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Ion beam analysis of T and He in tritiated thin films

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Motivation: metal tritide films

- Metal tritide films are essential for applications such as neutron generators.
  - Problems:
  - T decays to ^{3}He, forming bubbles and stressing the film.
  - O contamination influences hydriding efficiency and film stability.

- Tritide film composition and other properties are needed for understanding and controlling hydriding and aging problems.

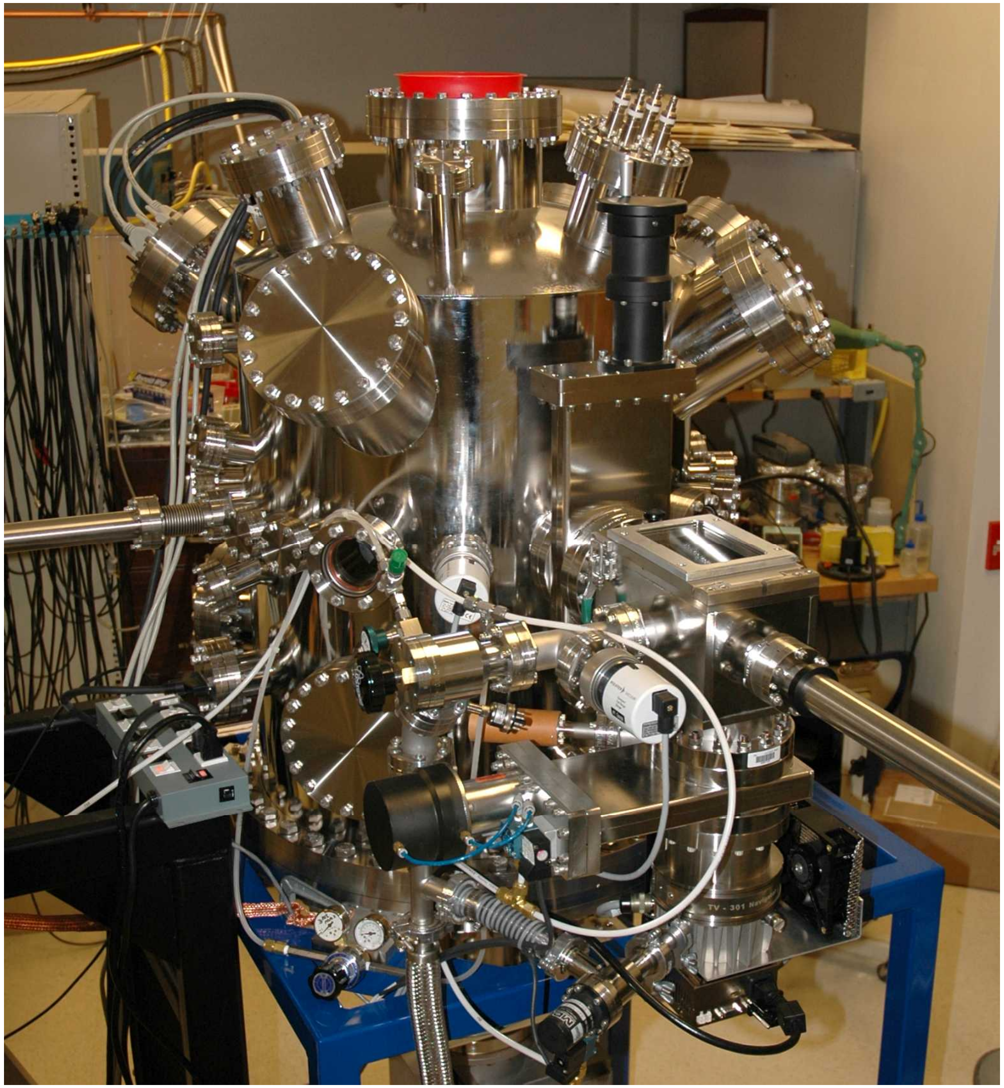
- Several types of films are being studied:
  - ErT_2 films with 100% T, studied as the films age
  - ErD_2 films hydrided under various conditions

- Here we present:
  - Examples of high energy, heavy ion ERD profiling of ^{3}He, T, D, H, O and C in these types of samples.

High energy ERD system

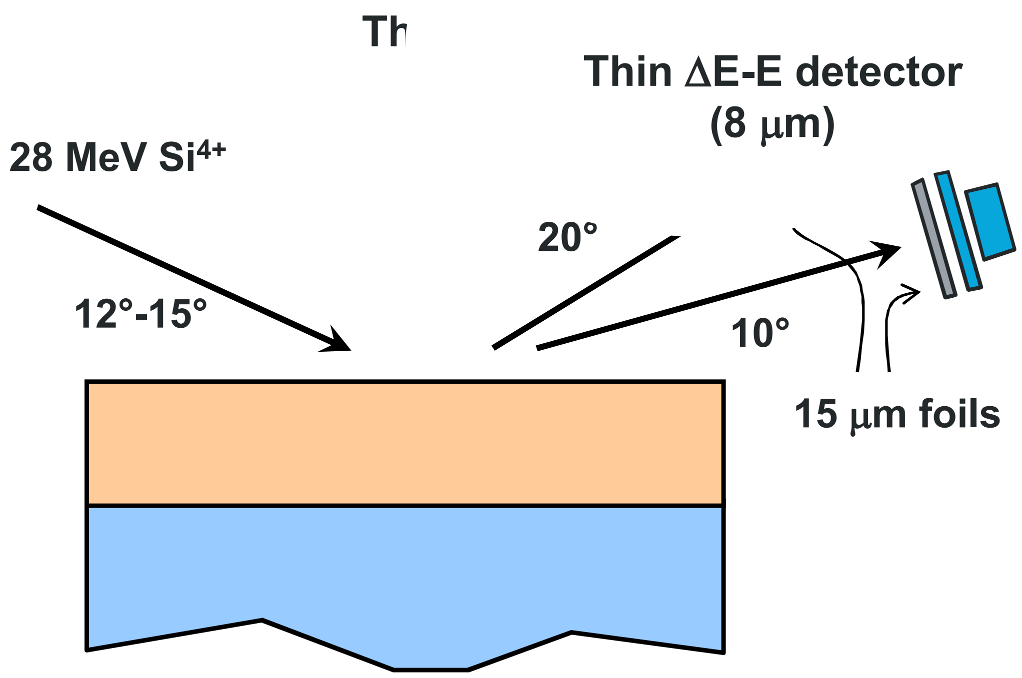
- **Problem:**
  - **- Need to analyze T, ^{3}He, D, H, O, and C.**
  - **- Our existing ERD analysis chamber used for T could not measure ^{3}He or O on Si.**
  - **- New system designed based on high energy, heavy ion beams and ΔE-E detectors.**

- **Analysis chamber:**
  - **- beam line on 7 MV Tandem**
  - **- 6-axis custom goniometer**
  - **- all-metal ion-pumped chamber**
  - **- load lock with heating stage**



Analysis method

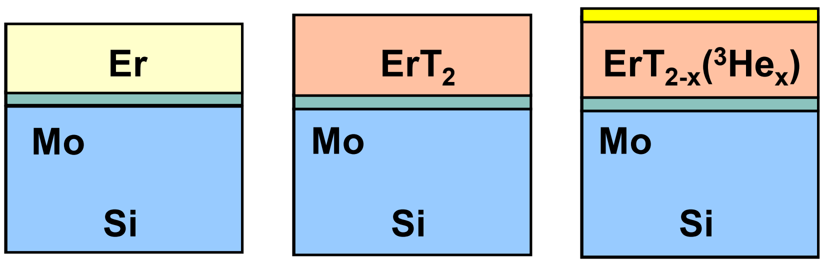
- Heavy ion elastic recoil detection (ERD) with 28 MeV Si^{+4} analysis beam incident at 12°-15°
- Thick ΔE-E detector to profile H, D, T, ^{3}He
- Thin ΔE-E detector to profile O, C
- Each detector pair has a 15 μm foil to block the Si analysis beam and curved slits to optimize resolution.



