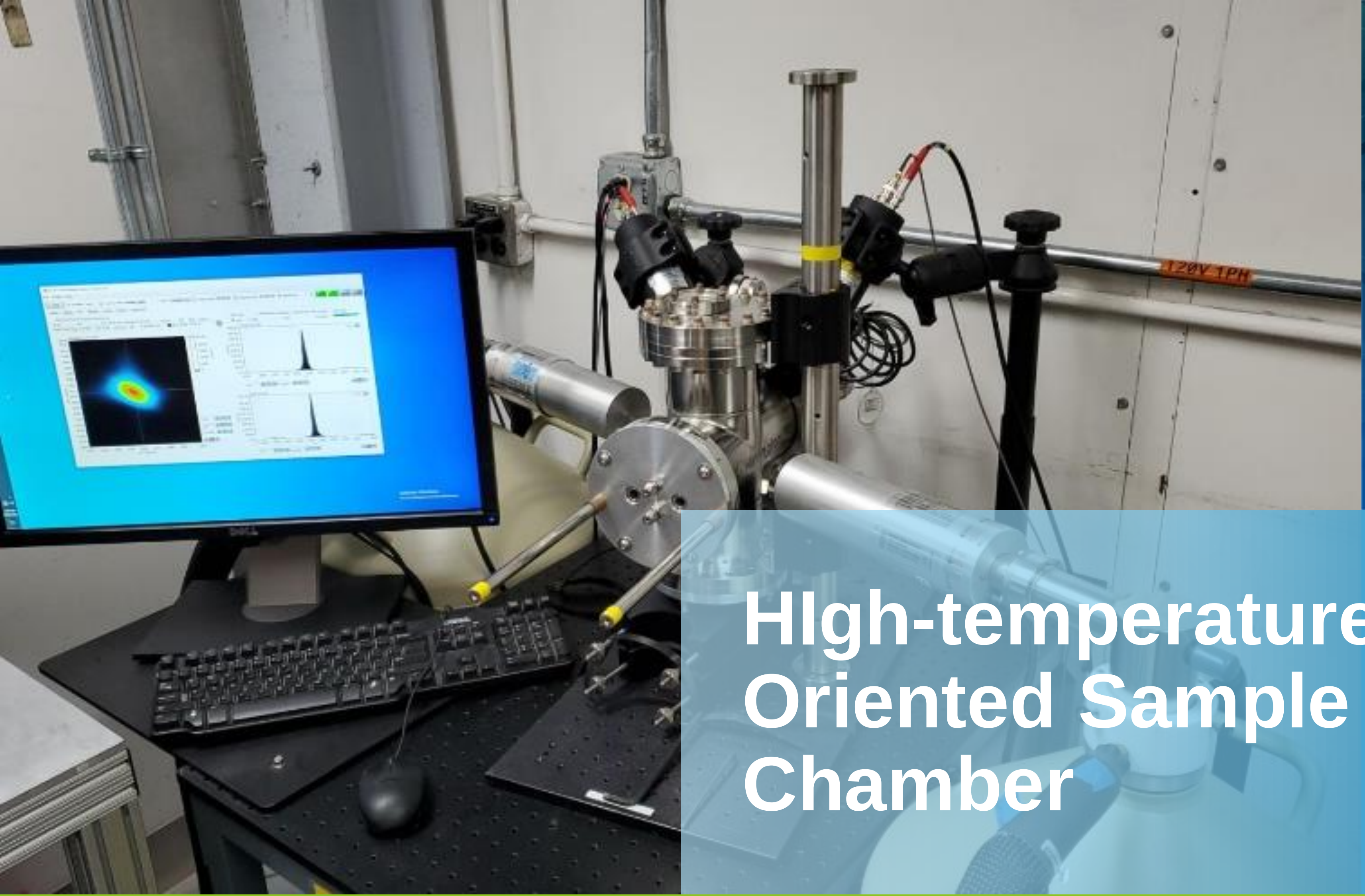


INL/CON-24-77578

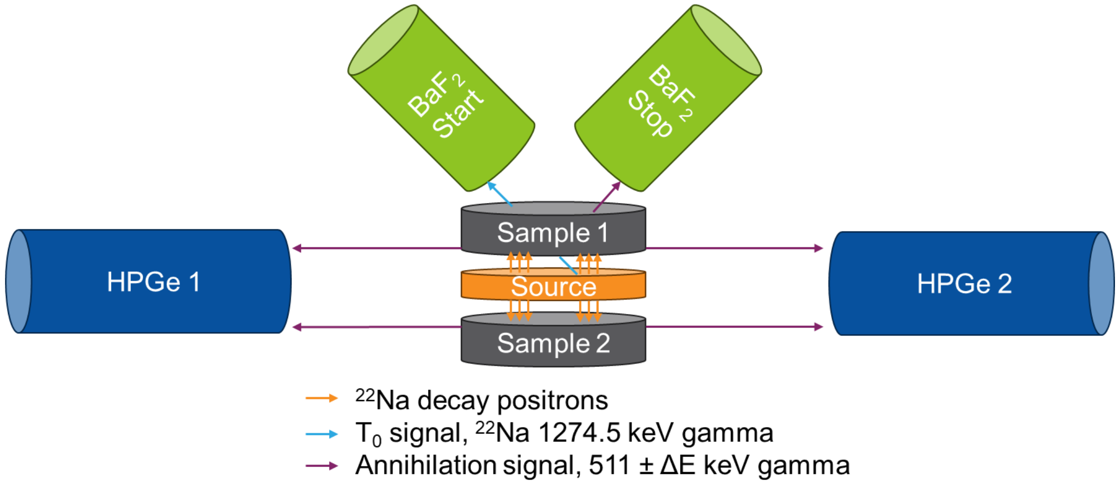
Chuting Tsai

PIE and Advanced Characterization Division

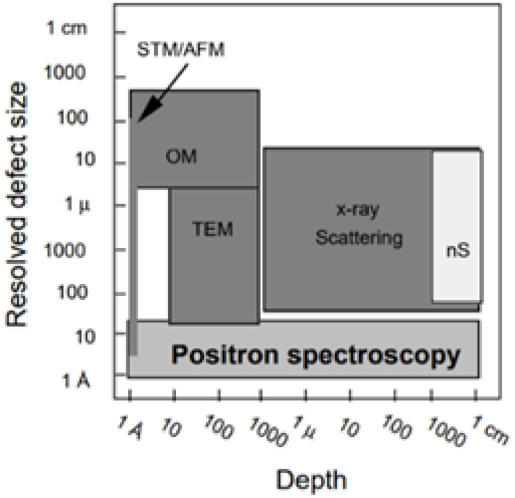
HIgh-temperature Portable PAS-Oriented Sample (HIPPOS) Chamber

