Ge Crystal pulling, Ge Processing and Recycling of Enriched Material in the US

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*For the **MAJORANA DEMONSTRATOR** Collaboration*

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Ge-76 Detector Production

Isotopic Enrichment
7.8% to 87% ^{76}Ge
Zelenogorsk Electrochemical Plant Zelenogorsk, Russia

Reduce Oxide to Ge Metal
Electrochemical Systems Inc.
Oak Ridge, Tennessee

Zone Refine to 10^{13} cm^{-3}
Electrochemical Systems Inc.
Oak Ridge, Tennessee

Detector Fabrication and Testing
ORTEC, Oak Ridge Tennessee
Delivered to MAJORANA

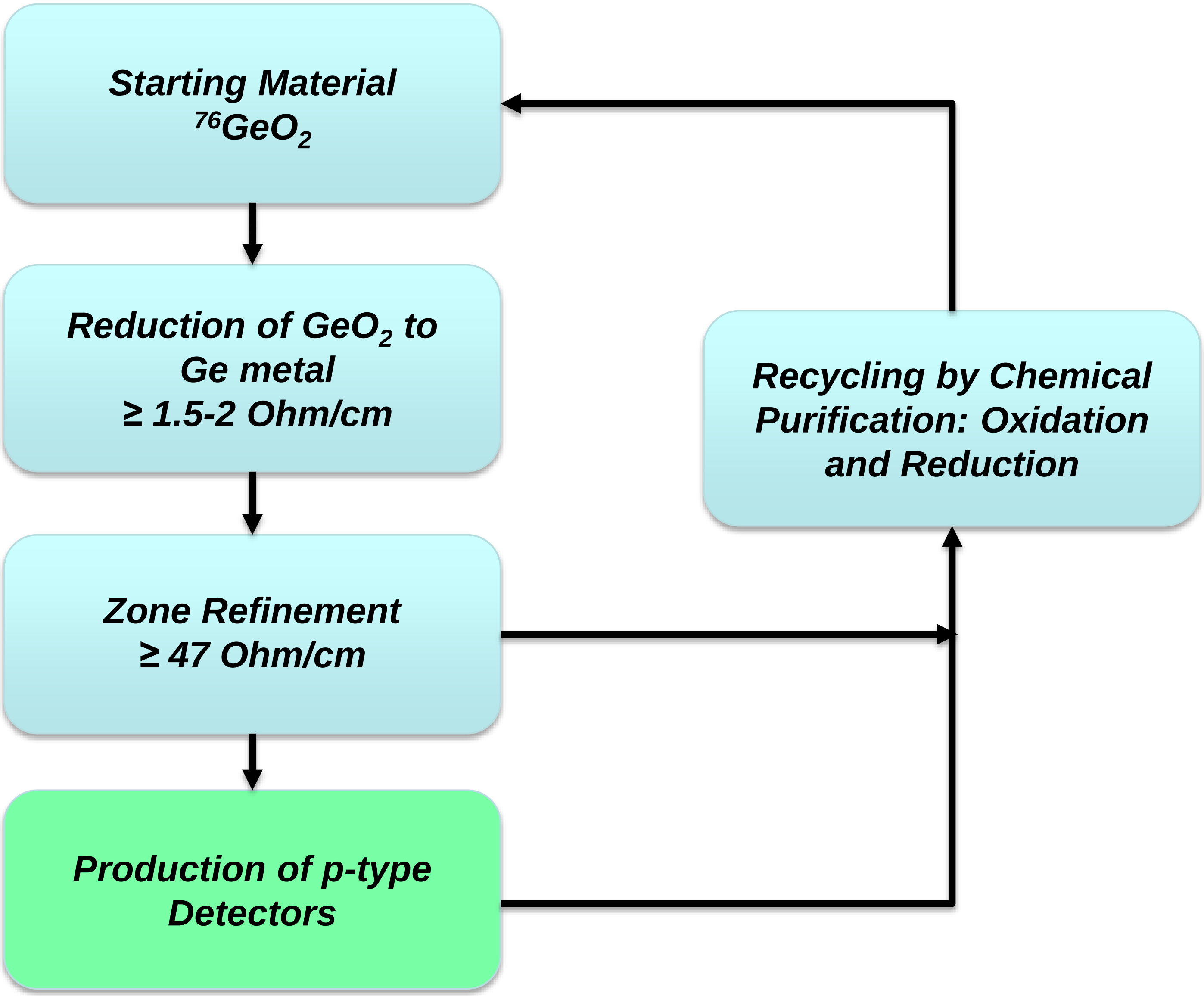
Oxidation to GeO_2
Electrochemical Systems Inc.
Oak Ridge, Tennessee

