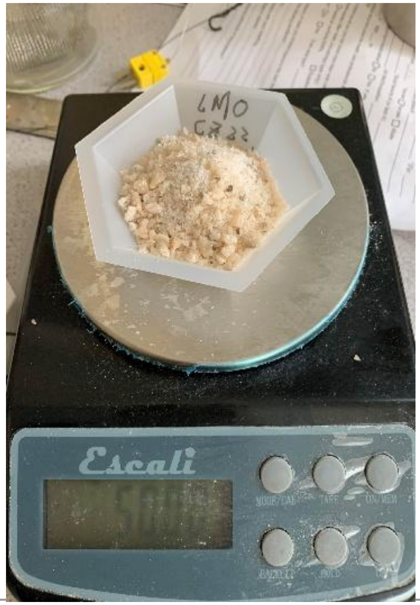


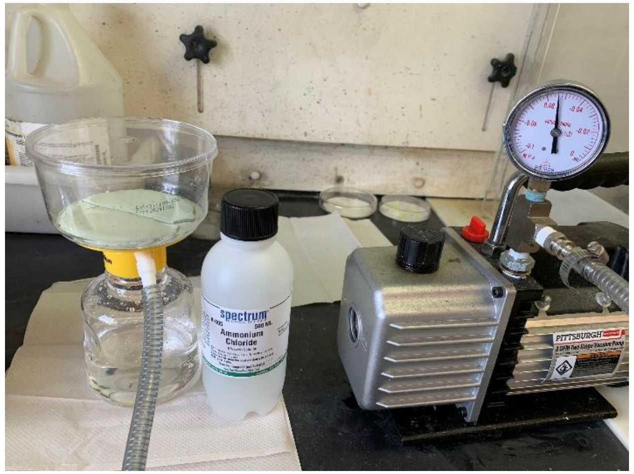


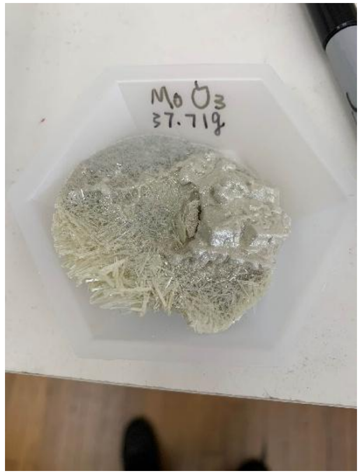


Reclamation of Mo from LMO Scraps

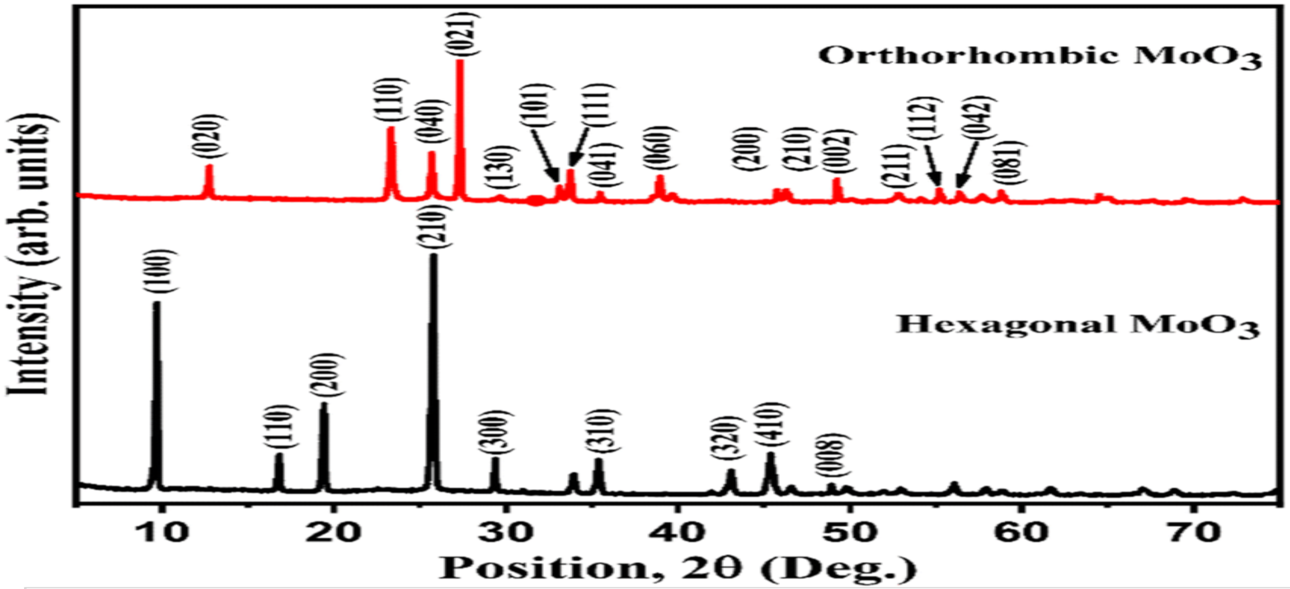
1. Dissolve LMO Residuals in DI Water and Add HCl to PH 0.9
2. Dissolve NH_4Cl in DI Water and Mix with LMO+HCl
3. Use vacuum filter to remove liquid and harvest a cake of APM (ammonium polymolybdate)
4. Bake in air to convert APM to MoO_3 (93.5% recovery measured).