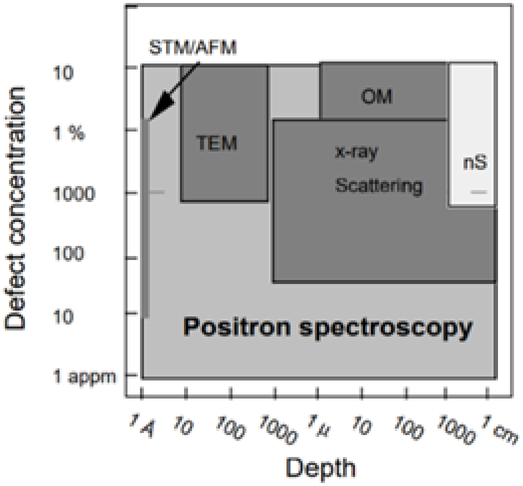
PAS Technique

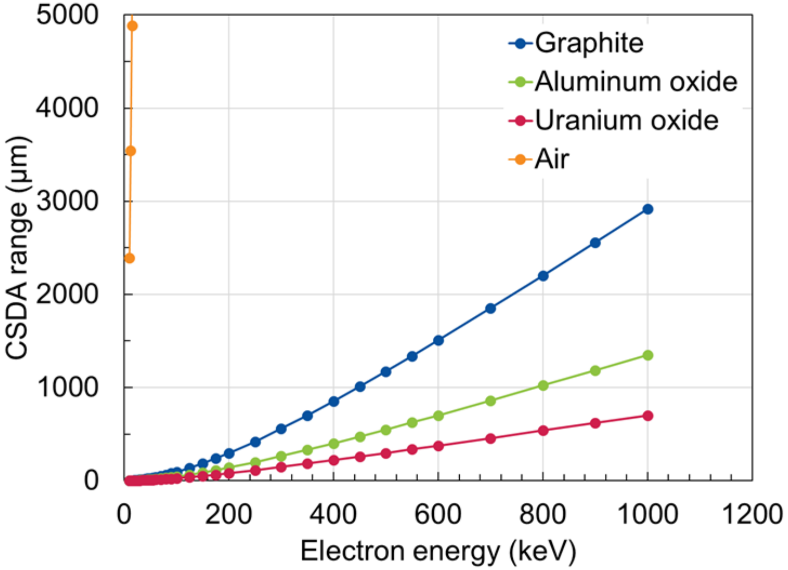
- Microstructure evolution plays an important role in material degradation
- Vacancy type of defects is not well understood in irradiated materials
- Two measuring modes simultaneously
  - Positron annihilation lifetime spectroscopy
  - Coincidence doppler broadening spectroscopy



PAS Technique – Continued







- **Why suitable for PIE?**
  - A bulk technique and a microscopic technique
  - Probing vacancy type defects
  - High sensitivity
  - Has been utilized in metallic, ceramic, semiconductors, polymers etc industries

- **Current Status**
  - Does not provide sufficient data as a stand-alone PIE technique*
  - No pretty images*
  - Utilizing samples from other techniques
  - More suitable for neutron irradiated specimens
  - Correlate well with mesoscale modeling

Mallon, P. E., and D. M. Schrader. *Principles and applications of positron & positronium chemistry*. World Scientific, 2003.

Positron Annihilation Spectroscopy (PAS) Development

2019

Purchased the digital unit

Purchased Na-22 source (LDRD)

2020

Assembled system

Commissioned PALS

Salvaged HPGe detectors and parts from previous system

2021

Rebuilt HPGe detectors (LDRD)

Commissioned CDB (LDRD)

2022

Started working on CDB-AP software (LDRD)

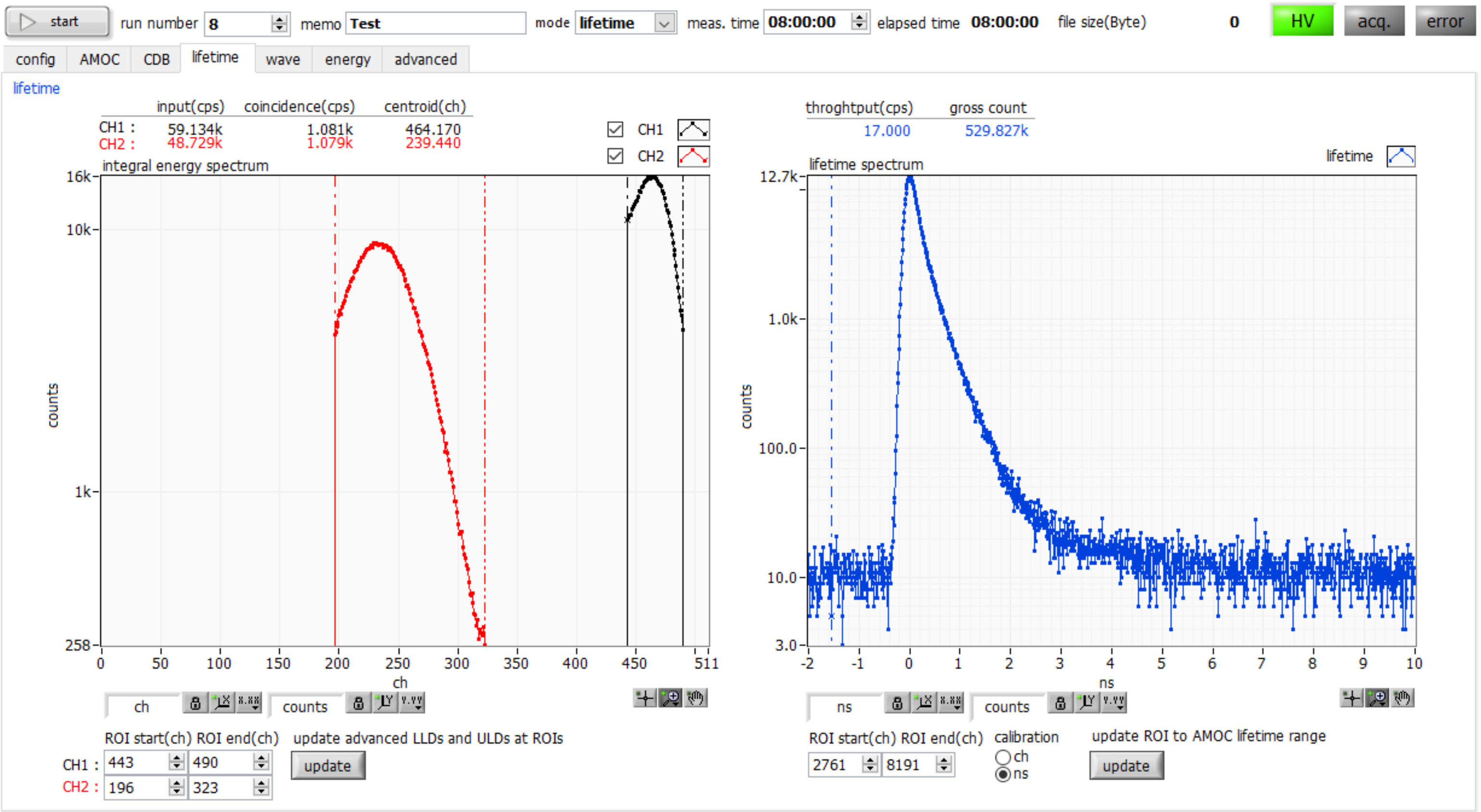
Upgraded PALS hardware for longer lifetime measurements (LDRD)

