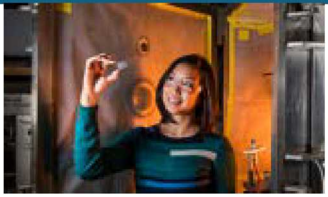
Sandia National Laboratories

SAND2018-9092C

Development of ERD Technique for Quantifying Light Isotope Concentrations in Irradiated TPBAR Materials



PRESENTED BY

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Sandia National Laboratories, Albuquerque, NM

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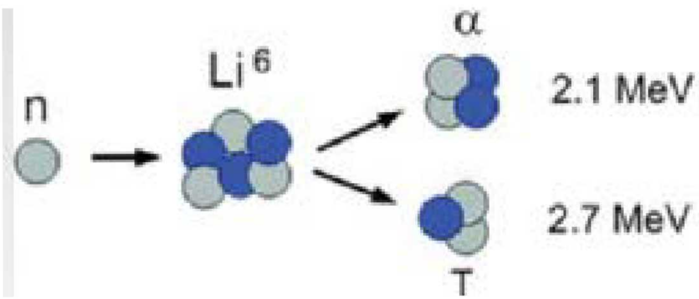
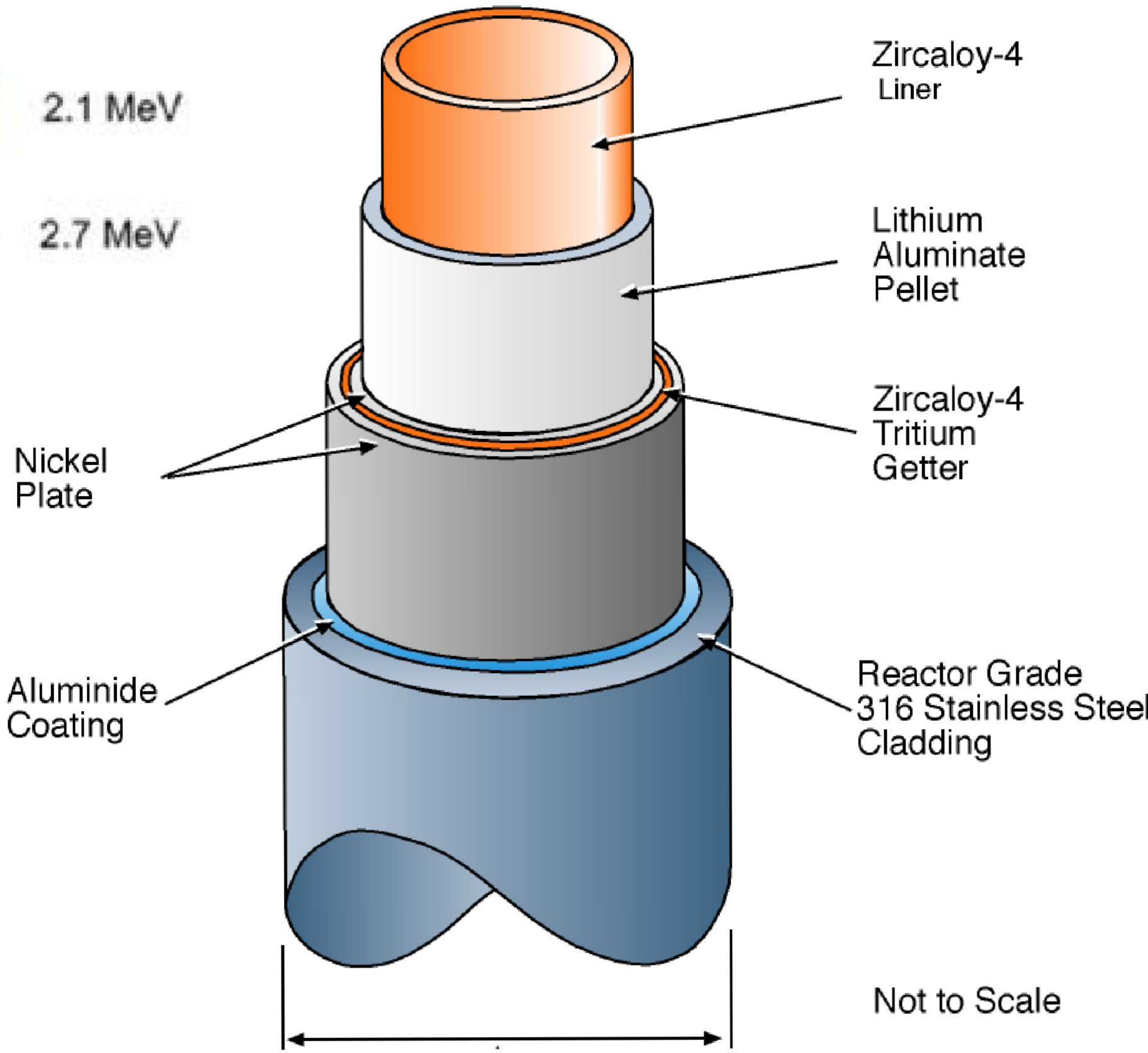
Tritium Producing Burnable Absorber Rod