Example: ErT_2 layers on Mo/Si

Sample preparation

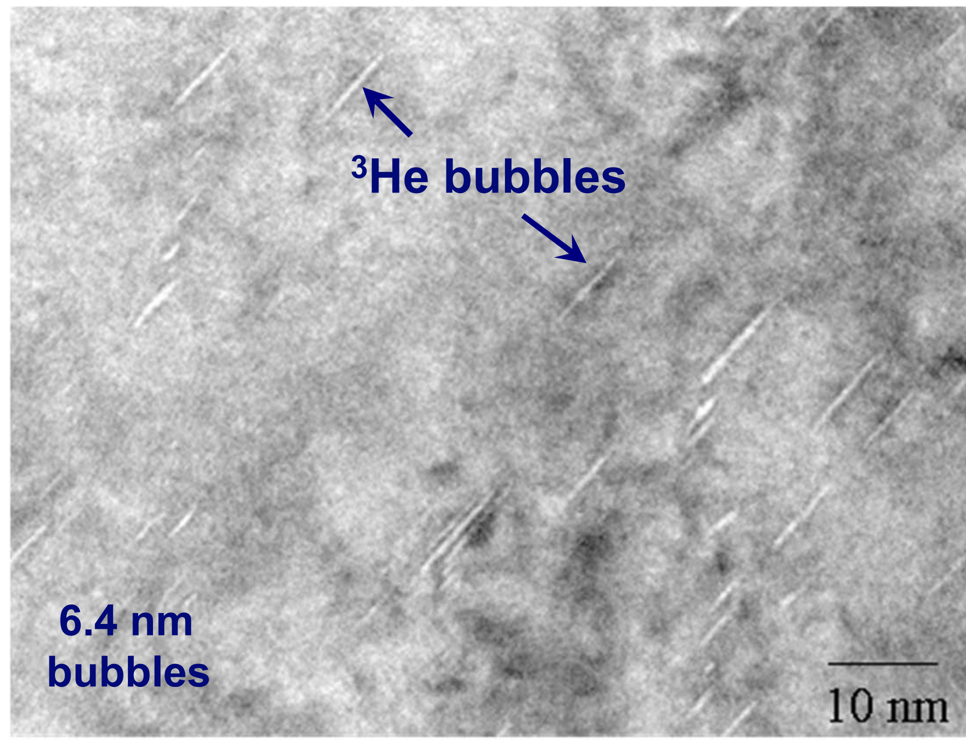
500 nm Er 95 nm Mo Silicon	Hydrided with 100% T at LANL	Aged in vacuum



Oxide forms during hydriding and upon air exposure.

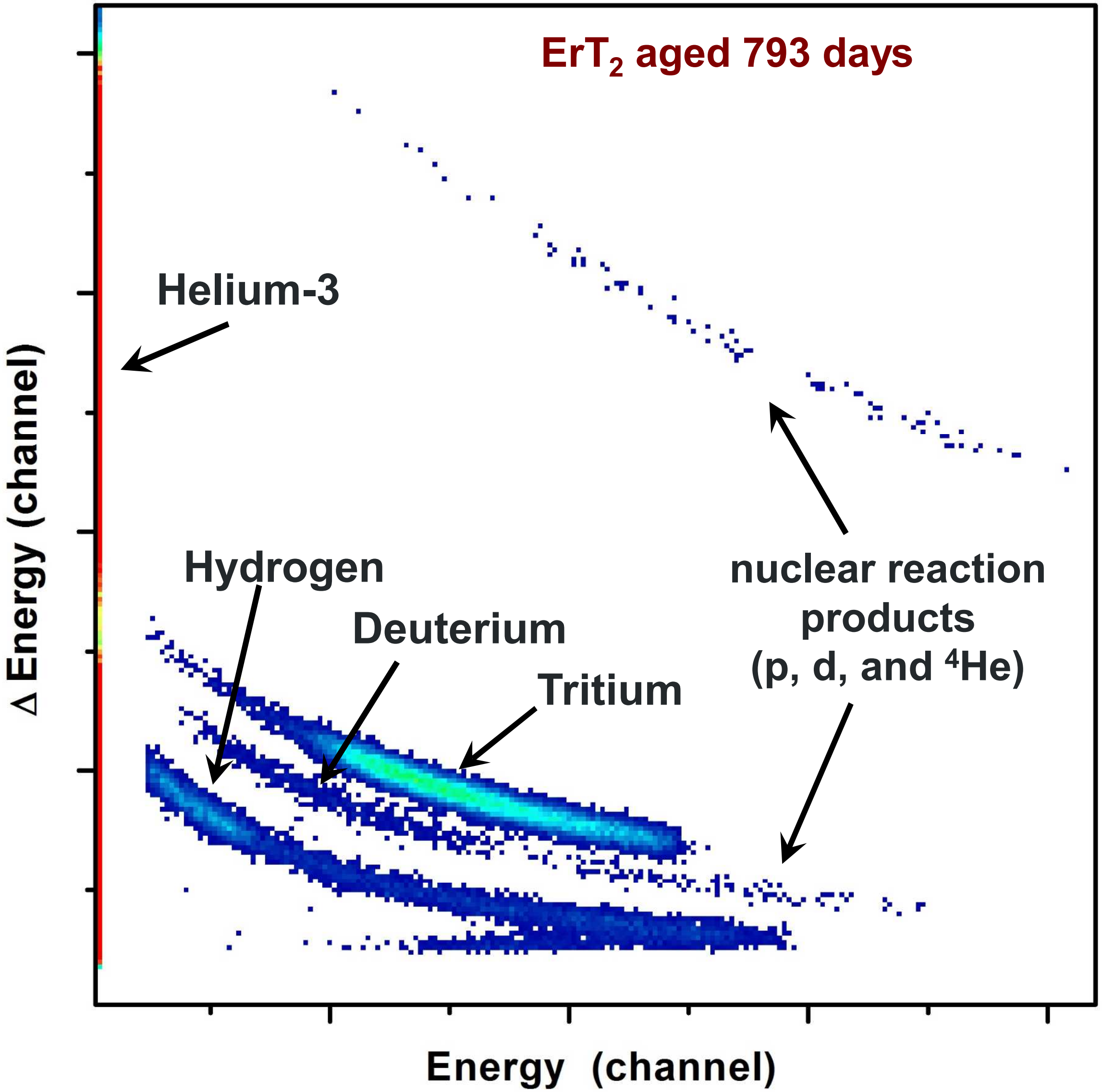
Tritium decays into ^3He, forming platelet-like bubbles on (111) planes.

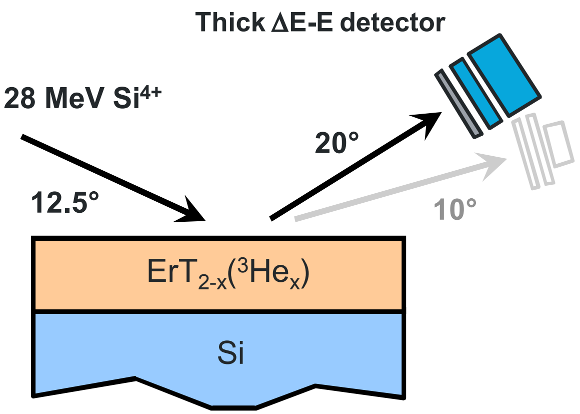
TEM cross-section bright-field, ~{110} zone 62 days after hydriding.