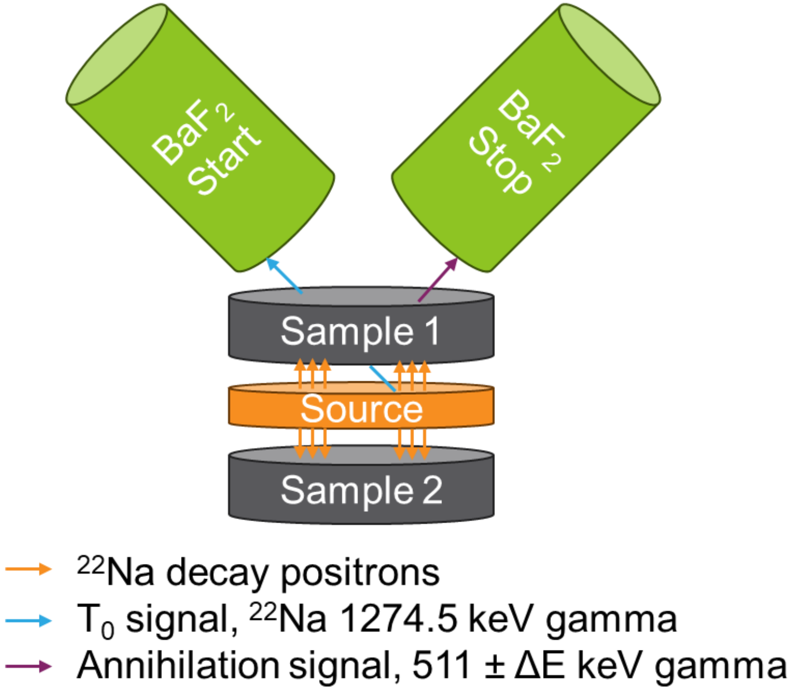
2023

Acquired HIPPOS chamber

Published CDB-AP software

Positron Annihilation Lifetime Spectroscopy

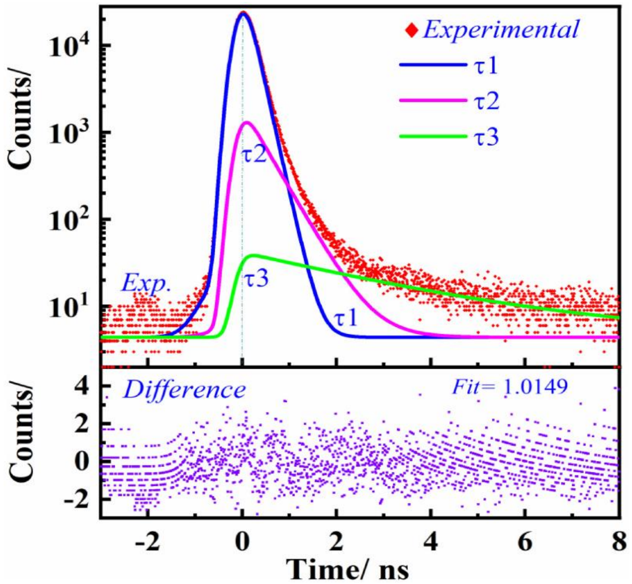




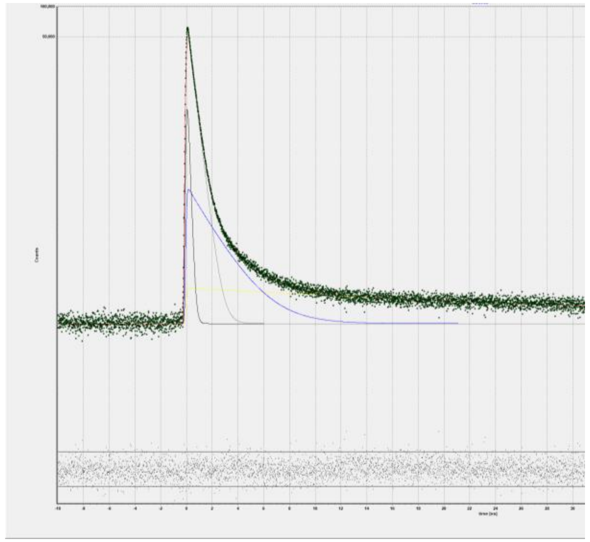
Defects Correlation Using PALS

$$N(t) = \sum_{i=1}^{n} \frac{I_i}{\tau_i} exp\left(\frac{-t}{\tau_i}\right)$$

- Size (τ_i) and concentration (I_i)
- Open volume – no positive ion
  - Lower potential for positron trapping
- Larger vacancies = longer lifetime
  - Decreased density of electrons
- No such correlation if
  - Cavity too big (> 500 ps)
  - Gas bubbles
- Absolute value of defect density can be obtained with theoretically determined trapping coefficient for specific materials*