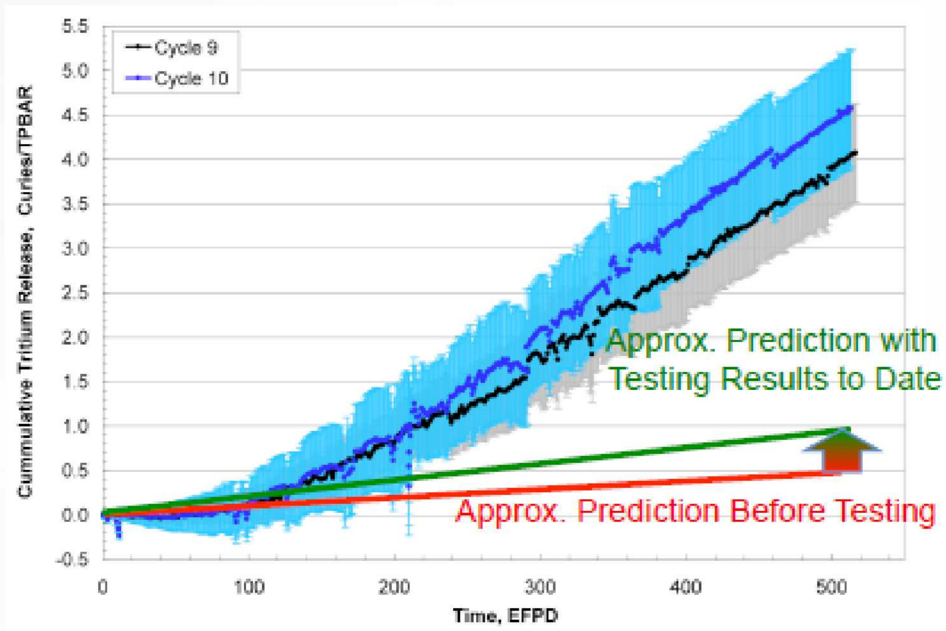


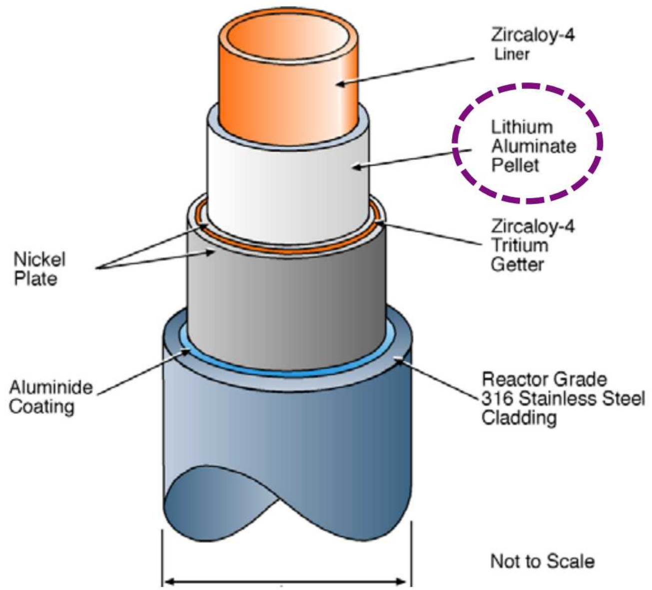
- Displacement Damage
- Helium Implantation
- Tritium Implantation
- Elevated Temperatures