Chemical Purification through Distillation
Electrochemical Systems Inc.
Oak Ridge, Tennessee

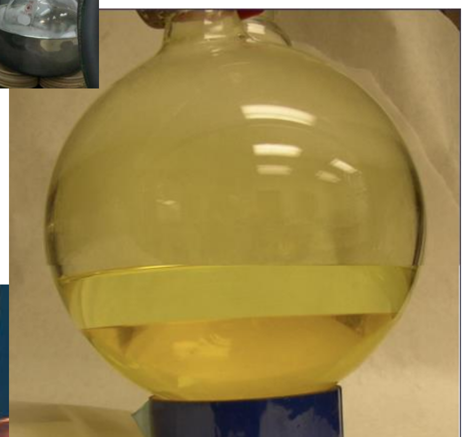
The Challenges and Formation of ESI

- ***There were no qualified commercial companies in the U.S. willing to process enriched Ge.***
- ***Use of a foreign company not practical because of cosmogenics and schedule considerations:***
    - ***a) Each recycling would require an overseas trip.***
    - ***b) Each trip would require the shielded shipping container but would add cosmogenic activation.***
    - ***c) Would require MAJORANA on-site presence.***
    - ***d) Multiple trips would have a serious impact on the schedule.***
- ***The only practical solution was to create an organization within the Collaboration for this task.***
- ***Contract with a private company (Electrochemical systems Inc.) to equip, operate and manage a Collaboration-controlled facility to reduce/purify zone refine and recycle the ^{76}Ge***
- ***A Task team of ten scientists was formed and augmented by two technical experts with industrial experience.***
- ***A procedures manual was written by the technical experts and reviewed by QA team and updated recently.***
- ***The required equipment was purchased and installed at Electrochemical Systems.***

Ge-76 Processing and Recycling



Recycling by Chemical Purification

Ge Recovered from Detector Manufacturing Process

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Chlorination to $GeCl_4$ and Purification through Fractional Distillation

↓

Hydrolyze $GeCl_4$

↓

Reduce GeO_2 to Ge metal

↓

Zone Refine and Test for Detector grade material

MJD ^{enr}Ge Detector Production

Phase-I (summer 2012): Trial run

[2 ^{nat}Ge detectors]

- ***~9 kg of ESI zone-refined ^{nat}Ge were delivered to ORTEC***
- ***One crystal was pulled (enough material to pull 2 crystals)***
- ***Two 1-kg-class PPC detectors were made from the crystal***
- ***MJD observer present to observe the whole process. Monitored dust count, Rn level, and materials in contact with Ge.***
- ***MJD and ORTEC further refined cleanliness, enriched material control and delivery, and QA plans collaboratively***

MJD ^{enr}Ge Detector Production

Phase-II (Nov. 2012 – Mar. 2014): ^{enr}Ge material [30 ^{enr}Ge detectors]

- ***Processed 41.78 kg of detector grade material from 42.5-kg of ^{enr}Ge (61.7-kg of GeO2) for a initial processing yield of 98.3%***
- ***Thirty ^{enr}Ge detectors: 25.24 kg (total mass)***
- ***For each delivery of 9 kg of materials: two crystals could be pulled, and up to 4 detectors could be fabricated. Total duration: ~2 weeks per delivery.***
- ***Unused ^{enr}Ge materials from each run returned to production stream in latter runs.***
- ***^{enr}Ge stored in a nearby cavern when not being used***
- ***Two levels of detector testing at ORTEC:***
  - ***Normal vendor QA/QC for their Ge detectors (analog)***
  - ***MJD set up a test stand at ORTEC to take digitized data for preliminary QA on pulse-shape-analysis performance***

MJD ^{enr}Ge Detector Production

Phase-III (Jan. 2015 – Jun. 2015): Reprocessed ^{enr}Ge material [5 ^{enr}Ge detectors]