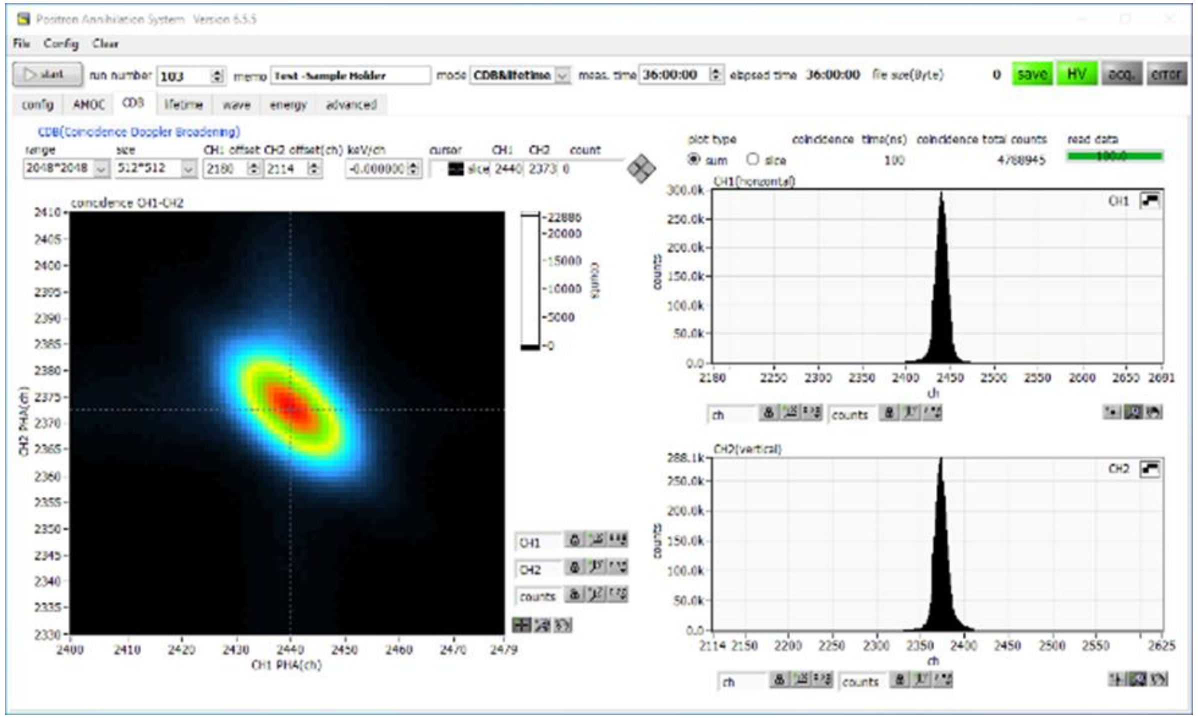
YAG

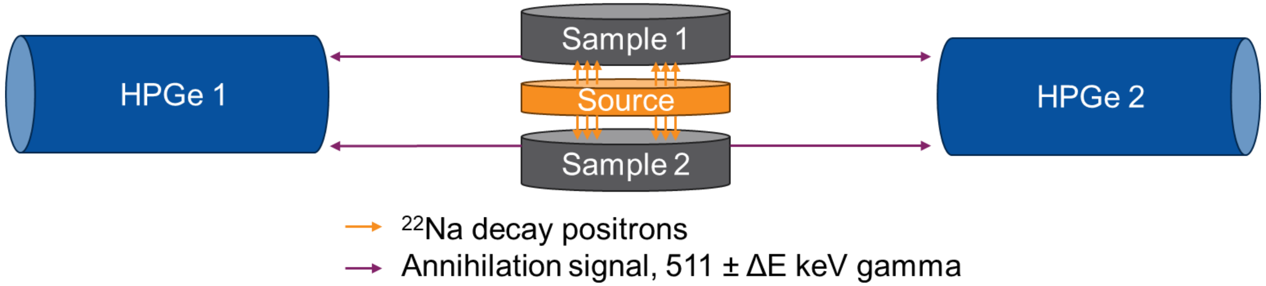


Polymer

Selim, F. A. "Positron annihilation spectroscopy of defects in nuclear and irradiated materials-a review." *Materials Characterization* 174 (2021): 110952.

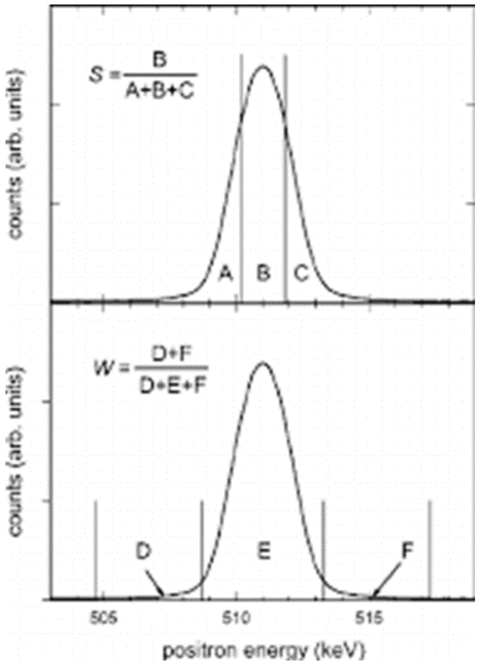
Coincidence Doppler Broadening Spectroscopy

