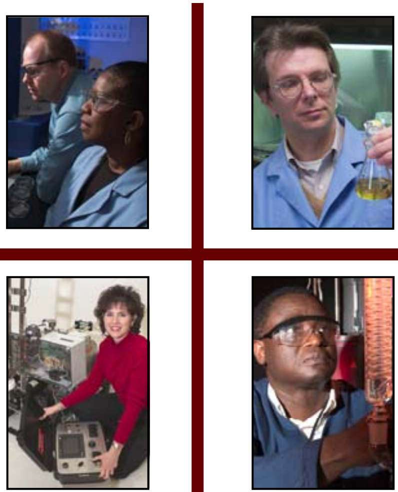
Positron Method For Detection And Measurement Of Helium-3 Bubbles



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SAVANNAH RIVER NATIONAL LABORATORY

We Put Science To Work

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INCENTIVE

- Stainless Steels applied for tritium processing and storage
  - Tritium diffuses into the steel
  - Decays to ^{3}He
  - Bubble formation
- Positron Annihilation Lifetime Spectrometry (PALS)
  - Scoping study
  - Potential for earlier detection of defect / bubble formation
  - Plant Directed R&D (PDRD) funded
- Complement previous and ongoing TEM studies

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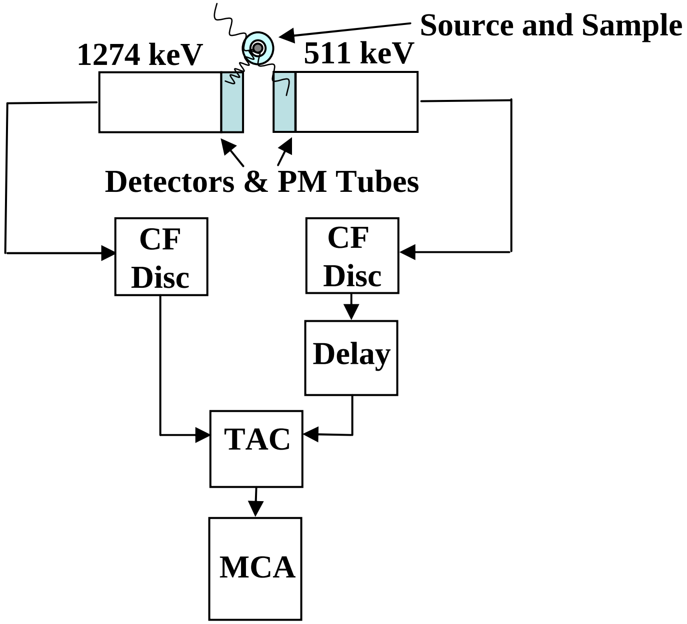
PALS Technique



- Expose material to positrons (β^+) from a ^{22}Na source
  - Sandwich source between stainless steel samples
  - Exposed surface: ~1 cm radius
- Measure time difference from decay to annihilation
  - Start signal: ^{22}Na gamma (1274 keV)
  - Stop signal: Annihilation photon (511 keV)
  - Select components for time resolution
  - “Fast-Fast” coincidence