Understanding Tritium Permeation in TPBAR

- TPBAR ^{3}H permeation is higher than predictive performance models
  - In 2004, during Cycle 6, the predicted levels were ~0.5 Ci/TPBAR/cycle and actual levels were ~4 Ci/TPBAR/cycle (0.04% of total ^{3}H produced)
- Mechanisms responsible for differences between predictions and observations are not well understood
- Isotopic concentrations in the $LiAlO_2$ pellet after ^{3}H extraction are currently unknown.

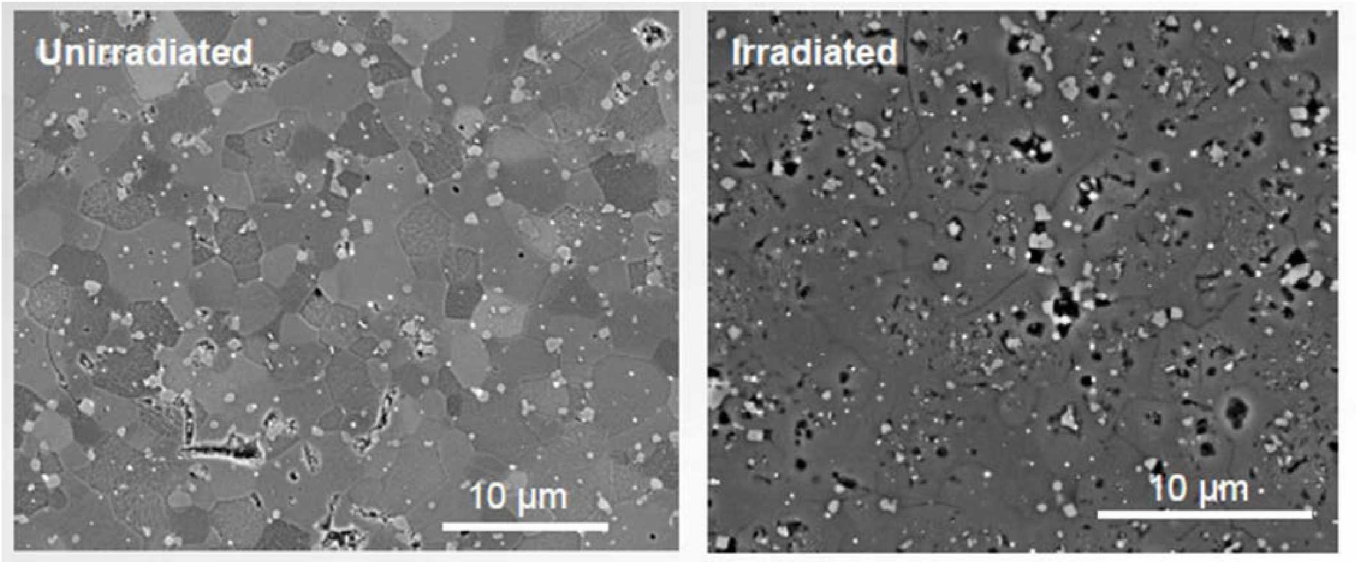




Burkes, Senor, Longoni and Johns, TFG Meeting 2016, Rochester, NY

Isotopic concentrations in the $LiAlO_2$ pellet after ^3H extraction are currently unknown