TEM with Gillian Bond, New Mexico Institute of Mining and Technology

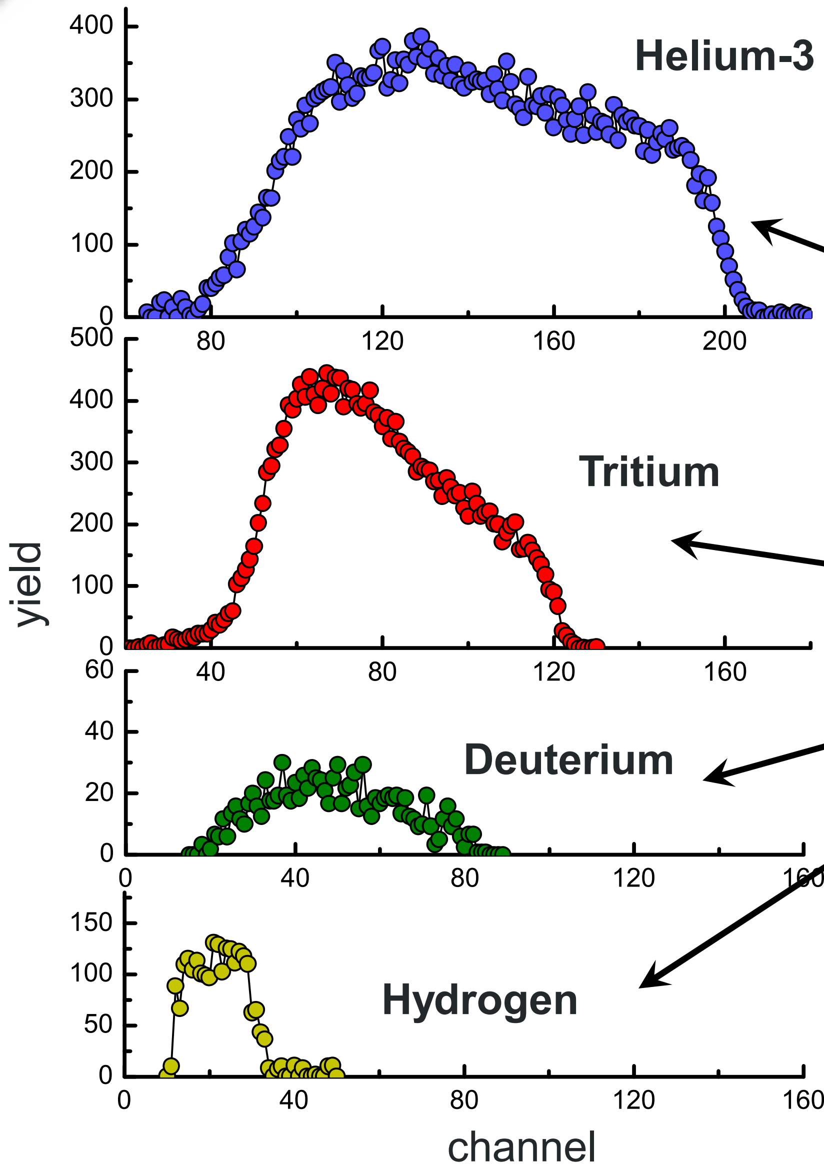
Coincidence map for **thick** ΔE-E detector