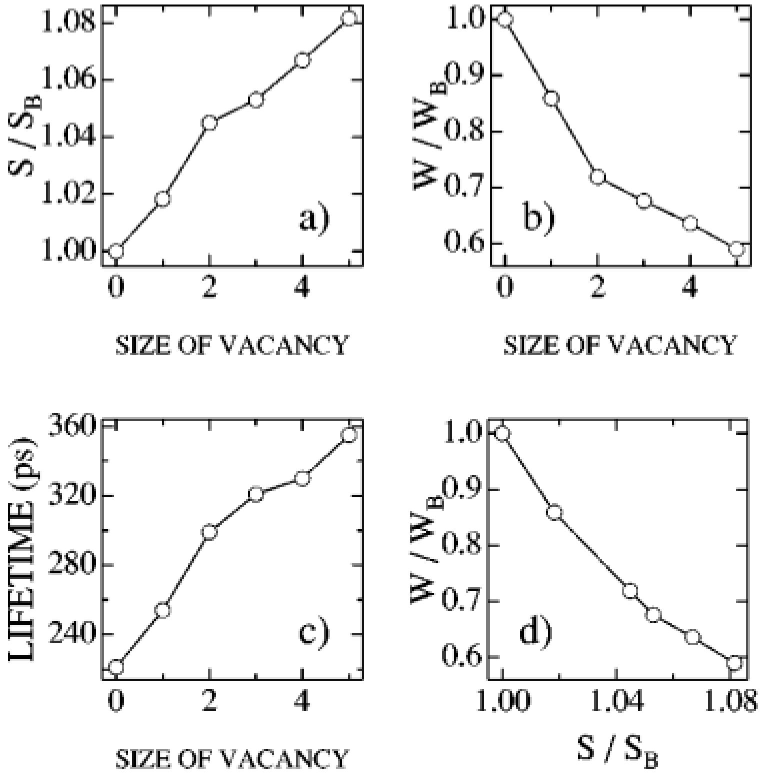




Defects Correlation using CDB

- Non zero electron momentum at annihilation site
- Coincidence not necessary
  - Reduce background
  - Irradiated materials
- S parameter represents annihilation with electrons with low energy/momentum, hence the unbound and valence electrons
- W parameter correlates with the core electrons
- Larger size vacancies a = higher S parameter and a suppressed W parameter



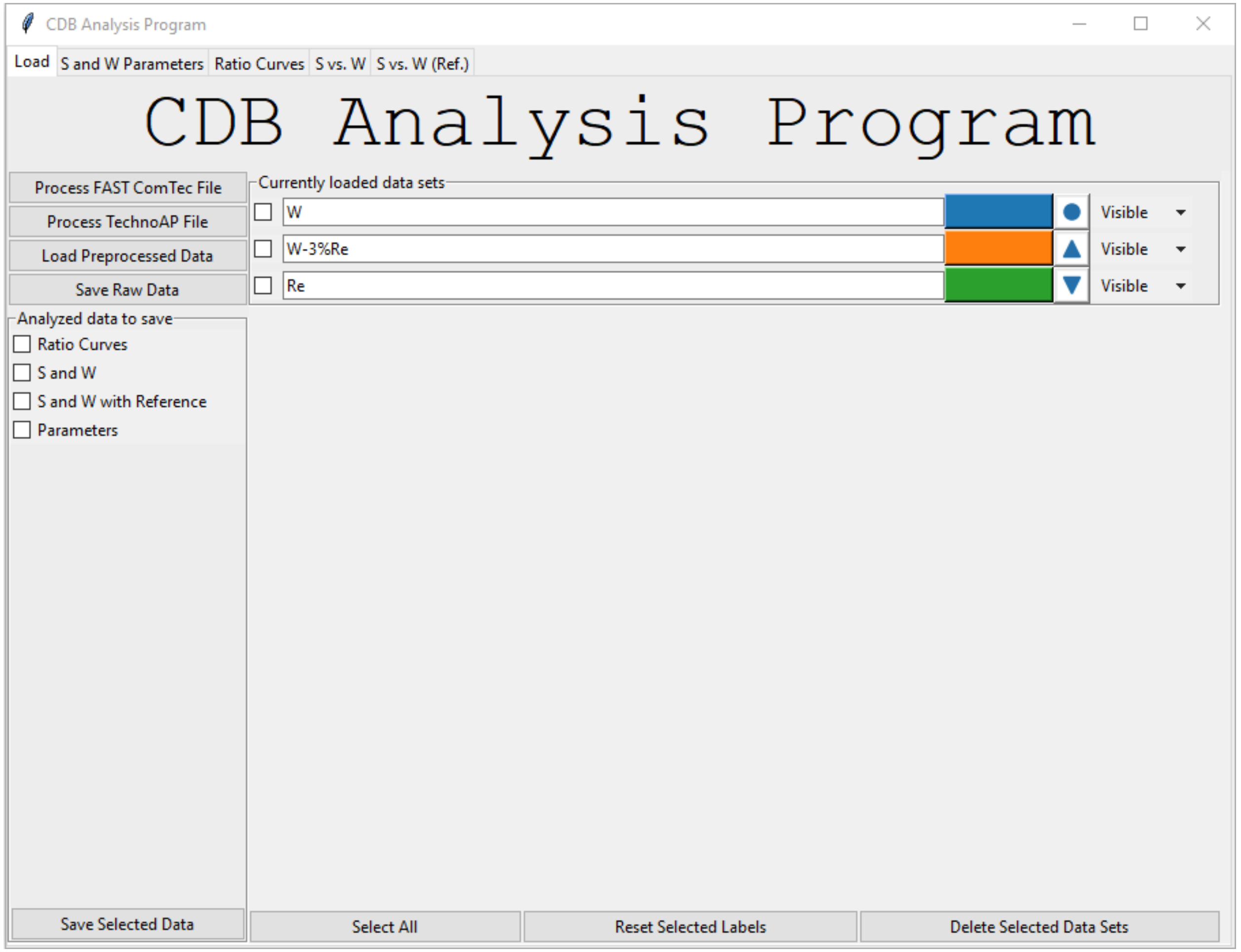


Van Huis, M. A., A. Van Veen, H. Schut, C. V. Falub, S. W. H. Eijt, P. E. Mijnarends, and J. Kuriplach. *Physical Review B* 65, no. 8 (2002): 085416.
Hakala, M., M. J. Puska and R. M. Nieminen (1998). Physical Review B 57(13): 7621.