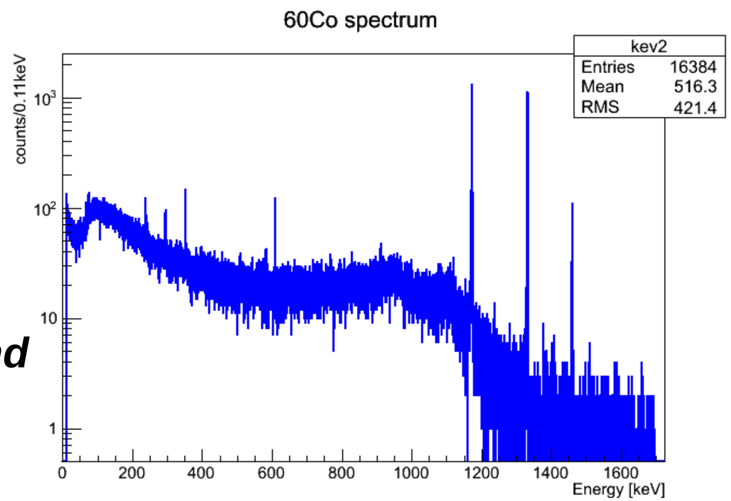
- *Previously used, but no longer acceptable ^{enr}Ge material (metal pieces and of a sludge containing shavings, grindings and machining scraps) reprocessed for use in detector production by ESI.*
- *Input material through reprocessing:*
  - ✓ *8.4-kg of starting "scrap" from Phase-II*
    - ✓ *2.87 kg of metal from detector manufacturer reject.*
    - ✓ *5.53 kg metal in sludge*
  - ✓ *5.87 kg of ^{enr}Ge with a ≥47 Ohm-cm recovered through reprocessing*
  - ✓ *Combined with 3.22 kg of unused Phase-II material to yield 9.1 kg of detector grade material*
  - *Produced two 4.5 kg boules*
- ***Five additional ^{enr}Ge detectors (4.44 kg) to bring the total detector mass to 29.68 kg.***

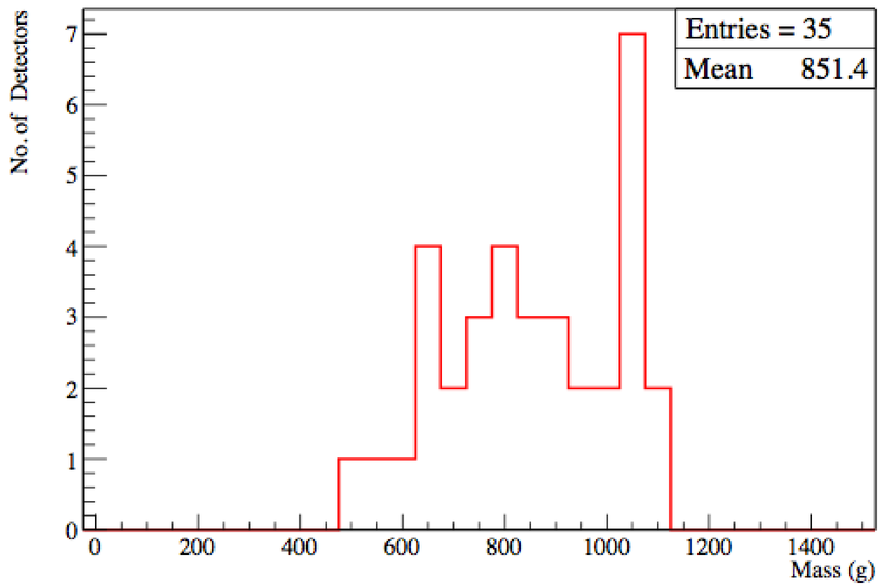
MJD Enriched Detectors

- ***Final yield of detectors: 70% (29.68 kg detectors from 42.5 kg starting ^{enr}Ge)***
  - ***Best to date for overall yield Ge experiments***
  - ***Unused ^{enr}Ge inventory: 1.49 kg (crystal) and 1.15 kg (zone refined)***