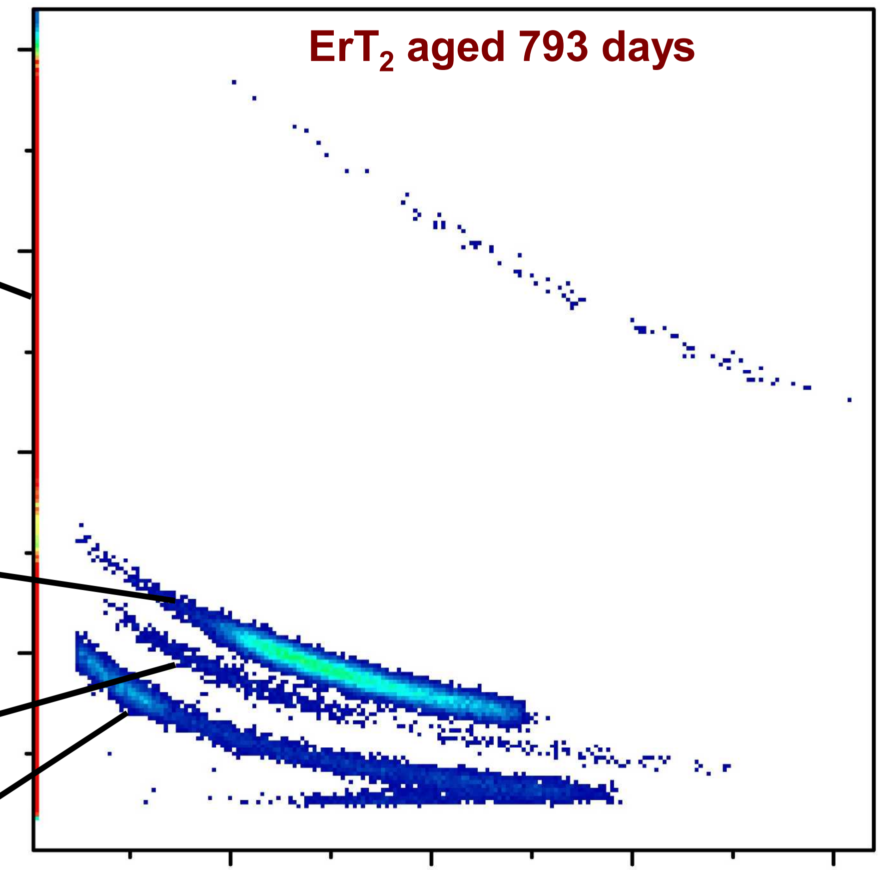




12.5° in, 20° out
28 MeV Si^{+4} beam
15 μm foil
44 μm ΔE detector

Spectra for **thick** ΔE-E detector

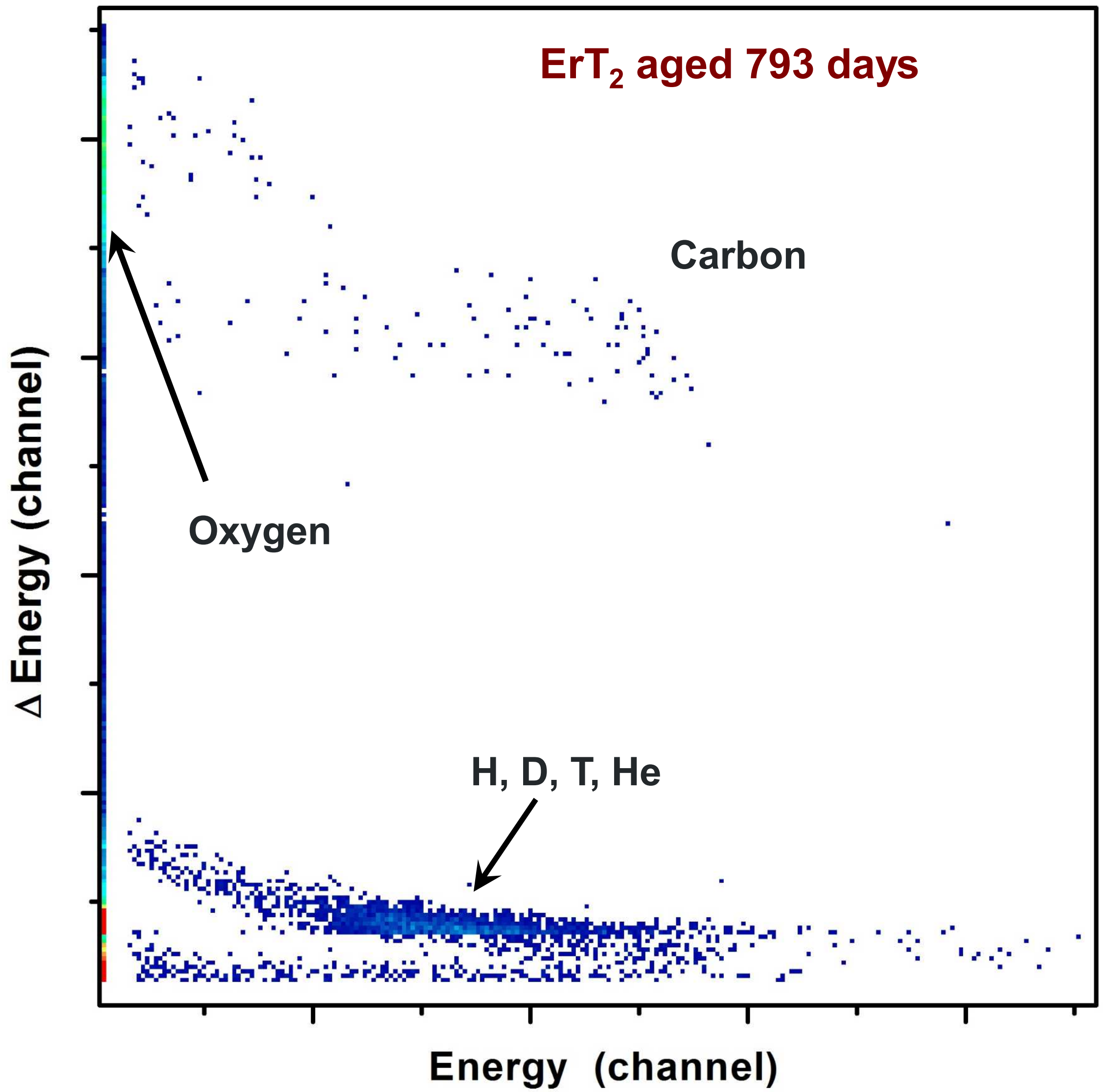


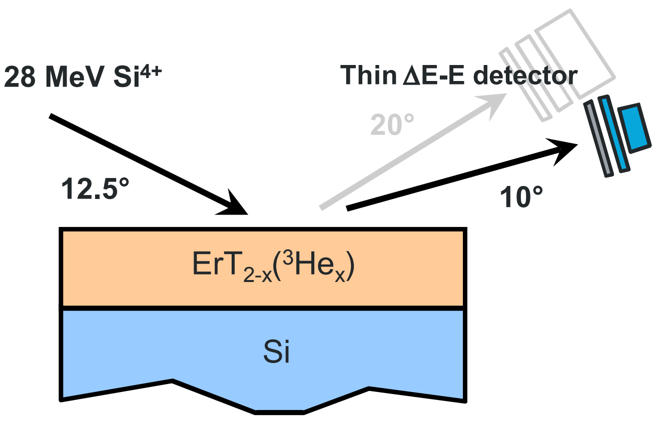


12.5° in, 20° out
15 µm foil

28 MeV Si^{+4} beam
44 µm ΔE detector

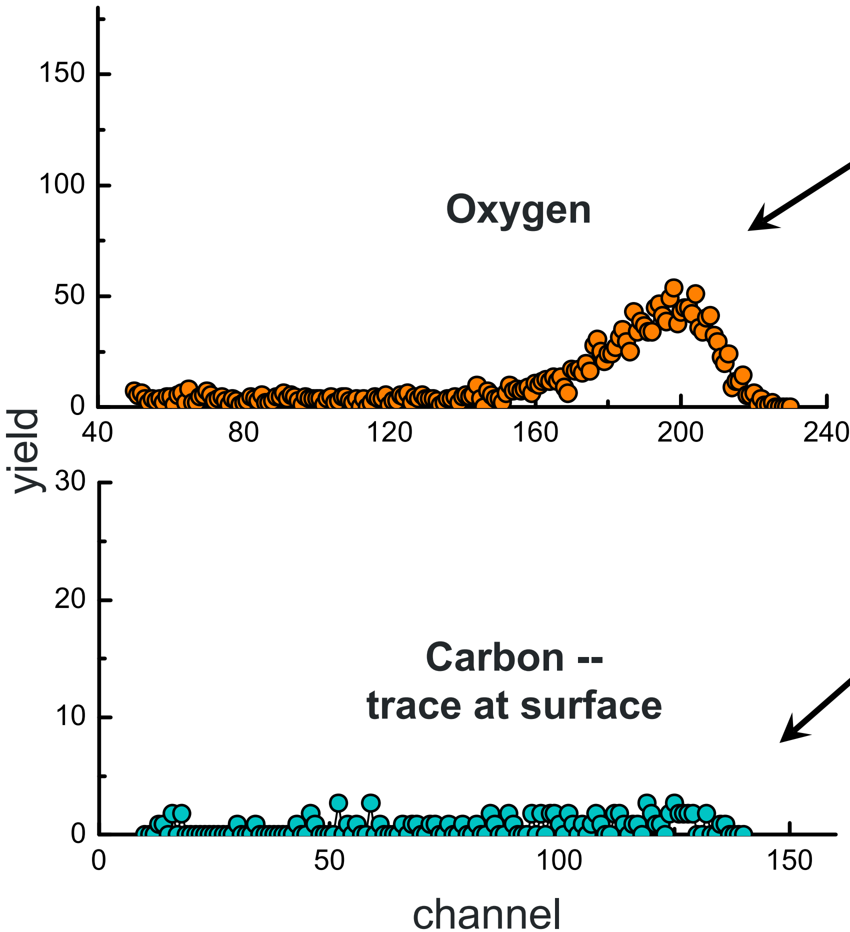
Coincidence map for thin ΔE-E detector

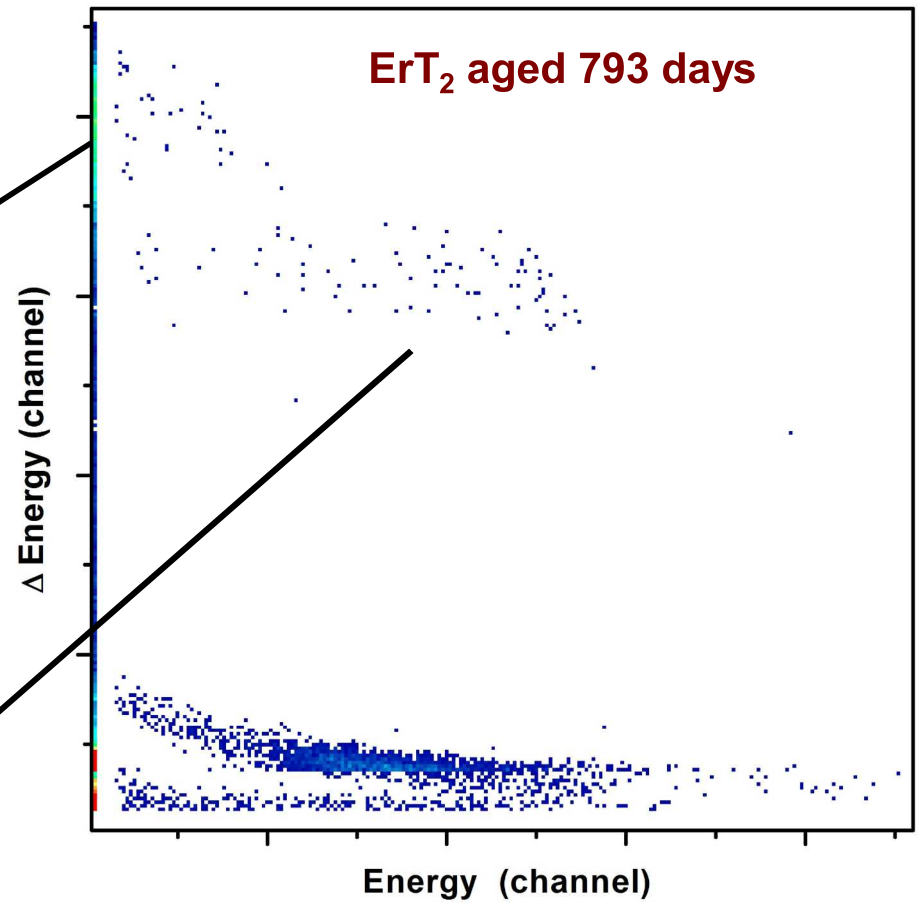




12.5° in, 10° out
28 MeV Si^{+4} beam
15 μm foil
8 μm ΔE detector

Spectra for thin ΔE-E detector





12.5° in, 10° out
15 μm foil

28 MeV Si^{+4} beam
8 μm ΔE detector

Analysis using SIMNRA