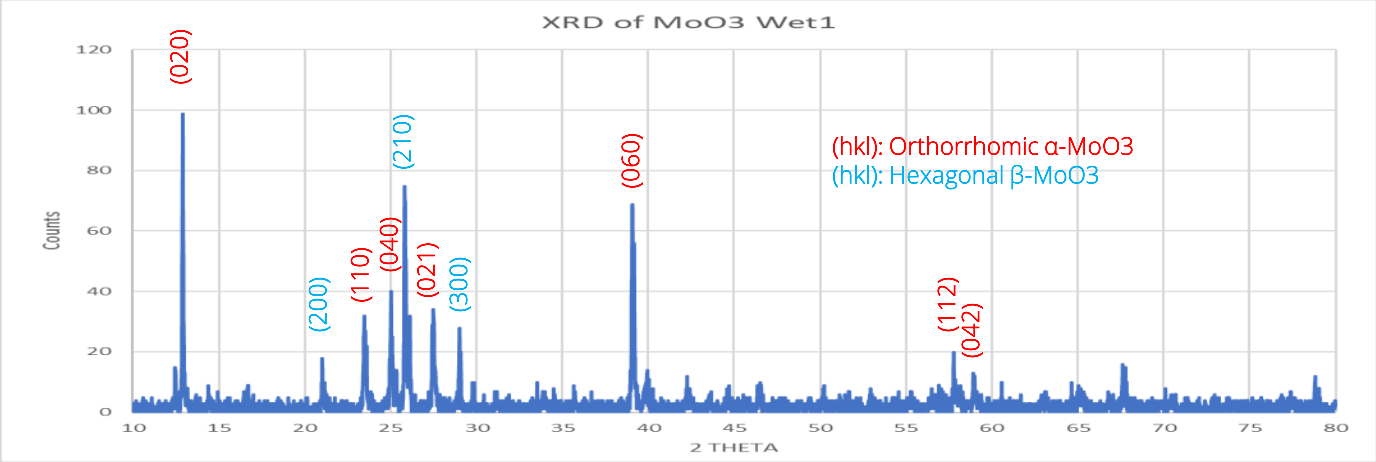






XRD Confirms that MoO_3 was Recovered from LMO

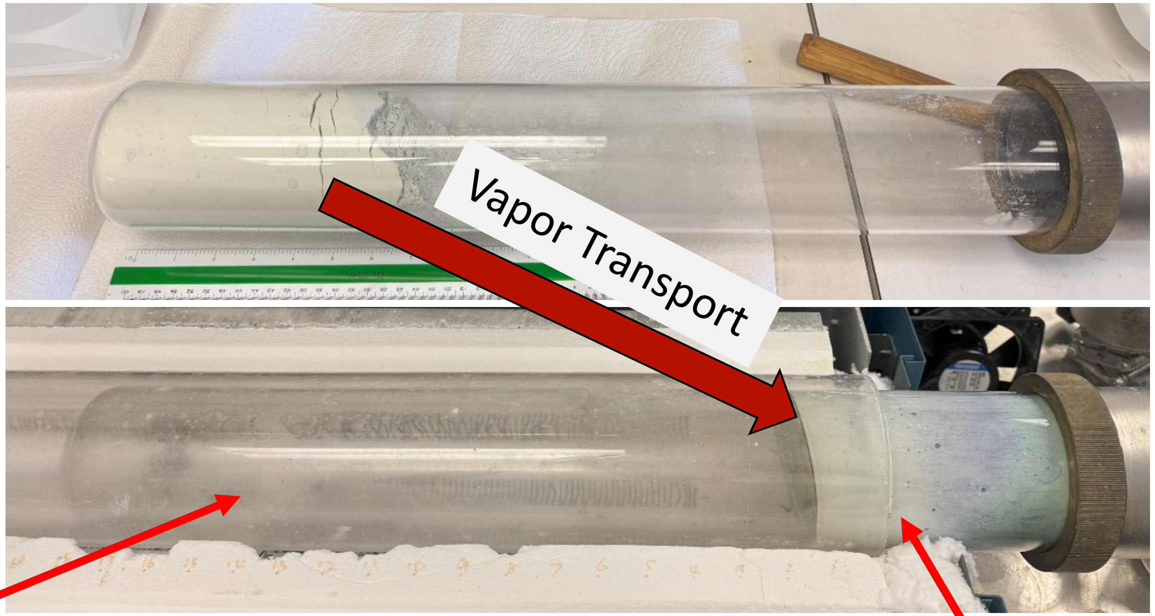




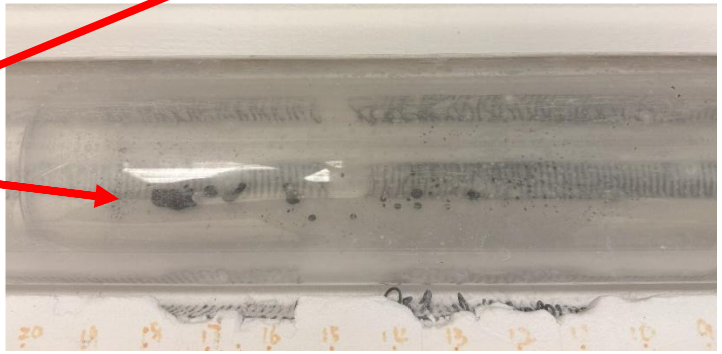