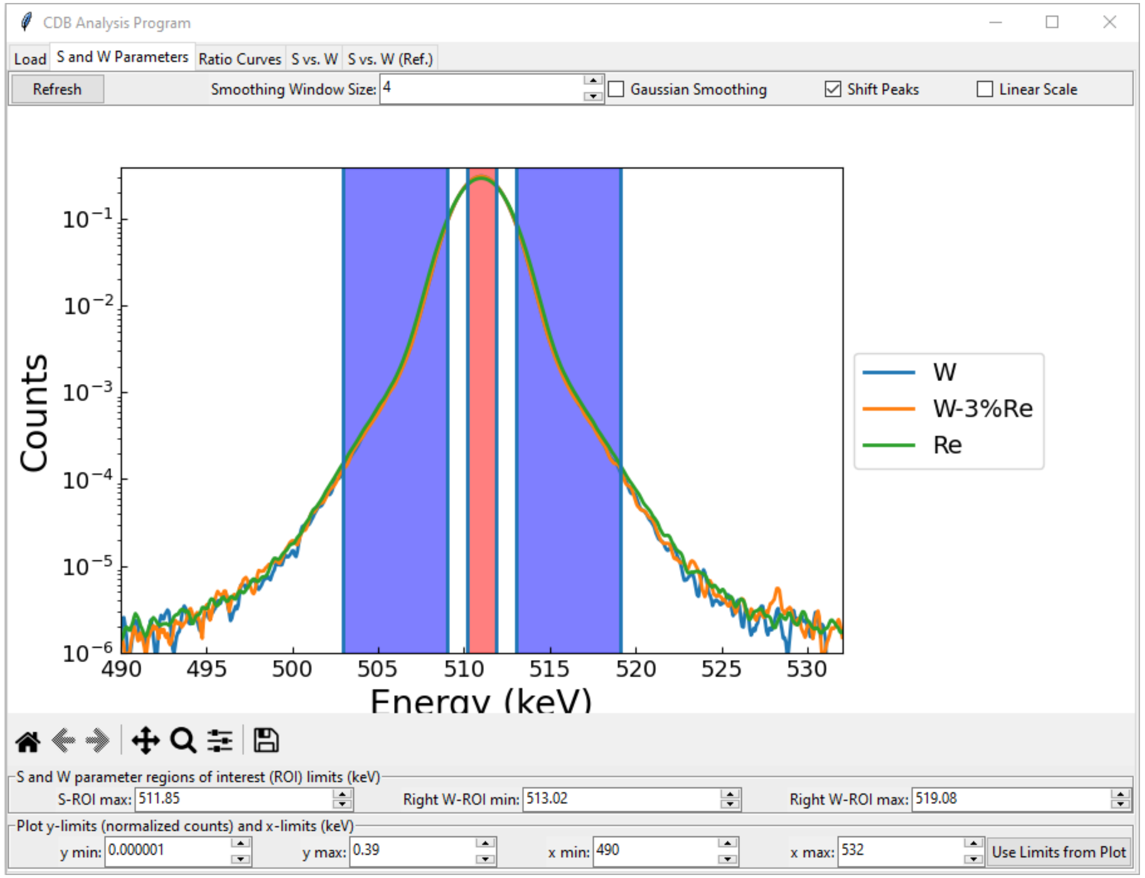
Software Development of CDB-AP

- Python based, open source, transparent
- No commercially available CDB data reduction packages
- Works for all CDB data acquired using INL PAS systems
- Rapid processing of large data sets

https://github.com/ElsevierSoftwareX/SOFTX-D-23-00218



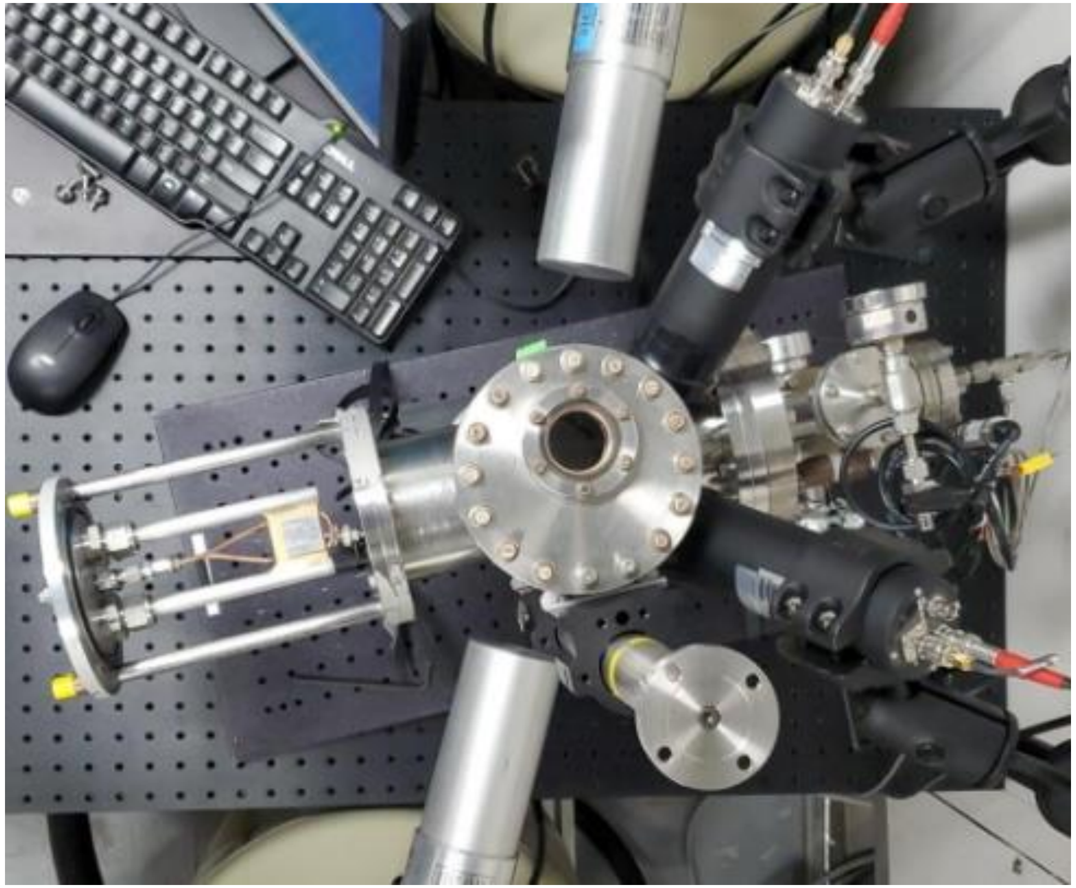
Extract S and W parameters



- Users have freedom adjusting S and W parameter calculating ROIs
- Automatic folding of 2D spectra
- Generate plots vs reference
  - Perfect material bulk parameters
  - Non irradiated materials