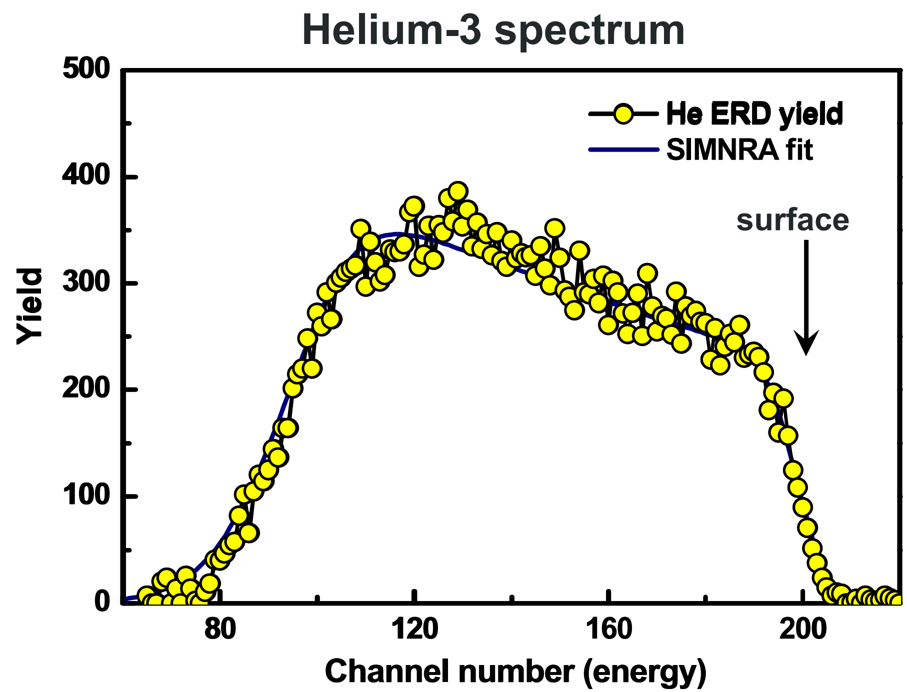
-- Direct conversion of ERD data to a depth profile is generally not possible.

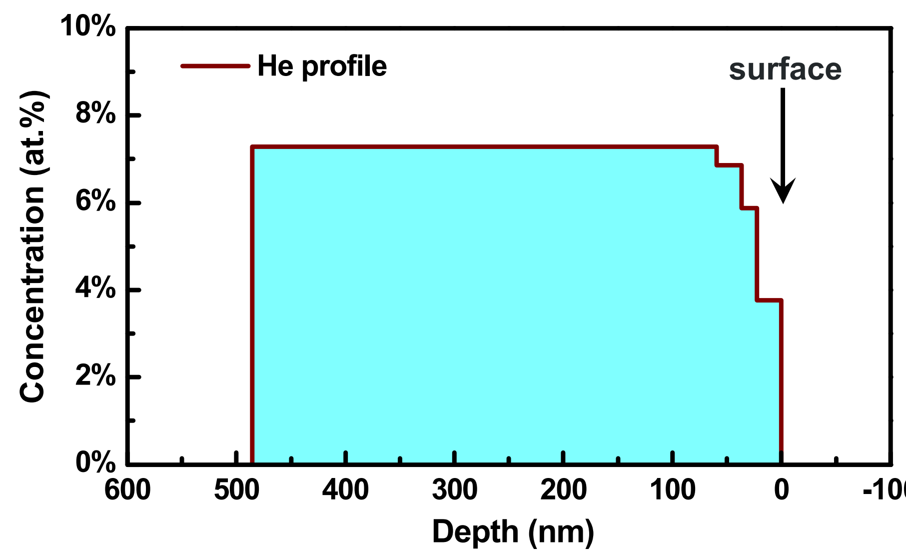
-- Instead, the data must be analyzed by fitting with an ion beam interaction code such as SIMNRA*.

-- A test profile including all elements is input, along with energies, angles, etc.

-- Ion interaction cross-sections, stopping powers, straggling and geometrical broadening are all calculated by the simulation.

-- The concentration profile is varied until a best fit is obtained.





*SIMNRA vs. 6.03, Dr. Matej Mayer, Max Planck Inst.