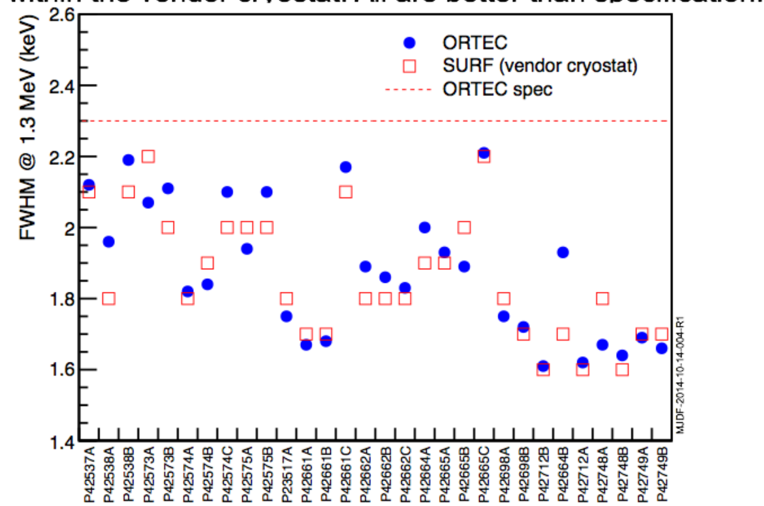
- ***MJD Processing facility decommissioned at the end of April 2015 and relocated to SDSMT.***

Enriched ORTEC PPC UG (60Co Calibration)



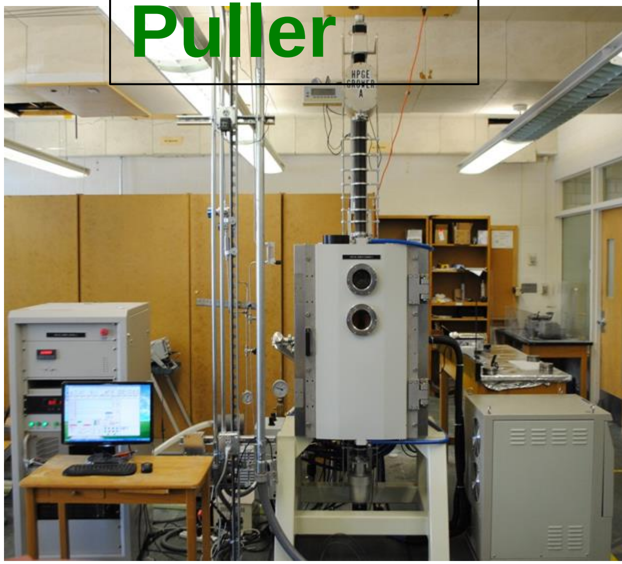


Comparison of measurements done at ORTEC and SURF within the vendor cryostat. All are better than specification.