- Neutronics calculations only provide an estimate of the ^6Li burnup
- Some ^3H may remain inside the sample, trapped in He bubbles or other defects
- Complex microstructures have been found in irradiated $LiAlO_2$ pellets



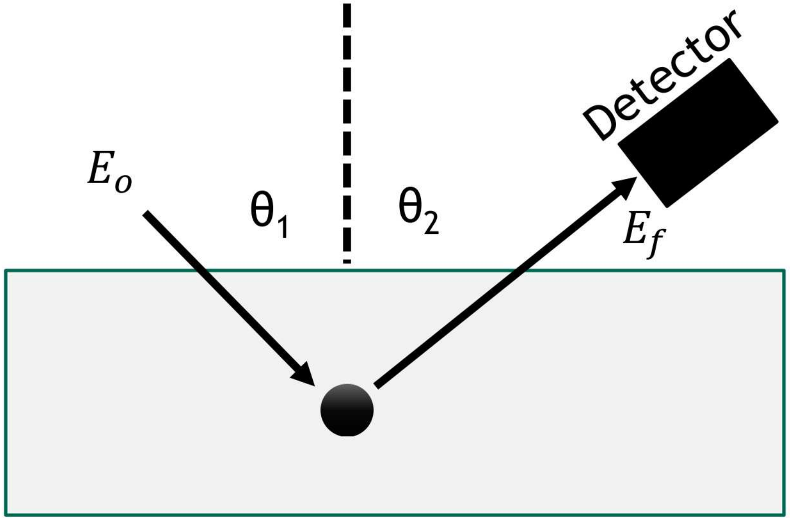
- Dissolution of $LiAlO_2$ on GBs
- Voids form inside grains
- $LiAl_5O_8$ form inside grains

- Li diffuses to GB
- Al, O, and vacancies diffuse to the center of the grains.

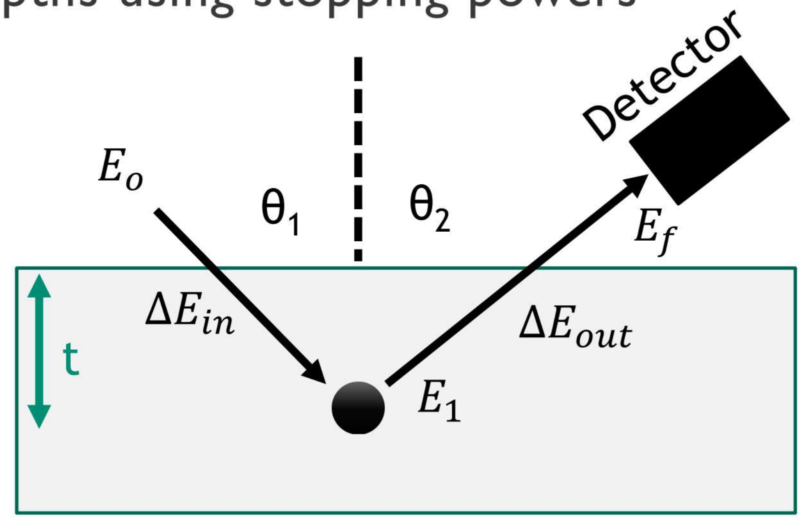
Burkes, Longoni, Hu, Whitman, Palmer, Greaney, Senor, Tritium Focus Group Meeting 2018, Oak Ridge, TN

Elastic Recoil Detection (ERD) analysis is well suited for determining isotopic concentrations as a function of depth

- Standard-less method of chemical analysis with depth profiling. Depth and composition resolutions are dependent on sample and experimental parameters specific to the system.
- Heavy MeV range ion impinges the target, forward scattering target atoms into the detector.