Sublimation of 1500g MoO_3 Powder

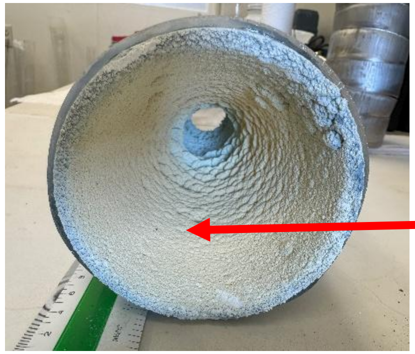
Before Sublimation

After Sublimation



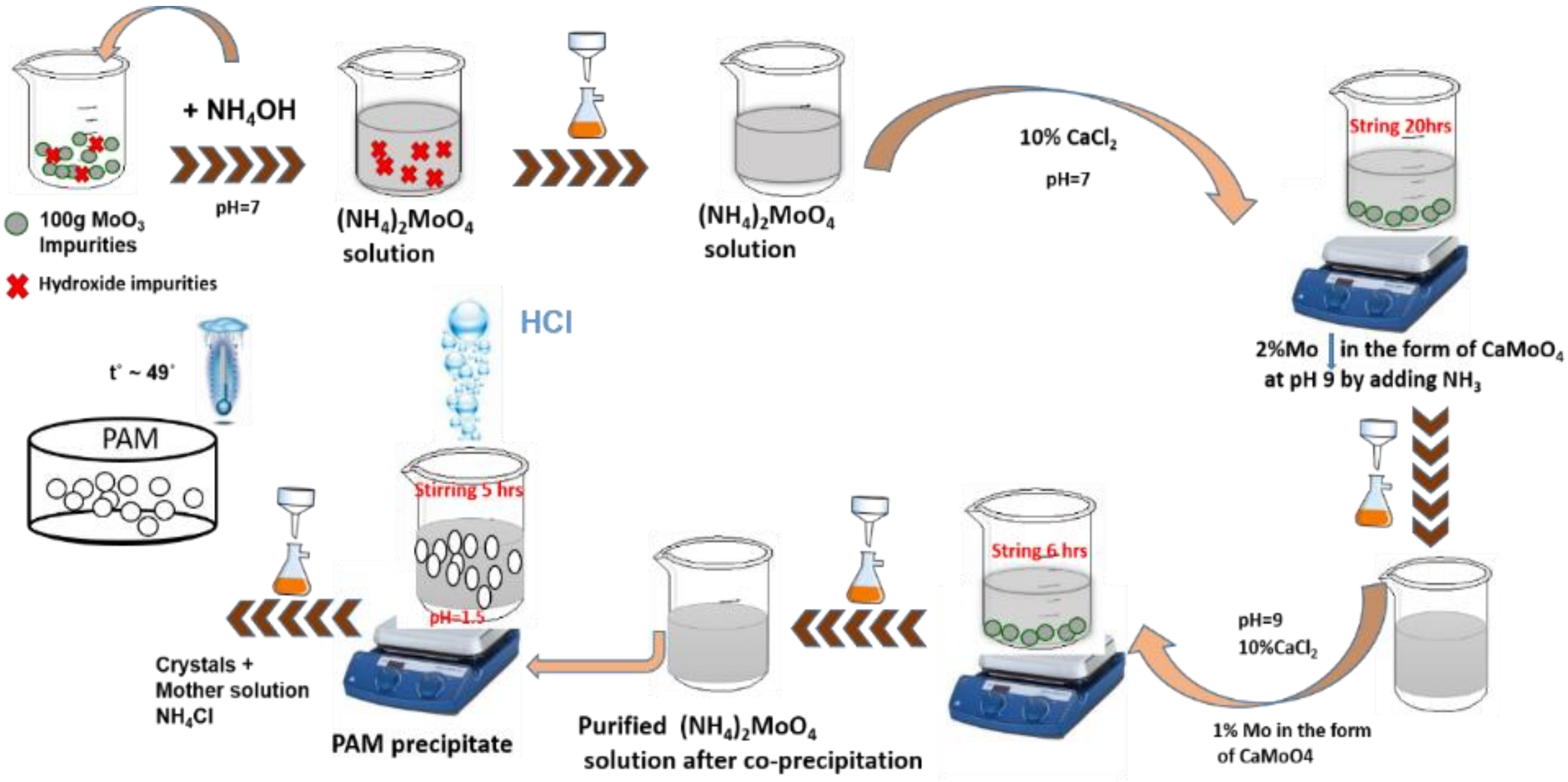
Black Impurity Residuals