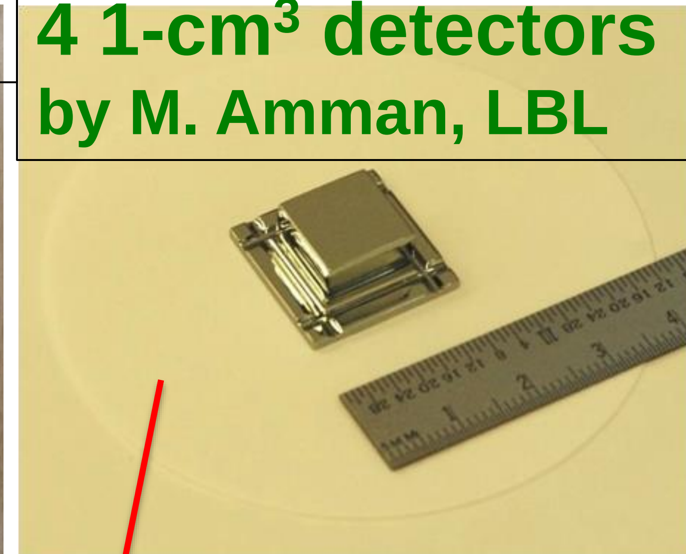
He-Ge Crystal Growth at USD

Puller



3 kg crystal

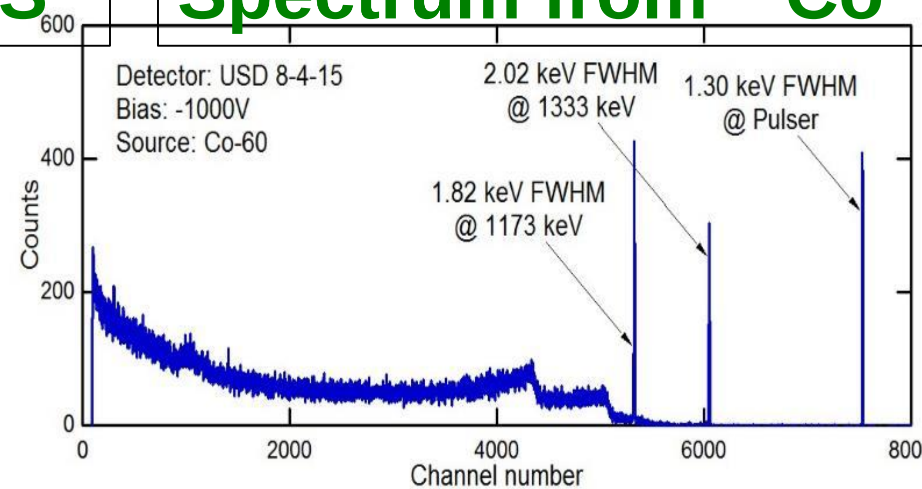
4 1-cm^3 detectors by M. Amman, LBL



2.3 kg crystal

SuperCDMS

Spectrum from ^{60}Co

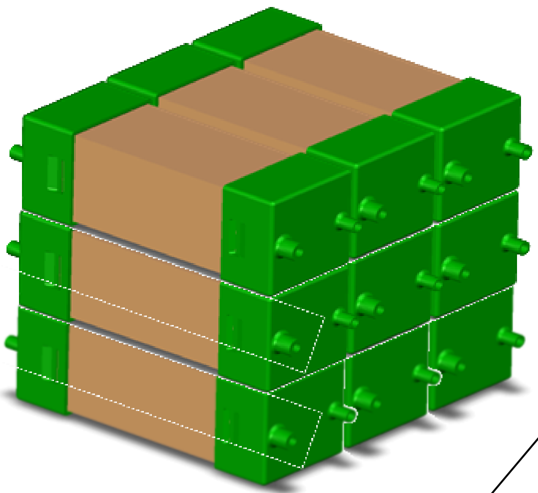


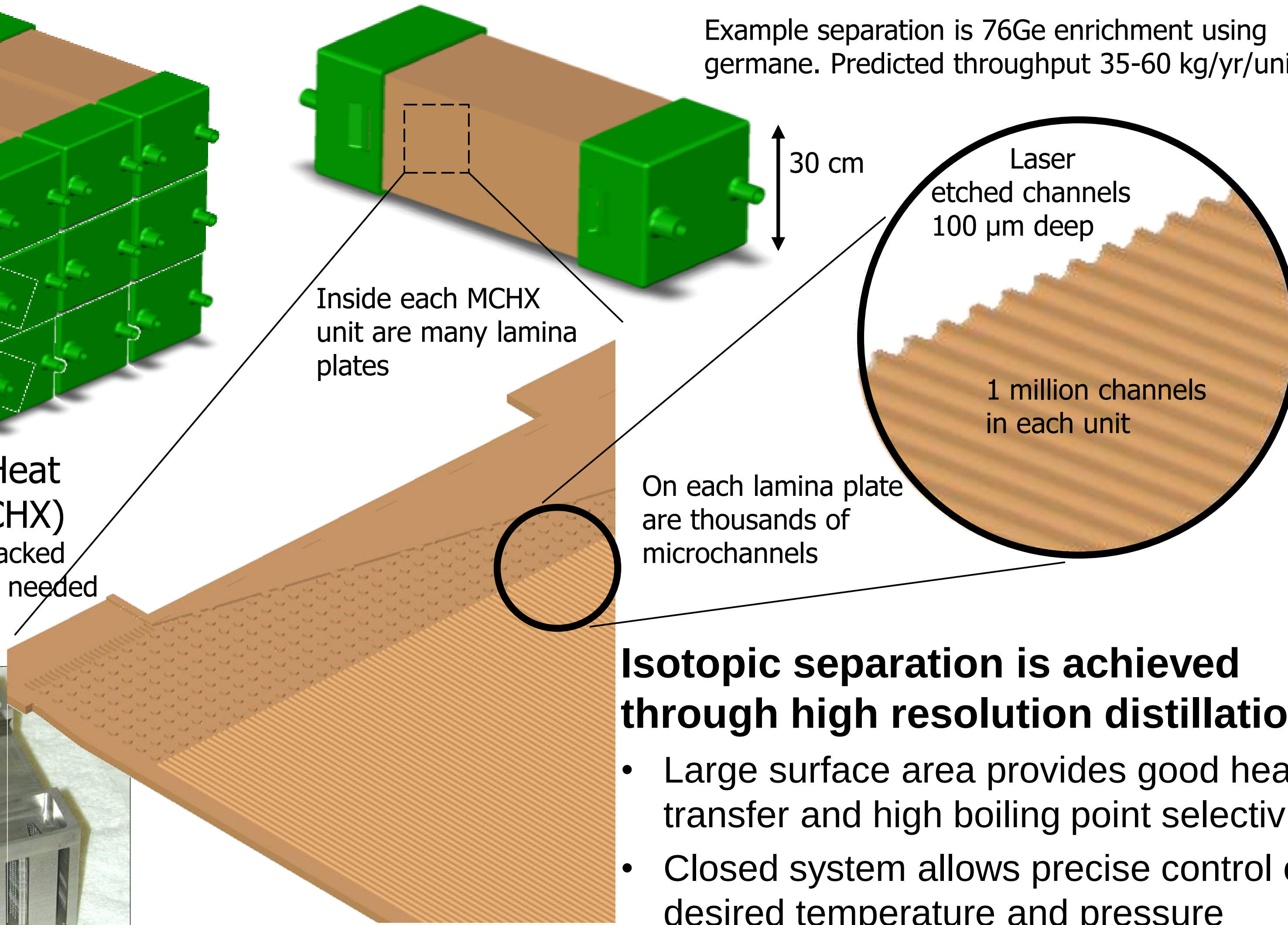
U.S. ^{76}Ge Future Capabilities

- ***Crystal Growth***
- ***Detector Fabrication and Recycling***
- ***Enrichment of Ge***

PNNL Isotopic Separation from High Resolution Distillation

Example separation is 76Ge enrichment using germane. Predicted throughput 35-60 kg/yr/unit





Microchannel Heat Exchanger (MCHX)

Scalable-Multiple stacked units can be used if needed

Isotopic separation is achieved through high resolution distillation

- Large surface area provides good heat transfer and high boiling point selectivity
- Closed system allows precise control of desired temperature and pressure