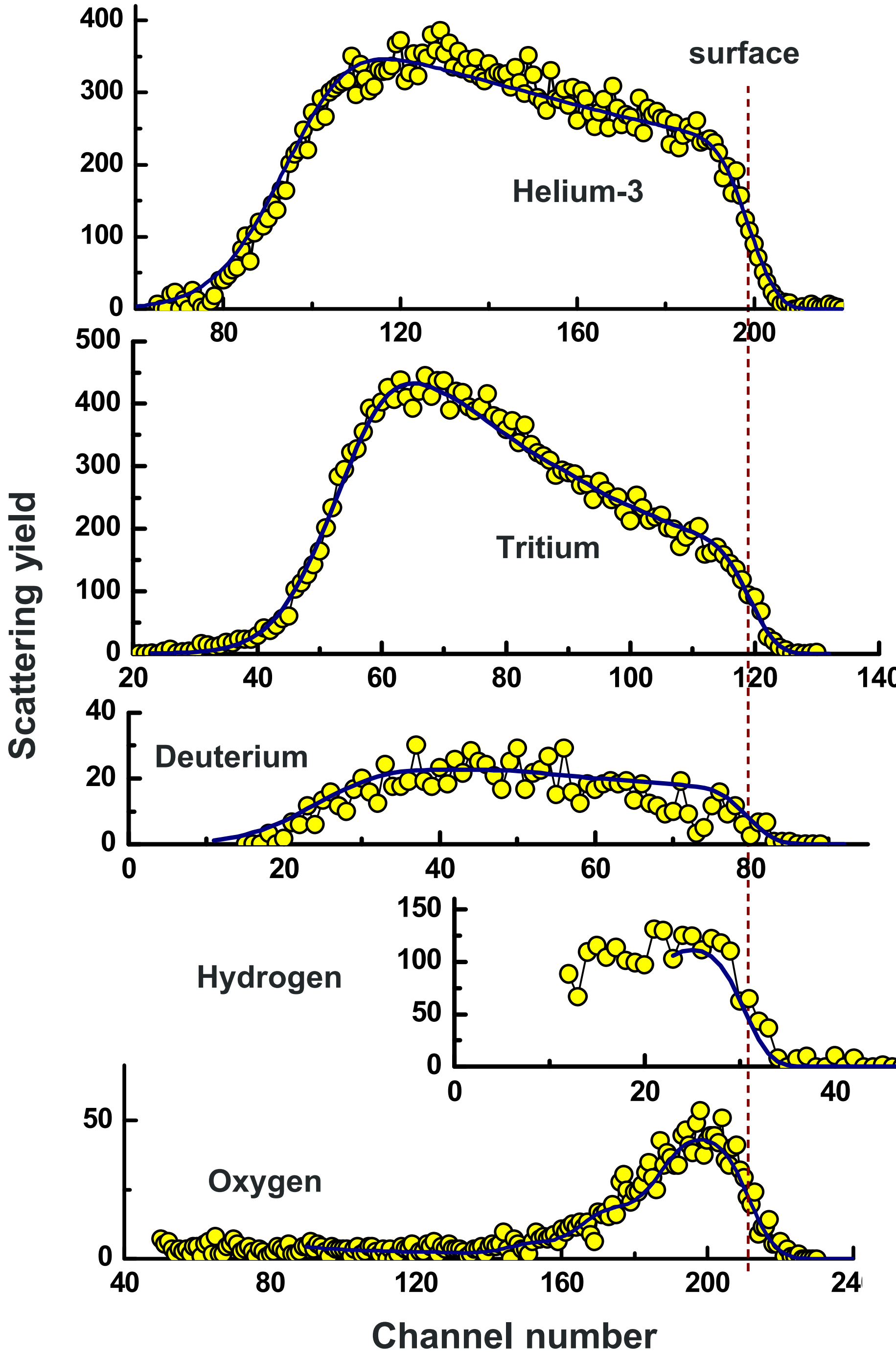
Analysis using SIMNRA

SIMNRA fits of the elemental spectra from the ErT_2 sample aged 793 days

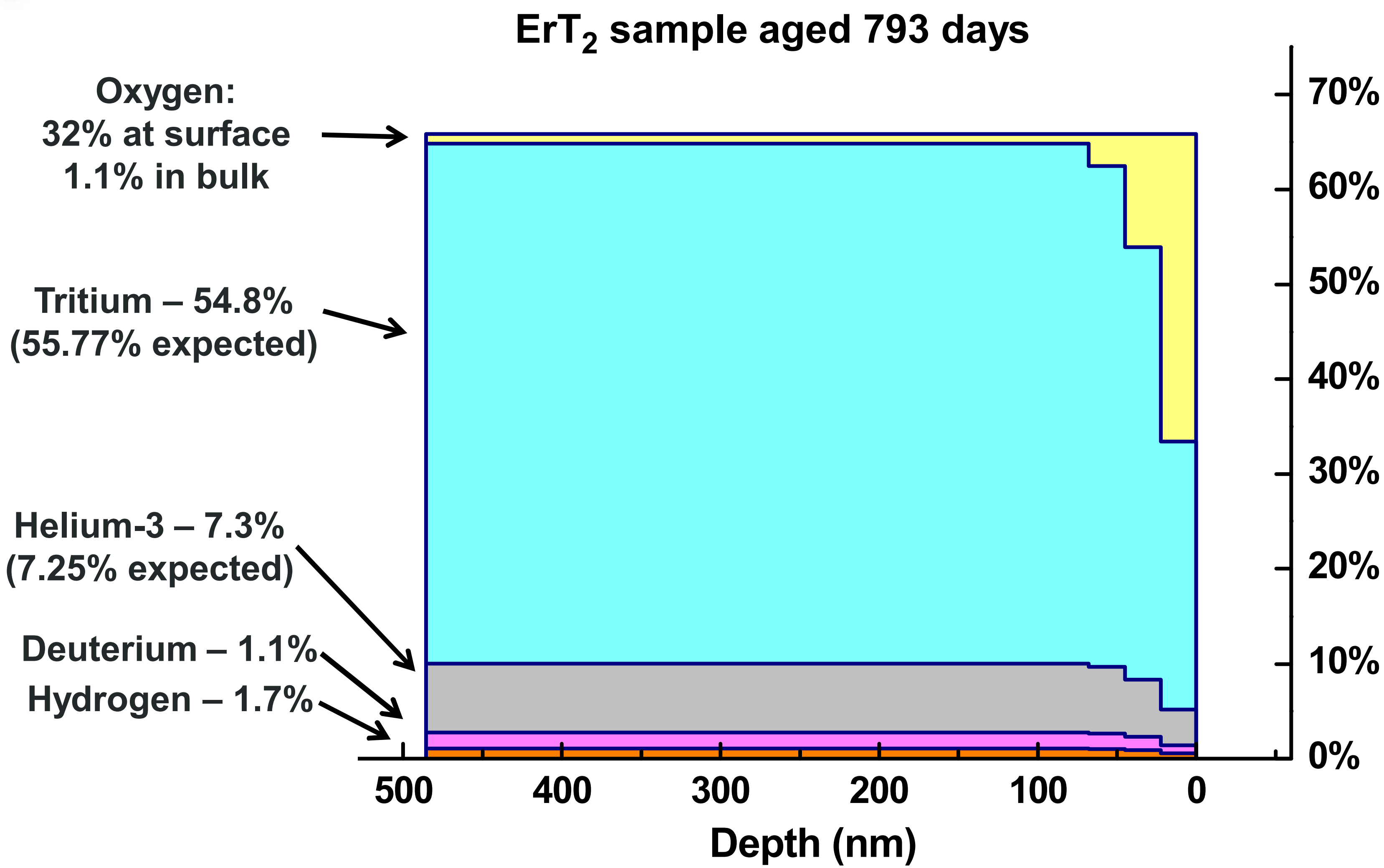
-- Broadening is due to both straggling and sample roughness.

-- T cross-section is non-Rutherford and had to be deduced by fitting T spectra obtained at 24–30 MeV, using a sample calibrated at lower energy.

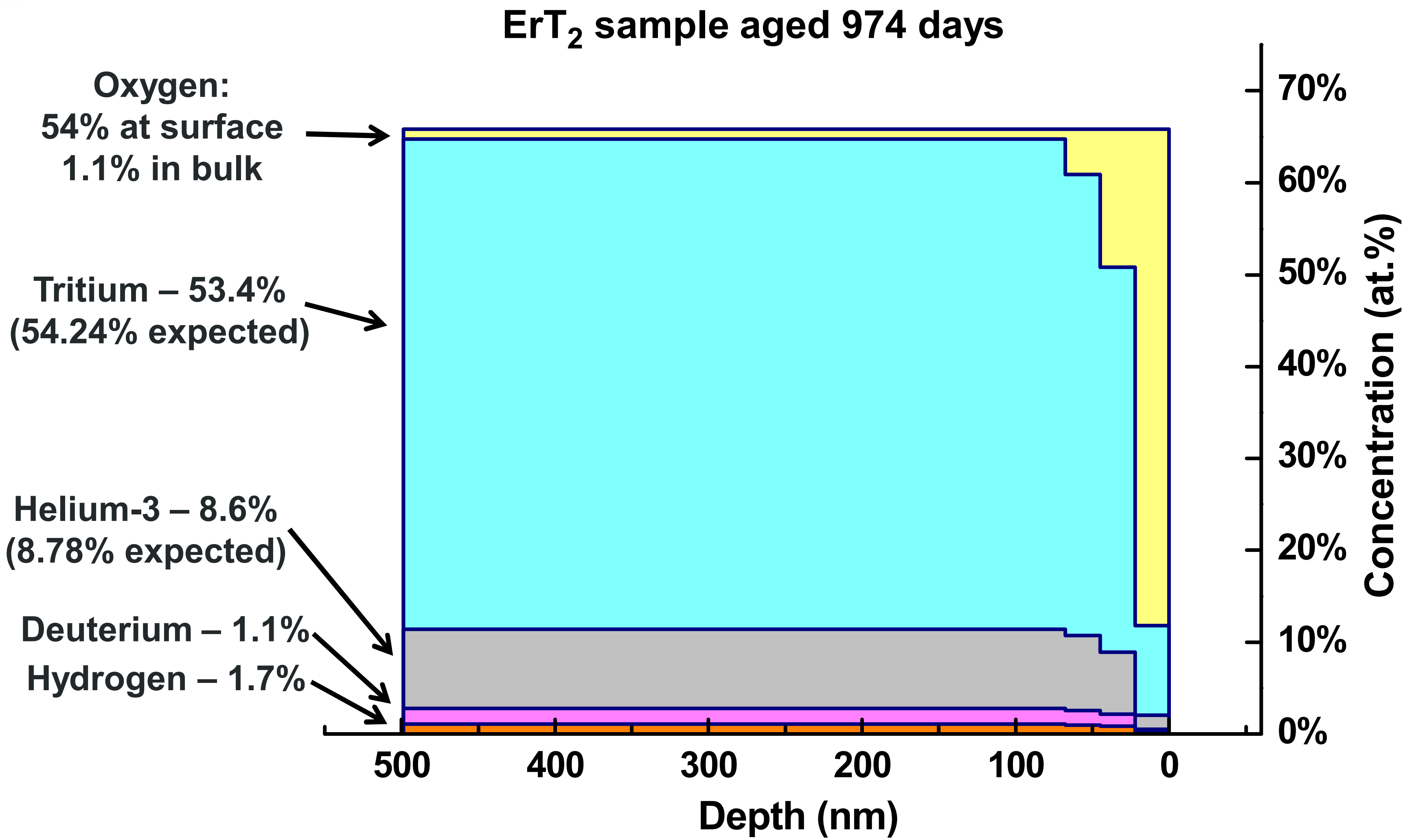
-- Energy and hence depth resolution vary with element – the best depth resolution is for O.