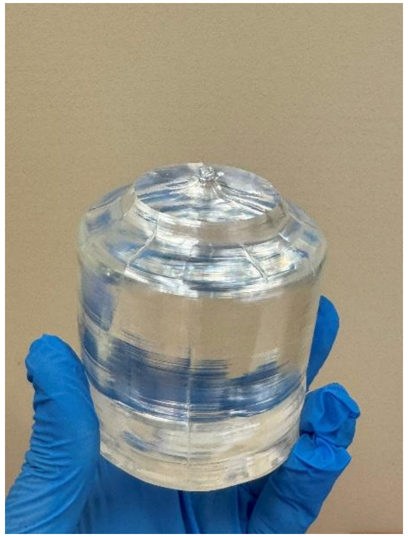


Sublimed MoO_3

Diagram of Co-Precipitation Purification of MoO_3



LMO Grown with Sublimed MoO_3



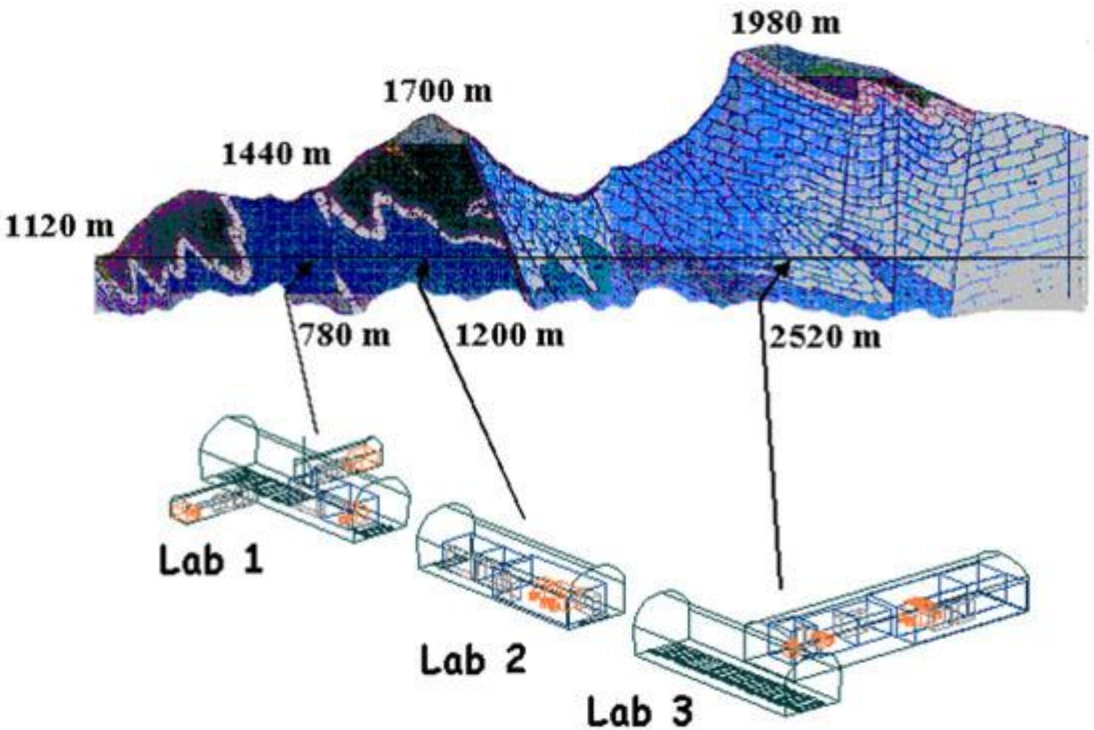