- **Needs improvements**
  - GUI
  - HIPPO incorporation
  - in-situ PAS on neutron beams

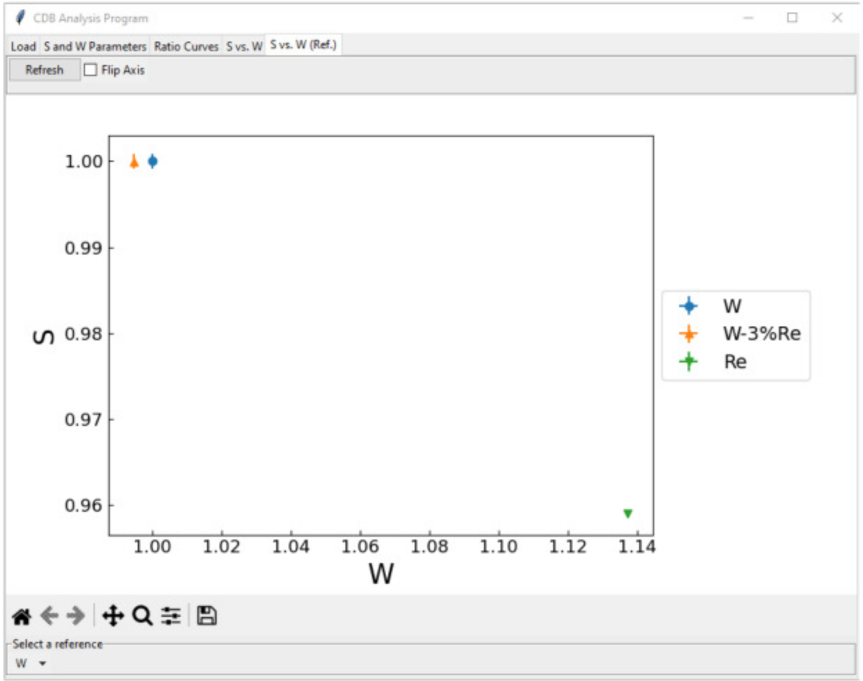
HIPPOS chamber

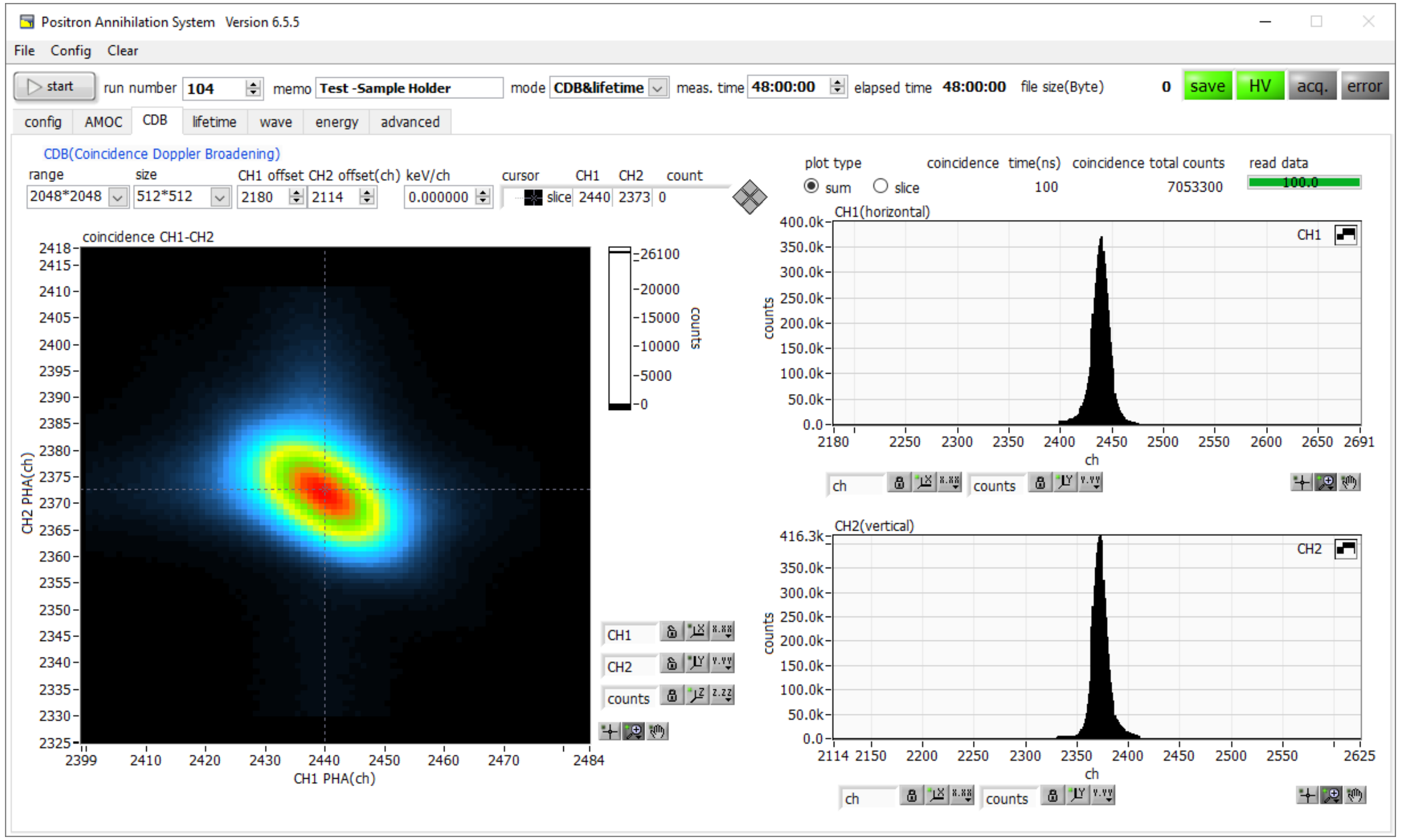


- PAS does not provide sufficient results as a standalone PIE technique
- With HIPPOS
  - In-situ annealing
  - Simultaneous PALS and CDB
- HIPPOS features
  - Dual localized cartridge heaters
  - Vacuum
  - Static inert gas
  - Compact design allows for good count rate*