Concentration profiles using SIMNRA

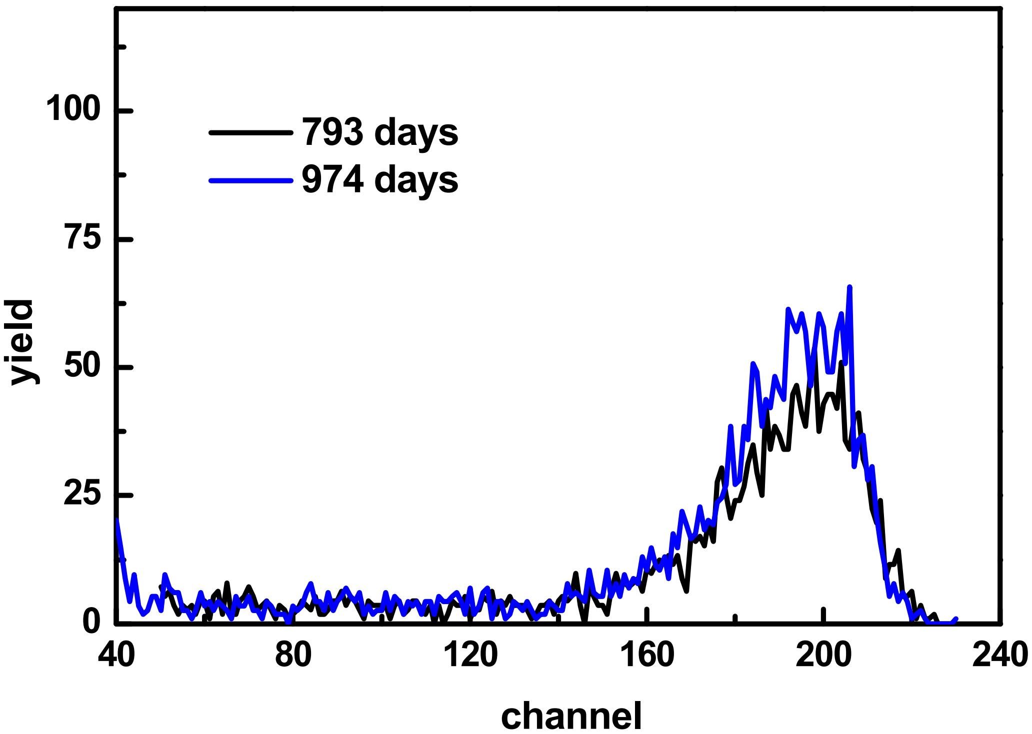


Concentration profiles using SIMNRA

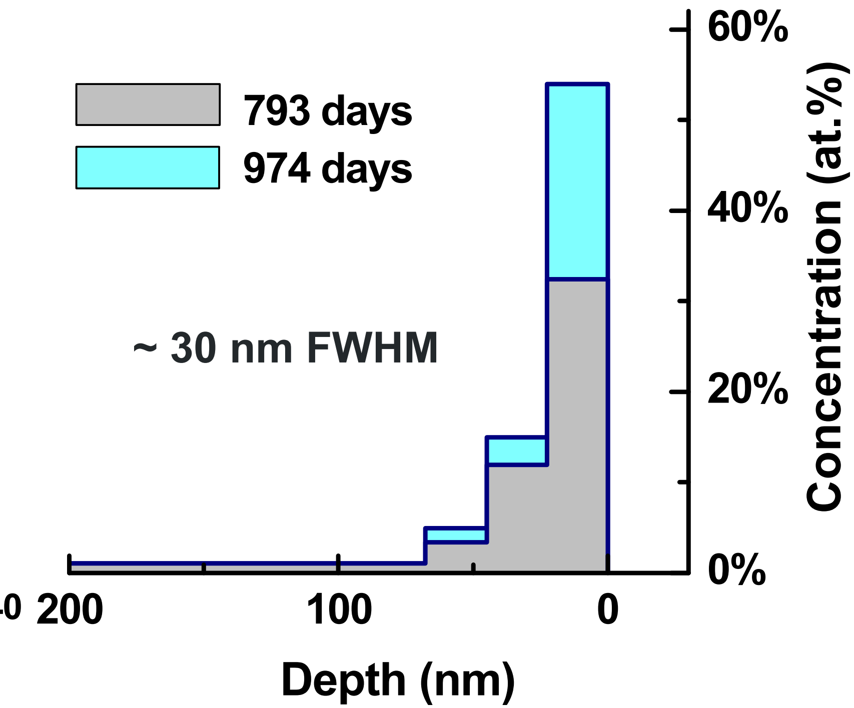


Oxide growth with aging

Normalized ERD Oxygen spectra (same ErT_2 sample)