Colorless crystal is very promising for good light output scintillator!

Sublimation Purification of MoO_3

Not purified MoO_3

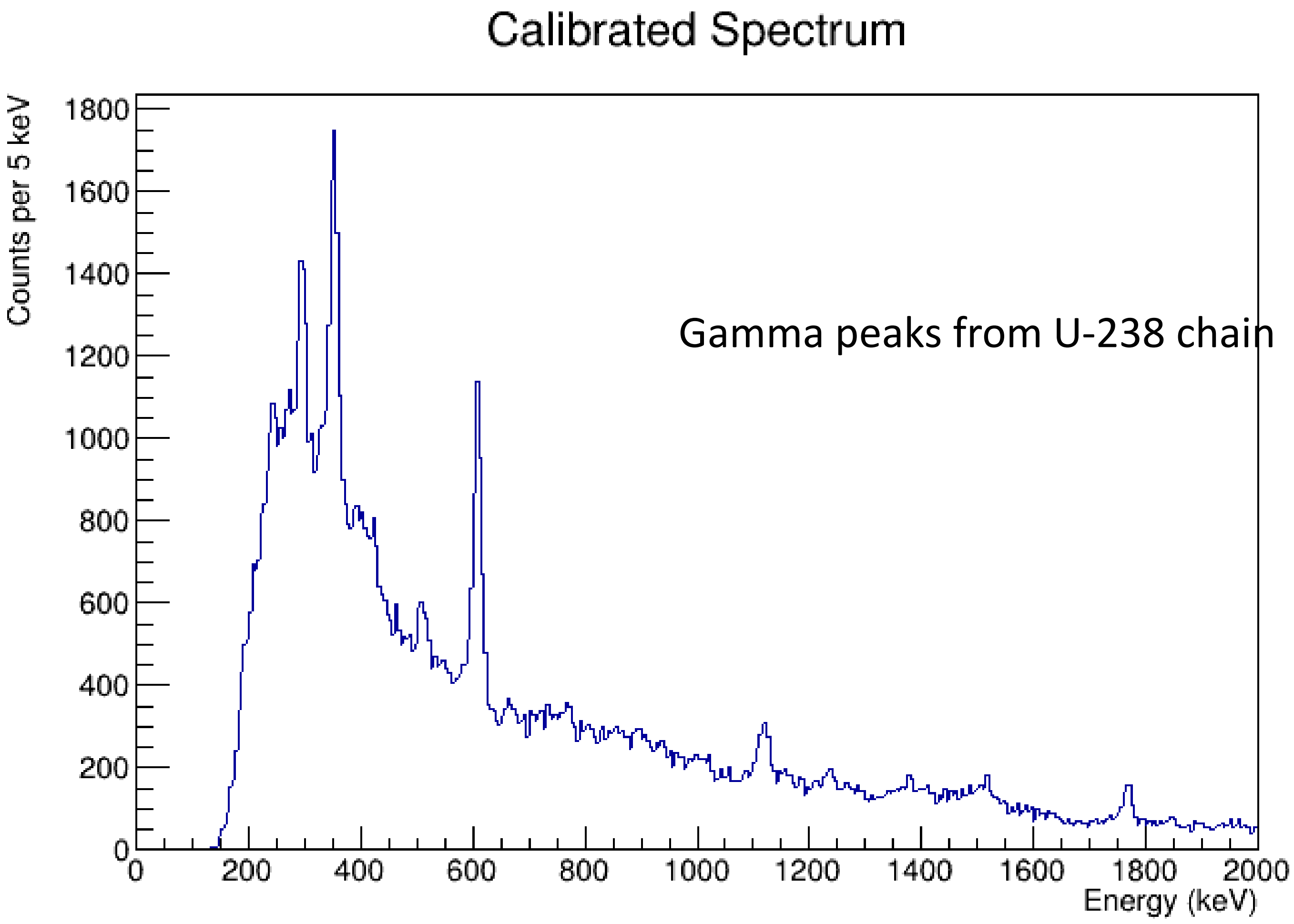
Underground Cryogenic Testing at Canfranc, Spain