- Large number of channels provides high throughput

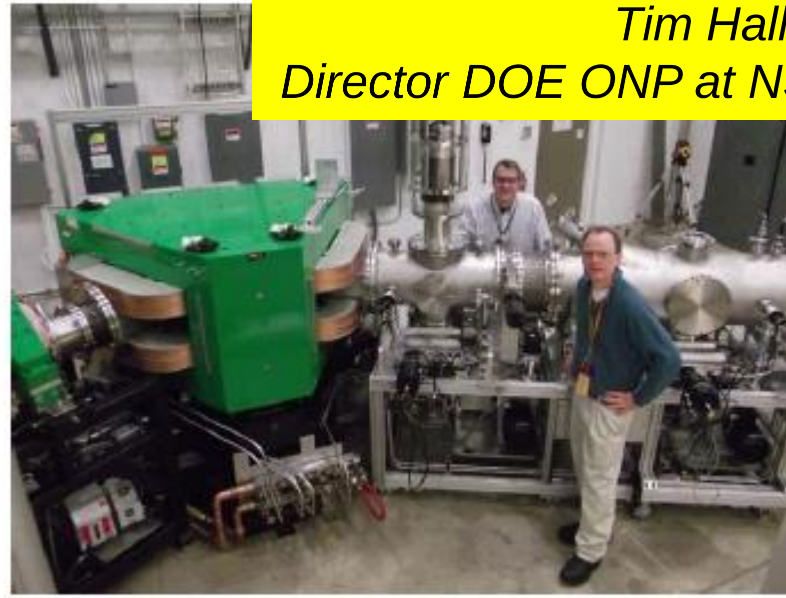
MCHX is a proven PNNL technology, actual device shown above

Small and Mid-Scale Projects: Stable Isotope Production Facility (SIPF)

Tim Hallman
Director DOE ONP at NSAC

- The Request initiates the SIPF MIE, which directly supports the DOE Isotope Program mission, restoring domestic capability that has been lacking since 1998.
  - Renewed enrichment capability will benefit nuclear and physical sciences, industrial manufacturing, homeland security, and medicine.
  - Nurtures U.S. expertise in centrifuge technology and isotope enrichment that could be useful for a variety of peaceful-use activities.
  - Addresses U.S. demands for high priority isotopes needed for suite of activities: neutrinoless double beta decay, dark matter experiments, target material for Mo-99 production.
  - Help mitigate U.S. foreign dependence on stable isotope enrichment.