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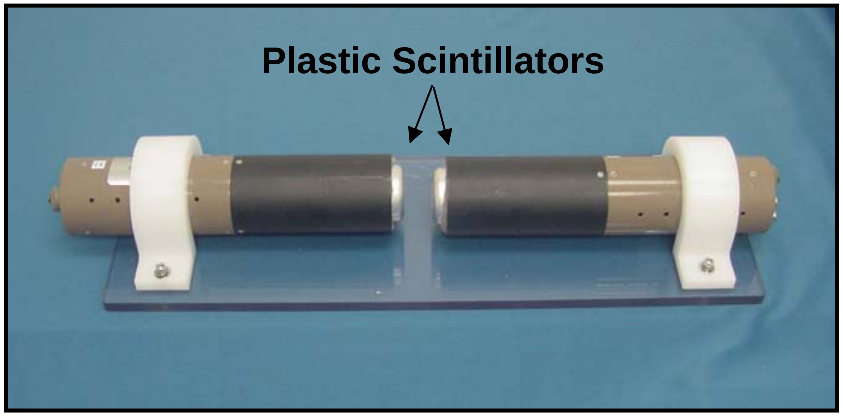
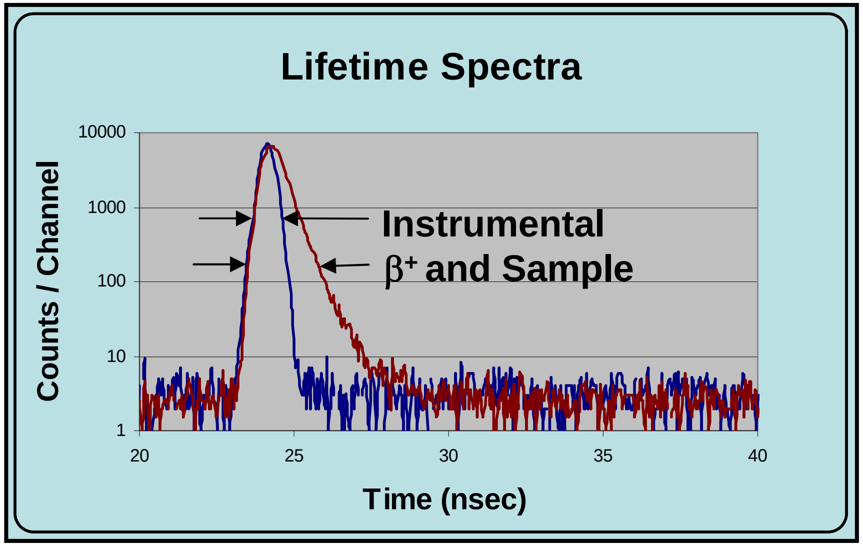
PALS Technique





- Time spectra
  - Instrumental resolution
    - ^{60}Co
      - Coincident gamma-rays
      - No β^+ diffusion and annihilation delay
    - 0.25 nsec FWHM
  - Sample and positron source
    - Broad tail
    - Thermalization
    - Diffusion
    - Trapping
    - Annihilation

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SAMPLES

- 304 and 316LN Stainless Steels
  - Control samples: No tritium exposure
  - Tritium-exposed samples
    - Tritium loaded: Pressurized, 350^{o} C, 2 weeks
    - Ingrowth: -23^{o} C, Duration: 6 to 9 months
    - Tritium removal: Vacuum, 450^{o} C, 3 weeks
  - ^{3}He Concentrations
    - Measured at PNNL
    - 304: 68.1 and 85.6 appm
    - 316LN: 65.7 and 91.3 appm

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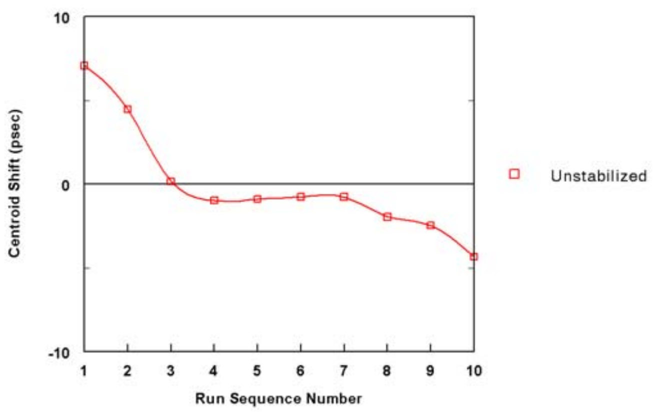
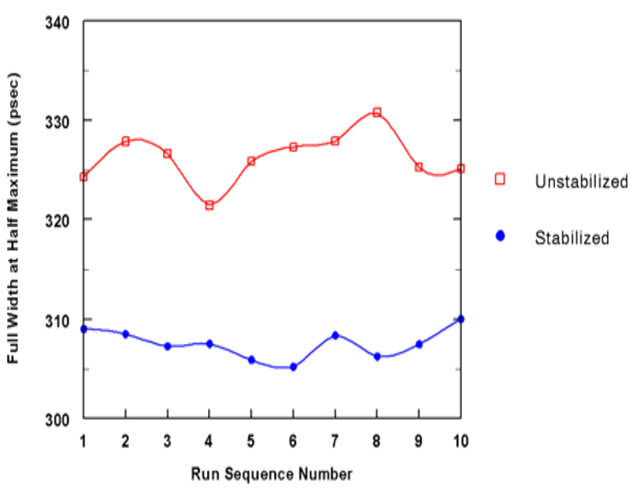
Stability