Incoming ion and projectile energies can be estimated at various depths using stopping powers



$$\Delta E_{in} = \int_0^{t/\cos\theta_1} \left(\frac{dE}{dx}\right)_{incoming\ ion} dx$$

$$E_1 = K(E_o - \Delta E_{in})$$

$$\Delta E_{out} = \int_0^{t/\cos\theta_2} \left(\frac{dE}{dx}\right)_{recoil} dx$$

28 MeV Si → $LiAlO_2$

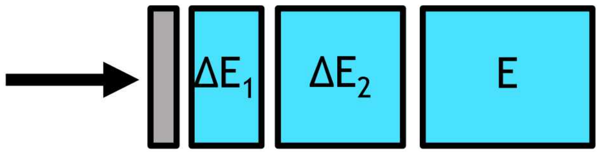
Bragg Correction = 0.00%
Stopping Units = eV / Angstrom
See bottom of Table for other Stopping units

Ion Energy	dE/dx Elec.	dE/dx Nuclear	Projected Range	Longitudinal Straggling	Lateral Straggling
20.00 MeV	4.192E+02	8.503E-01	6.53 um	2445 A	2897 A
22.50 MeV	4.179E+02	7.698E-01	7.12 um	2600 A	2953 A
25.00 MeV	4.155E+02	7.042E-01	7.72 um	2747 A	3005 A
27.50 MeV	4.122E+02	6.494E-01	8.32 um	2887 A	3056 A
28.00 MeV	4.115E+02	6.396E-01	8.45 um	2895 A	3066 A

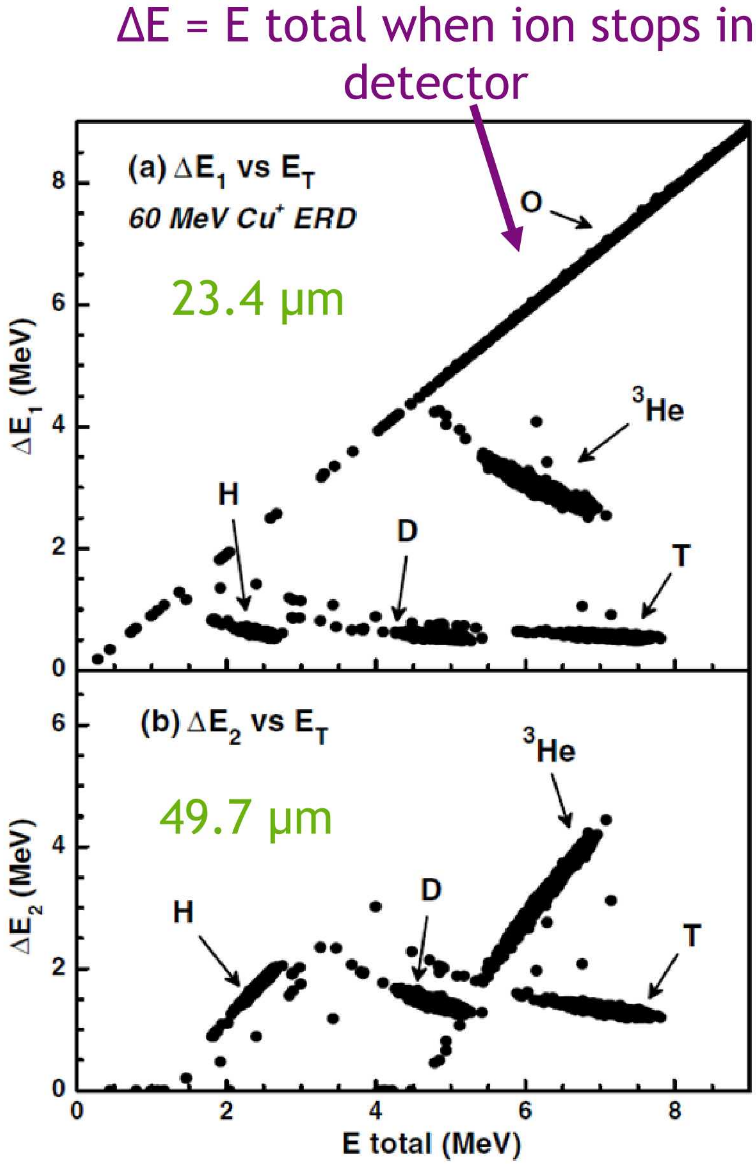
By dividing the sample into *n* layers called “slabs,” an iterative approach can be used to calculate the ERD yield