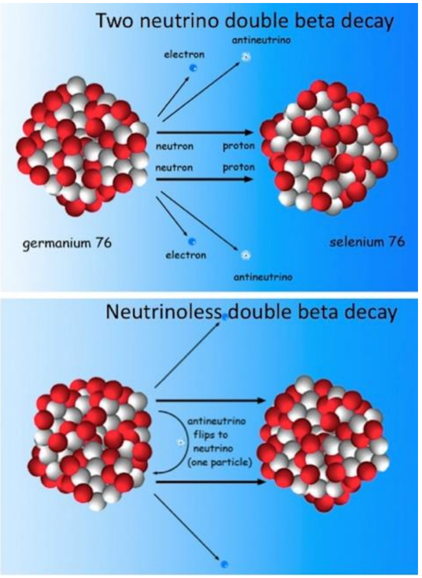
FY 2017 Request: $2,500,000
Estimated Total Project Cost: $9.5M-$10.5M
Estimated time frame for completion: FY 2020



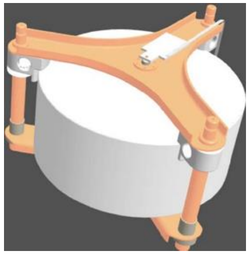
SIPF responds to Nuclear Science Advisory Committee – Isotopes (NSACI):

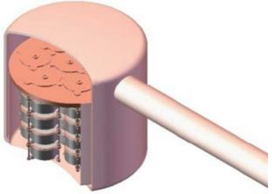
- 2009 Recommendation: "Construct and operate an electromagnetic isotope separator facility for stable and long-lived radioactive isotopes."
- 2015 Long Range Plan: "We recommend completion and the establishment of effective, full intensity operations of the stable isotope separation capability at ORNL."

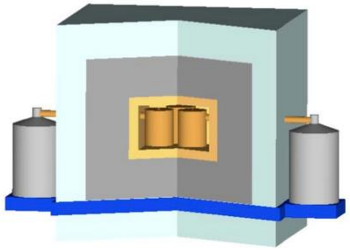
Large-Scale Enrichment Possibilities



- Support Large Scale, Fundamental, R&D Projects for example in Neutrinoless Double-Beta Decay [1]
  - ^{76}Ge (MAJORANA / US, GERDA / Italy)
  - ^{136}Xe (EXO-200 / US, NEXT / Spain, KamLAND-Zen / Japan)
  - ^{100}Mo (SuperNEMO / France) ; multi-isotope incl. ^{100}Mo
- May also support emerging, large scale medical/commercial applications

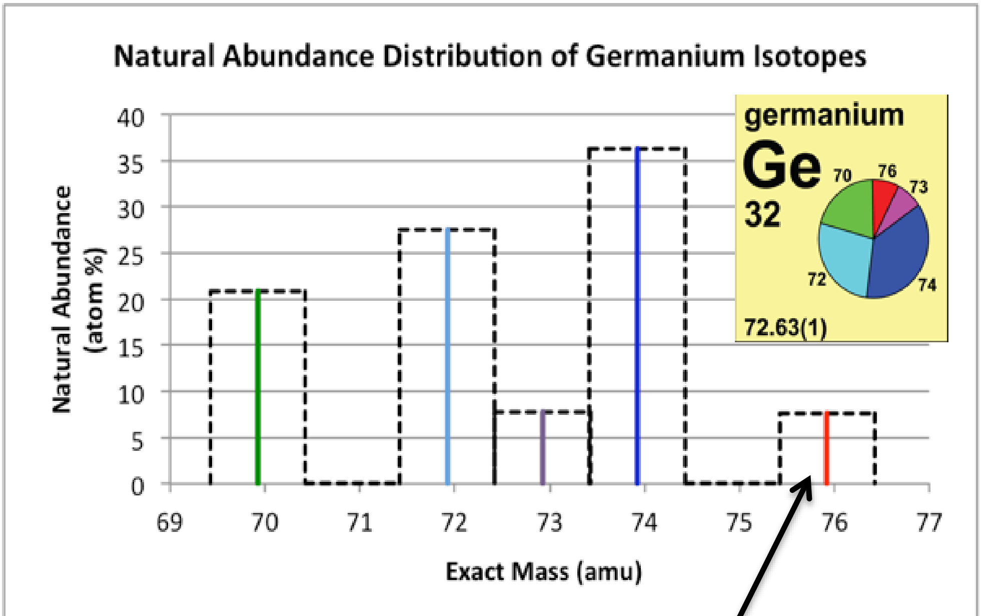






1: "Neutrinoless Double Beta Decay", Report to the Nuclear Science Advisory Committee, April 24, 2014
KEY: GCIS = Gas Centrifuge Isotope Separator