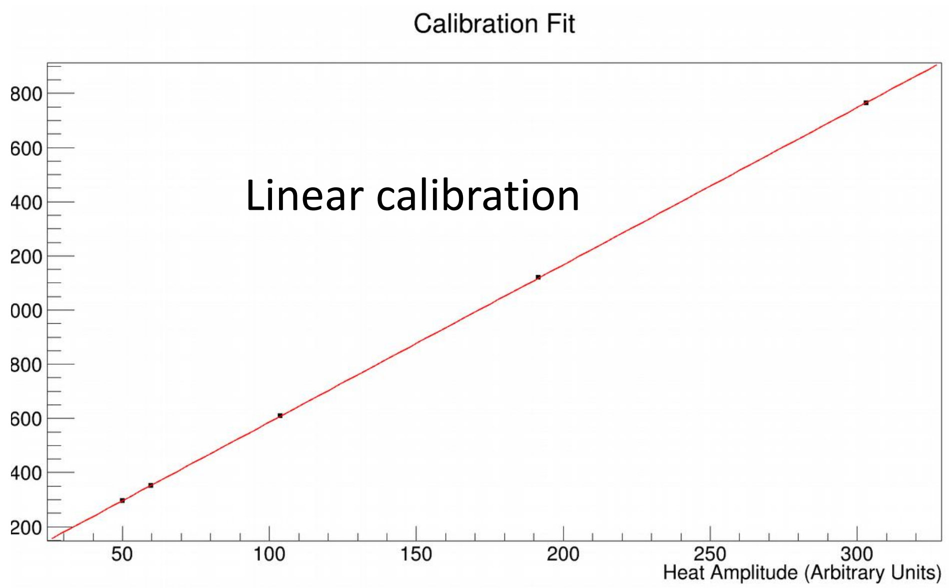
- New colorless LMO crystals to be tested for light output, energy resolution, and radioactivity at Canfranc.
- Previous testing on RMD crystals at Canfranc showed light output below CUPID spec due to color hue in crystals.