The HI-ERD system at Sandia was originally designed to quantify ^1H, ^2H, ^3H, and ^3He in aged metal tritides

- A Monte Carlo code called MCERD was utilized to simulate ERD experiments in potential system designs.
- In the original design, a solid state ΔE_1-ΔE_2 - E detector was planned.
  - The thickness of the ranging foil and both ΔE detectors were chosen in order to maximize separation between recoiling ^{3}He, H, D, T, and O using coincidence techniques.



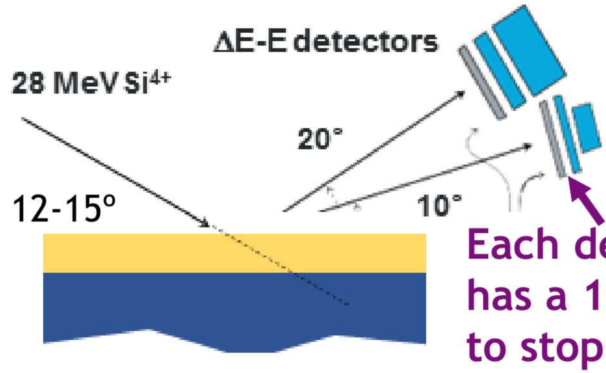
- The system design was simulated using an incoming beam of 60 MeV Cu.
- Simulations showed O stopping in the ΔE_1 detector, and H and ^{3}He stopping in the ΔE_2 detector.



Knapp et al. NIMB 219-220 (2004)

The HI-ERD system was eventually built using two particle telescope detectors at angles optimized for a 28 MeV Si beam.

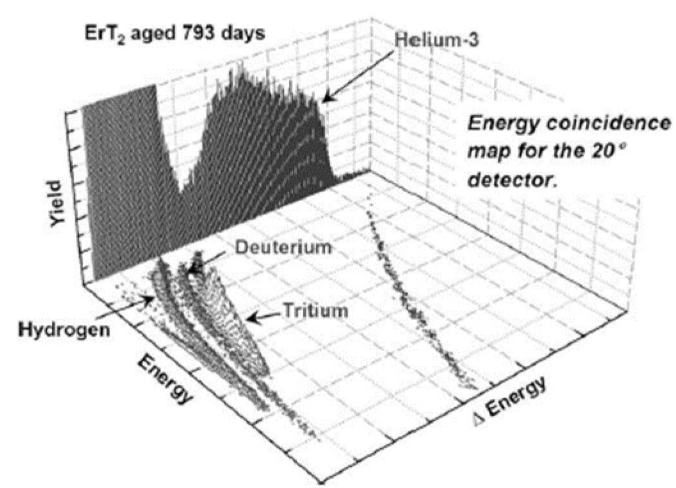
- One particle telescope (44 µm ΔE) was at a detection angle optimized for ^{3}He, H, D, and T, and the other telescope (8 µm ΔE) was at a shallower detection angle optimized for O and C.