Germanium Stable Isotope System



	Exact Mass (amu)	Natural Abundance (atom %)
	69.9242	20.85
	71.9221	27.54
	72.9235	7.735
	73.9212	36.29
	75.9214	7.614

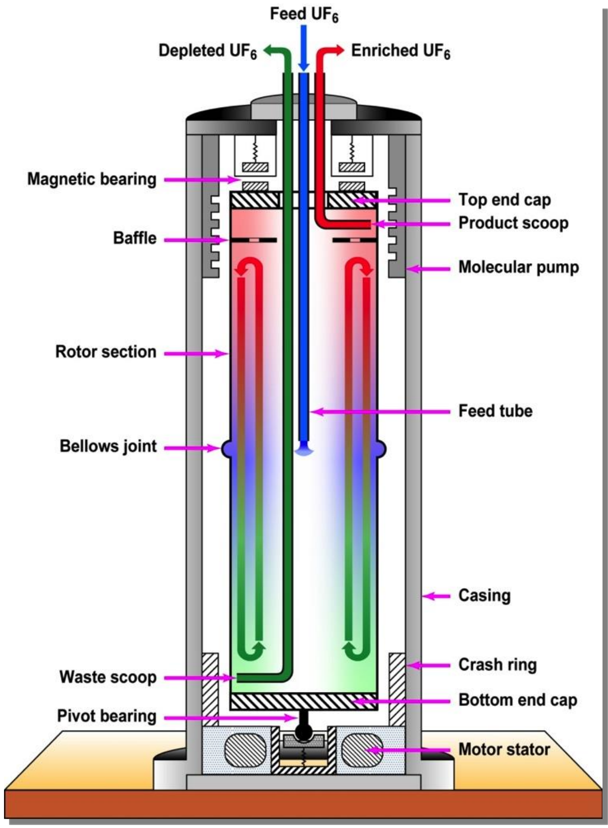
Note: dashed lines represent an arbitrary +/- 0.5 amu band for each isotope exact mass

Large scale separation

- Some R&D going on in micro-channel distillation (lower TRL)
- EMIS cannot provide the required quantity
- Proven technology: centrifuges (current supply from Russia)

^{76}Ge Feasibility Study

- The study was funded by the DOE Isotope Program at ORNL
- GCIS simulation / modeling study of ^{76}Ge separation
  - Newly developed and tested US stable isotope machine (SI-1) is part of the Enriched Stable Isotope Pilot Plant (ESIPP) for 10-100 gram/year stable production
  - FY17 President's Budget Request includes an expansion of ESIPP to kilogram production per year.
- Feasibility of 200 kg/year with caveats
  - Technical feasibility is not an issue (we know how to do this and performance maps on SI-1 demonstrate a machine that performs as designed)
  - Cost and facility drivers could be issues
  - SI-2 machine (3x taller) required for this scale of separation although enough SI-1 machines could do the same job
    - Feed material for total ^{76}Ge(metal @ 85%) required - assumed to be 1,500 kg
    - Facility built to house separators (ca 4 cascades of 200 units each)
    - Operations for 5-7 years (assumes 200 kg/year production)
  - Cost is close to Russian purchase of material, but with enduring facility



Summary

- *Processing and Recycling for tonne scale will be necessary for high yield in detector production. The MJD / ESI method is scalable.*

- *ORTEC worked with MJD to fine tune the detector fabrication yielding high purity crystals.*

- *Universities are developing crystal growth capacities.*

- *Stable isotopes program in the U.S. is being revived*
  - *- initially a small scale pilot plant being funded and constructed*
  - *- possible interest in larger scale expansion for bb-decay*
  - *- ORNL feasibility study for 76Ge bb-decay*

- *Isotope enrichment (^{76}Ge, ^{82}Se, ^{136}Xe) requires time and $s.*

  Preference of DOE Office of Nuclear Physics to, if possible, do some enrichment within the U.S., new possibilities are being explored.

- *Recent ORNL isotopes study shows that including investment in sufficiently sized enrichment infrastructure, the price is comparable to cost to buy from Russia (using MJD 76Ge isotope cost).*