Canfranc (Spain)

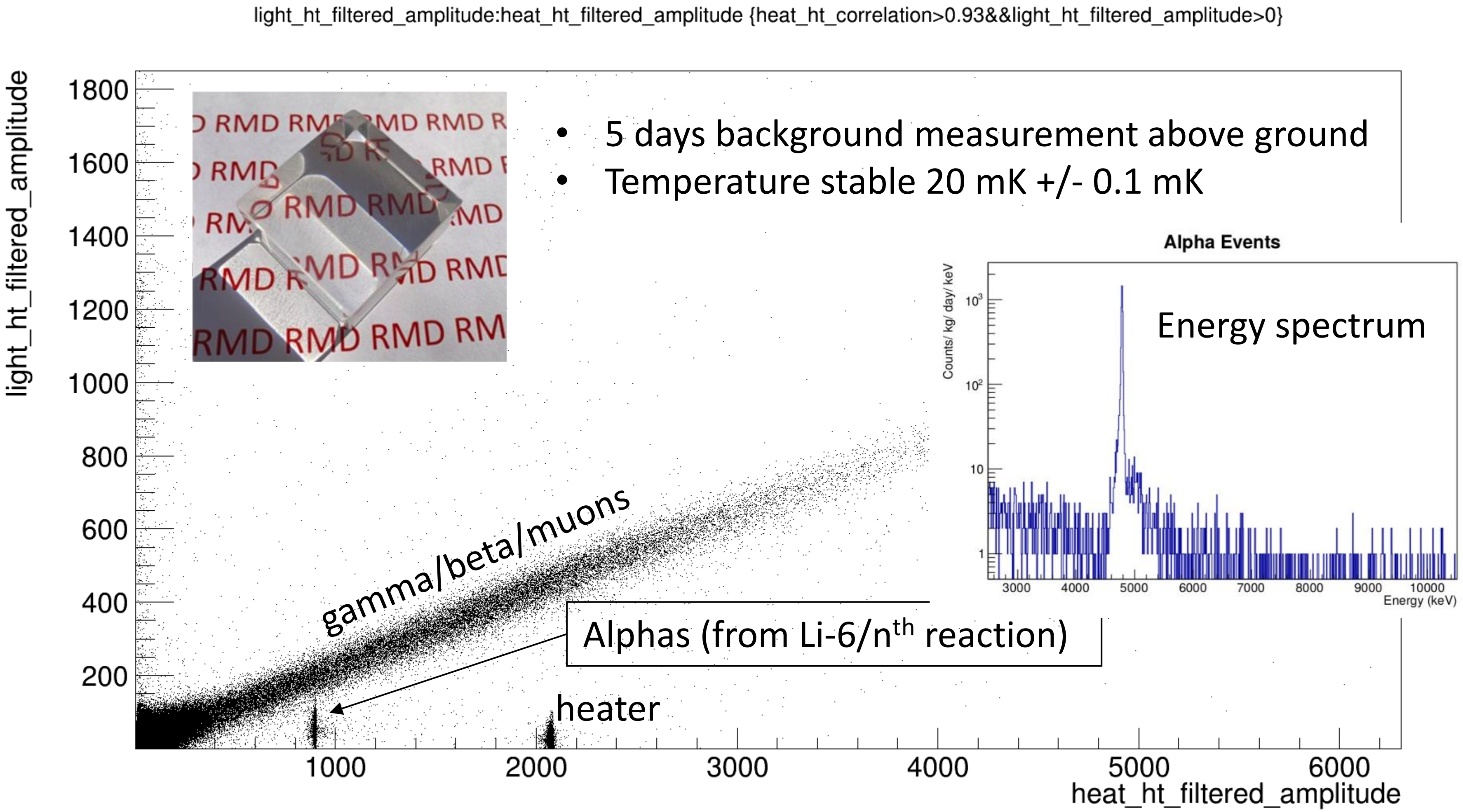
Calibrated Heat Spectrum for LMO
(previous above ground testing in France.)





Light versus Heat Chart for RMD-Grown LMO

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



Good separation of alphas!

Summary / Continuing Work

- ❖ Scale-up of reclamation and purification processes was demonstrated.
- ❖ Colorless LMO crystals were produced using reclaimed/purified MoO_3.
- ❖ All work to-date at RMD used natural Mo isotopes

TBD

- ❖ Use wet chemistry co-precipitation for further purification of MoO_3.
- ❖ Measure impurities in purified MoO_3 and LMO crystals.
- ❖ Testing of LMO cubes at Canfranc underground lab.
  - ❖ Radiation background, light output, energy resolution of heat signal
- ❖ Use enriched Mo-100 if/when the isotope is available.