Each detector has a 15 µm foil to stop Si ions

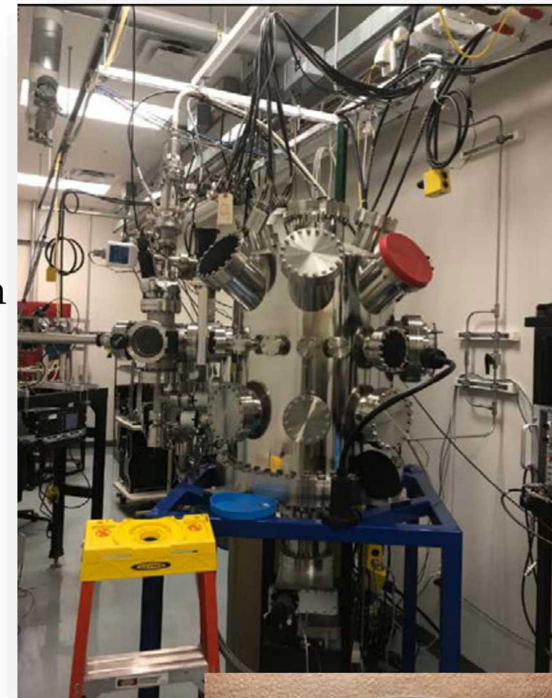
- Each particle telescope consists of a pair of detectors.
  - The first detector, the ΔE detector, is thin and most scattered particles scatter through
  - The particles are stopped in the second detector, the E detector
- The 15 µm foils in front of each telescope stop the directly scattered Si ions
- These experiments were found to have a depth resolution of ~40 nm and sensitivity of ~1 at.%.

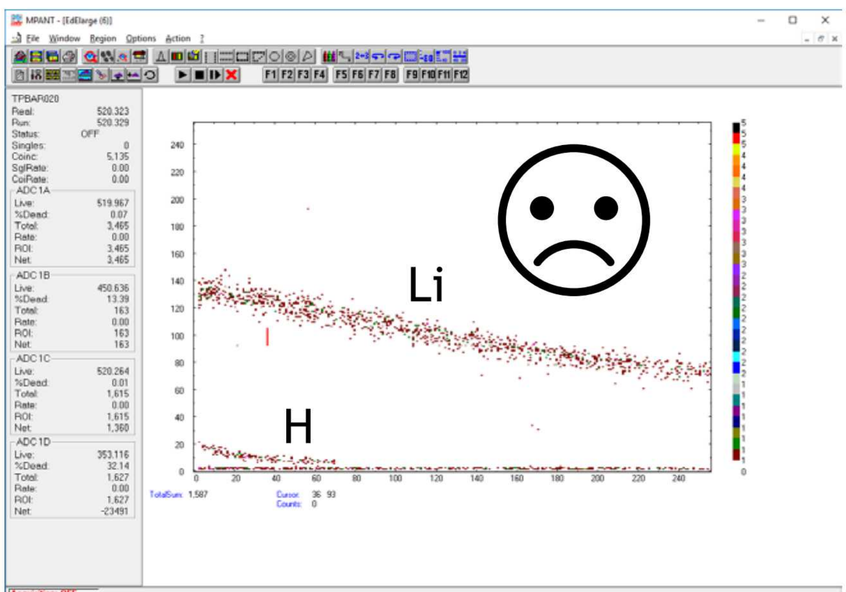
Energy coincidence map for the 20° detector with a 44 µm thin ΔE detector



For this project, the HI-ERD system was optimized for separation of ^{6}Li, ^{7}Li, ^{3}He, ^{4}He and hydrogen isotopes in $LiAlO_2$

- Parameters were optimized by trial and error using $LiNbO_3$ samples with a natural abundance of ^{6}Li, and on ^{6}Li enriched $LiAlO_2$ pellets with an isotopic fraction of 0.239 mol $^{6}Li/Li$ total.
- $LiAlO_2$ pellets were cross-sectioned, mounted in epoxy and polished, an engineered control to reduce tritium contamination from radioactive pellets from the reactor.
- First, the original HI-ERD configuration using 28 MeV Si to scatter isotopes into both particle telescopes was utilized, but detector mass resolution was not good enough to separate ^{6}Li and ^{7}Li signals.