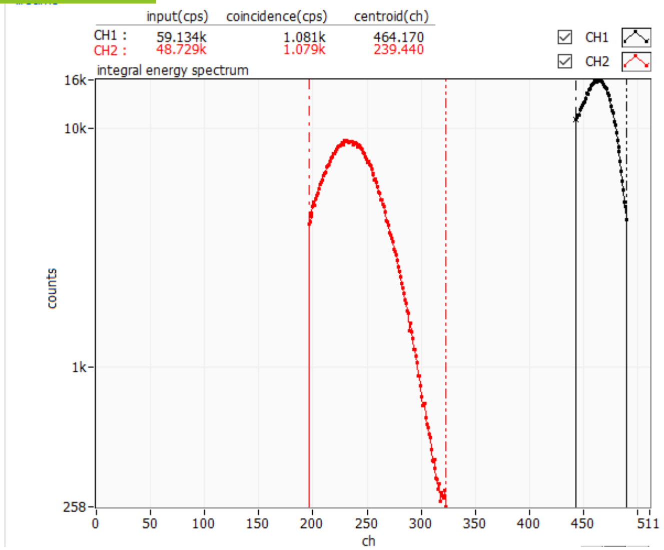
HIPPOS Chamber incorporation– CDB

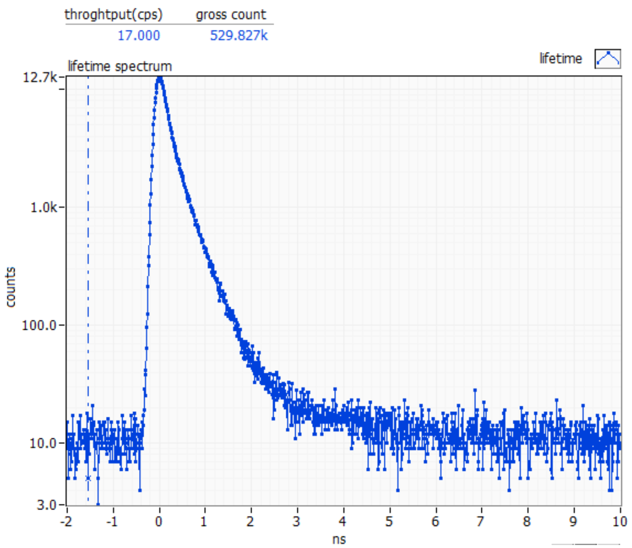
- The CDB copes well with the HIPPOS chamber
  - Raw data
- Processed CDB data would be even less impacted
  - Vs Reference (bulk parameters)



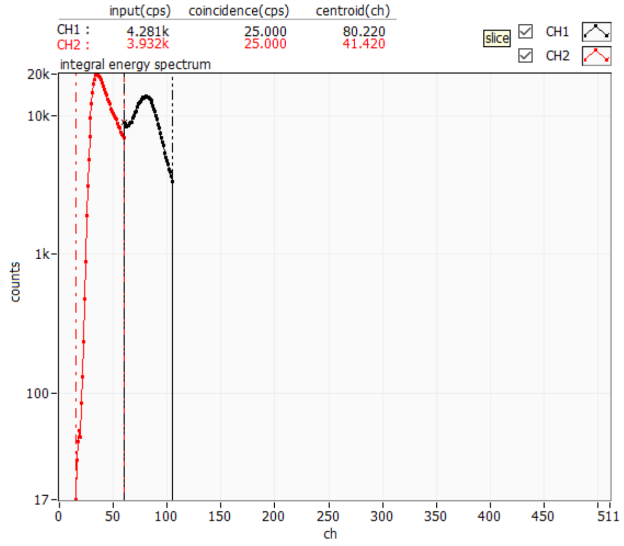


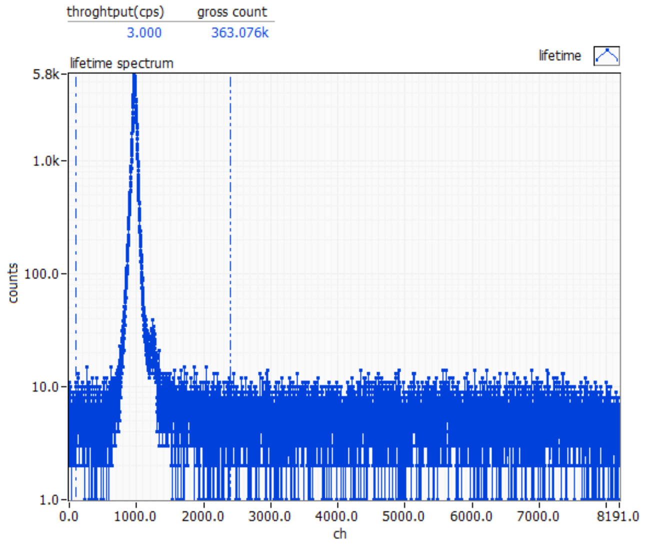
HIPPOS Chamber incorporation – PALS





Top: no HIPPOS Bottom: with HIPPOS





- Not ideal with HIPPOS
- PALS has a higher requirement of count rate
  - Count rate drops by 25%
- Stainless steel significantly attenuates 511 keV signal in BaF_2 detectors
- Lifetime spectrum shows artifacts
- Design improvements
  - Thinner walls
  - Detector slots

Battelle Energy Alliance manages INL for the U.S. Department of Energy's Office of Nuclear Energy. INL is the nation's center for nuclear energy research and development, and also performs research in each of DOE's strategic goal areas: energy, national security, science and the environment.