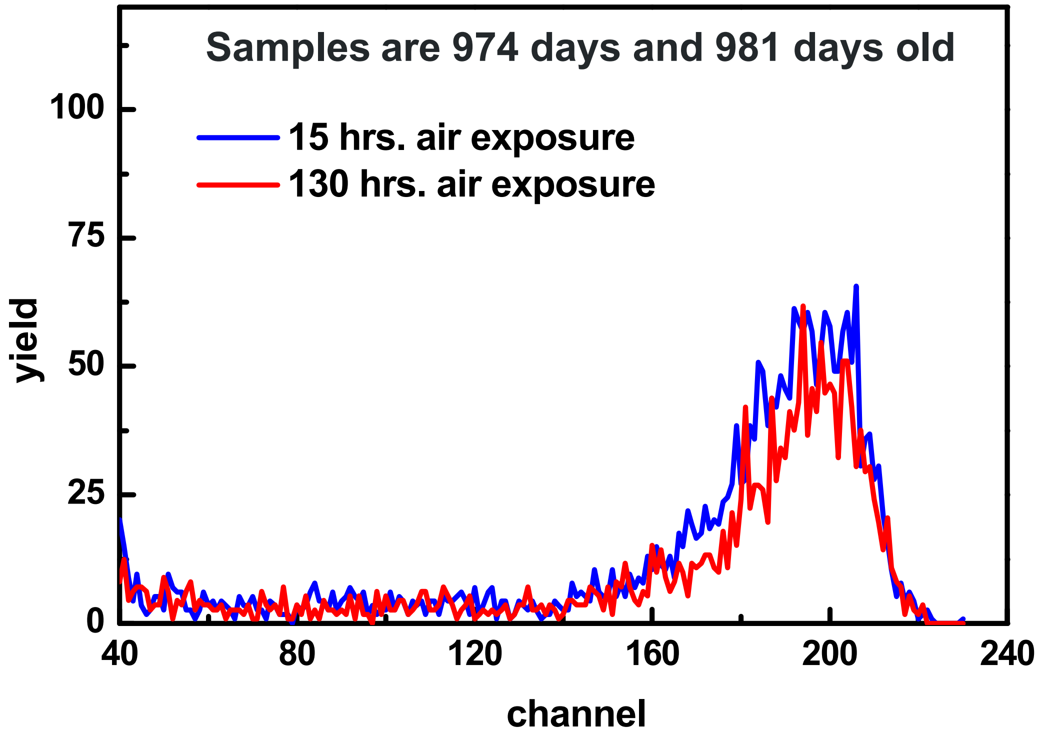
SIMNRA Oxygen profiles



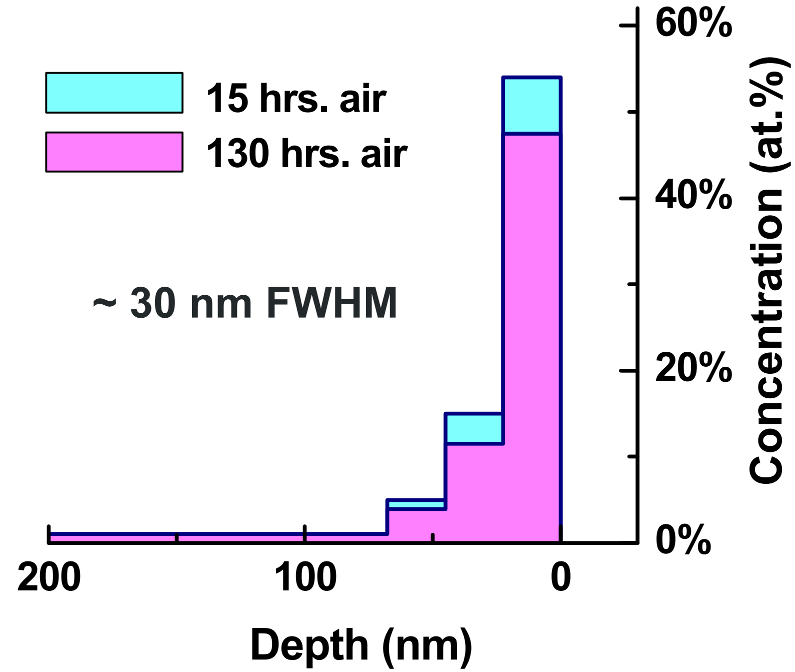
The oxide on this ErT_2 sample is growing slowly as it ages. Spectral broadening masks the effect somewhat in the raw data.

Oxide growth with air exposure

Normalized ERD Oxygen spectra (same age ErT_2 samples)



SIMNRA Oxygen profiles

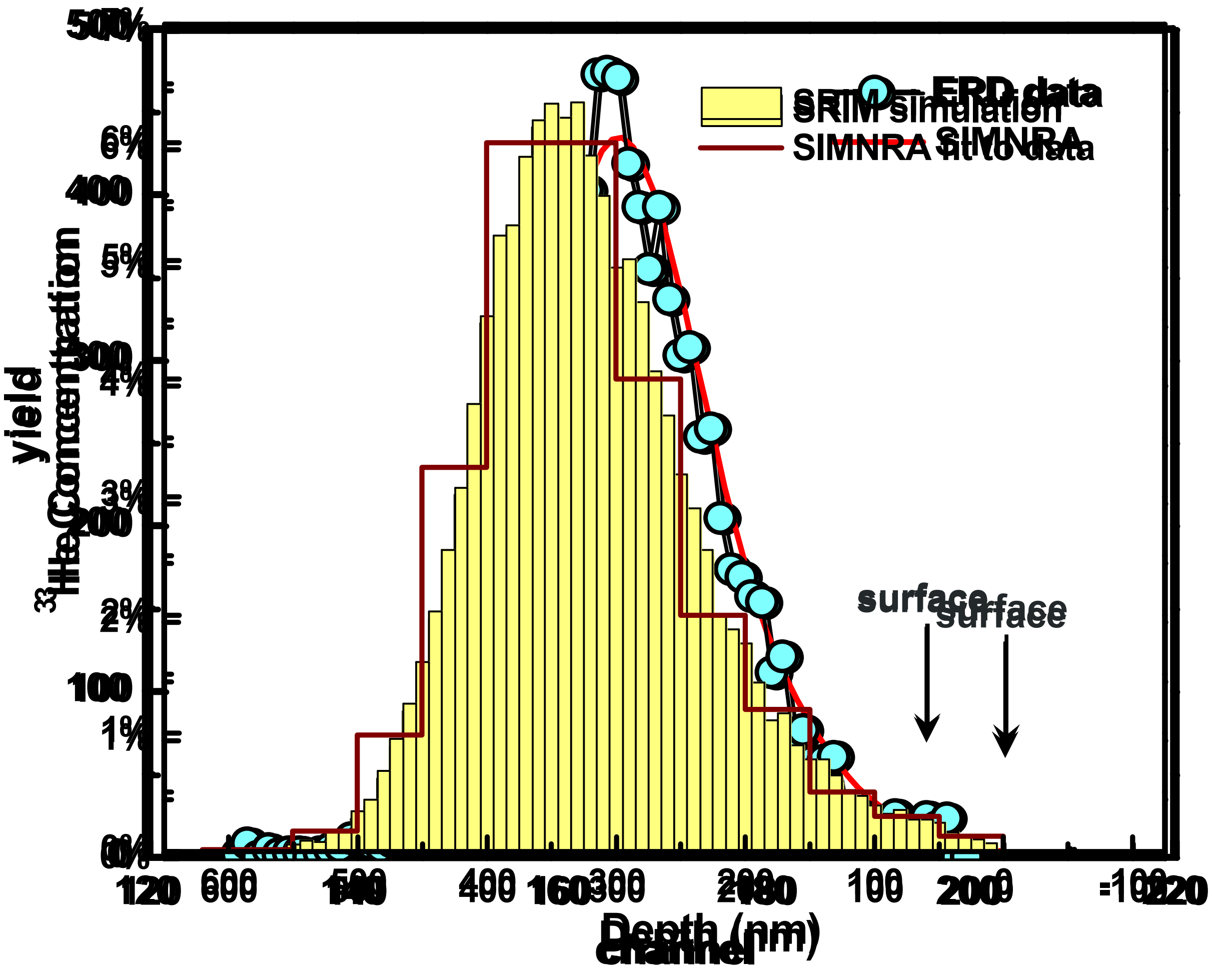


The oxides on these parallel ErT_2 samples are nearly identical, even though the 2nd sample has been out of vacuum nearly 10X as much.

⇒ Oxide growth may be controlled by the T activity.

Example: ^{3}He-implanted Si

1.6 x 10^{16} ^{3}He/cm^{2} implanted at 40 keV into Si.



- -- Single energy implant, resulting in a peak 6 at.% ^{3}He at 350 nm.
- -- Depth distribution calculated using the SRIM 2003 Monte Carlo code.
- -- 28 MeV ERD spectrum has the same general shape.
- -- SIMNRA fitting with a multi-layer profile matches the experiment.
- -- The SIMNRA profile is very close to the SRIM distribution.

Summary

A new ERD ion beam analysis system allows non-destructive profiling of ^{3}He, T, H, D, O, and C with down to 10 nm resolution.

Issues:

-- very sensitive to sample thickness and mounting angles.

-- cross-sections and stopping powers can be uncertain at these energies and for these elements (T, ^{3}He, D).

-- nuclear reaction products can interfere.

-- sample roughness limits depth resolution.

-- most problems solved by use of standard samples.