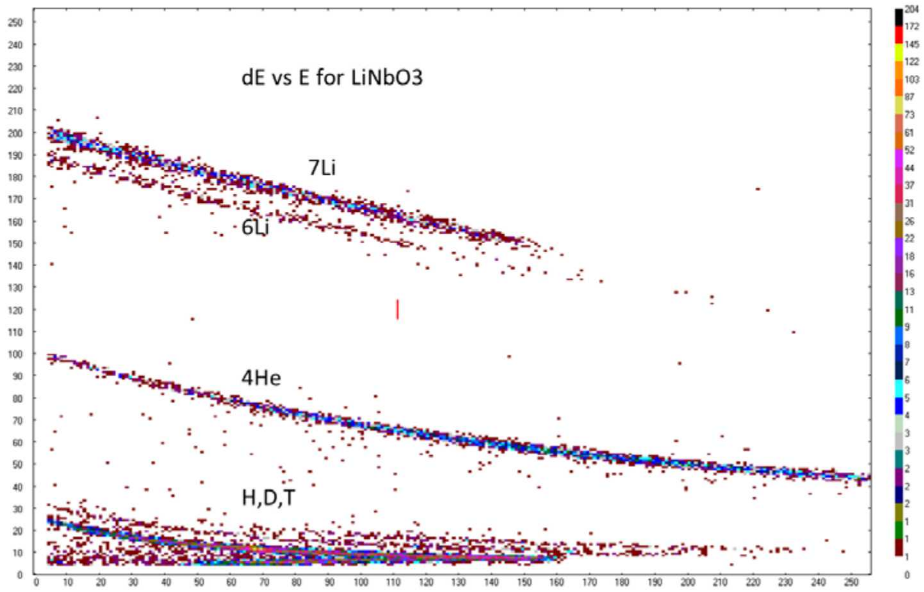




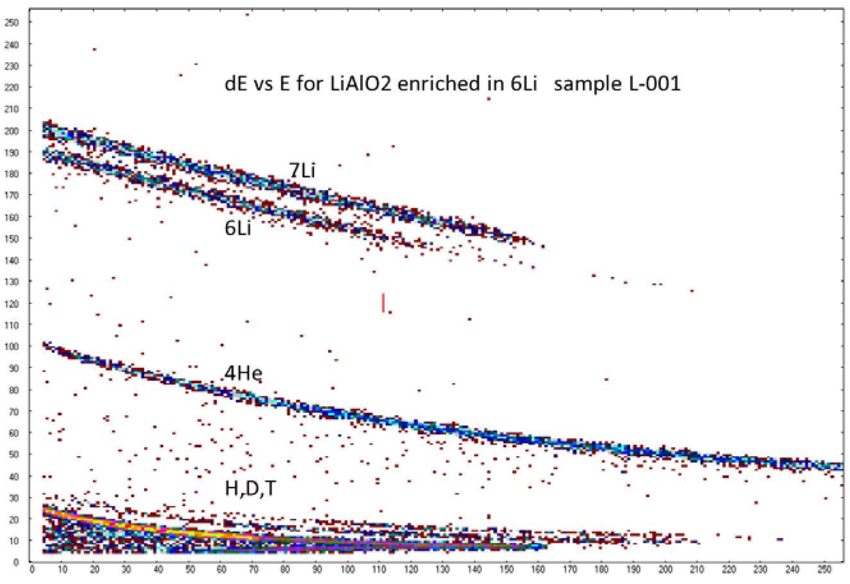
The ^{6}Li fraction has been successfully measured in unirradiated $LiAlO_2$ pellets

- ^{6}Li and ^{7}Li could be separated by using 42 MeV Si^{7+}, and an incident beam angle of 10º and an exit angle of 15º. Telescope with the 44 μm ΔE detector is used.
- The ^{6}Li fraction was measured to be 0.246 ± 0.016, compared to the known value of 0.239.

$LiNbO_3$



$LiAlO_2$



Barney's slab analysis program is used to confirm the position (in channels) of each isotope.

Future Work

- ^{6}Li fraction has been successfully measured in unirradiated pellets
- PNNL has prepared polished irradiated $LiAlO_2$ pellets, mounted in epoxy, that will be sent to SNL in the next few weeks
- We hope to measure the ^{6}Li/^{7}Li ratio, ^{3}H and ^{3}He concentrations for pellets taken from different regions of the reactor to compare with neutronics calculations
- Will implement a thinner ΔE detector that will allow us to reduce beam energy and improve the statistics.