- Gain instability
  - Multiple spectra
  - Centroid shifts
- Corrected
  - Rebinned data
  - Constant centroid
  - Improved FWHM

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DATA ANALYSIS

- Spectra comprised of
  - Sum of several discrete exponential decay components
    - Sample
    - Source
  - Convolved by Gaussian broadening function
- Software
  - Lifetime 9
    - Least squares fit
      - Lifetimes
      - Intensities
      - Background
      - Zero offset channel (T_0)
    - J. Kansy, *Nucl. Instr. Meth*. A374: 235 (1996)
  - Maximum Entropy Lifetime (MELT) Method
    - Yields more consistent lifetime estimates with fewer counts
    - Input to LT9 to reduce free parameters
    - M. Shukla et al, *Nucl. Instr. Meth.* A 335: 310 (1993)

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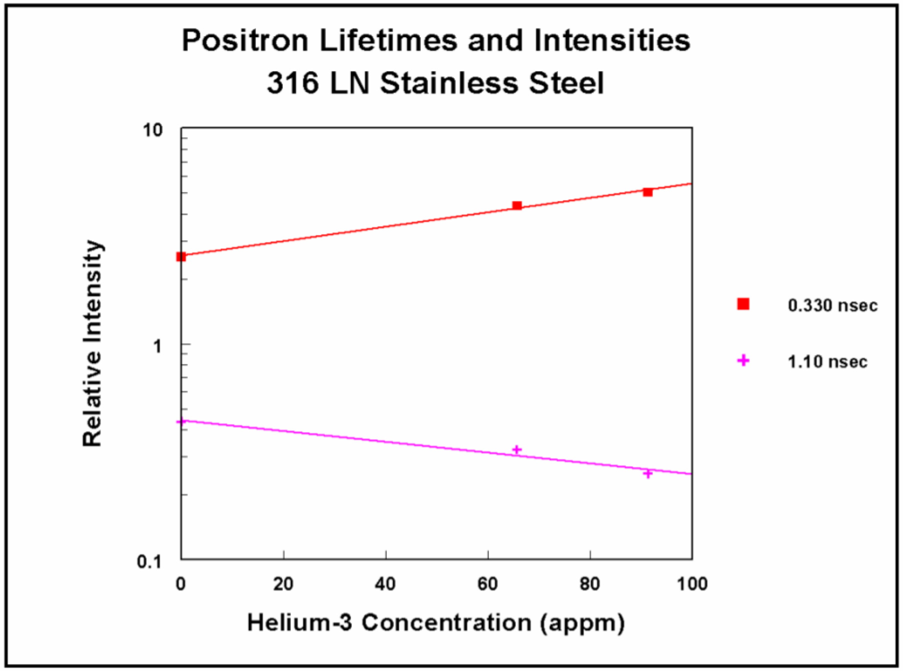
PALS Results



- PALS
  - Best for lifetimes >~ 0.050 nsec
  - Data for 0.024 nsec not shown
  - No clear changes in observed 0.165 nsec lifetime component
- Trendlines show changes
  - 0.33 and 1.10 nsec components
  - Intensities vary with ^{3}He concentration
- Relatively few samples
- Low concentration range

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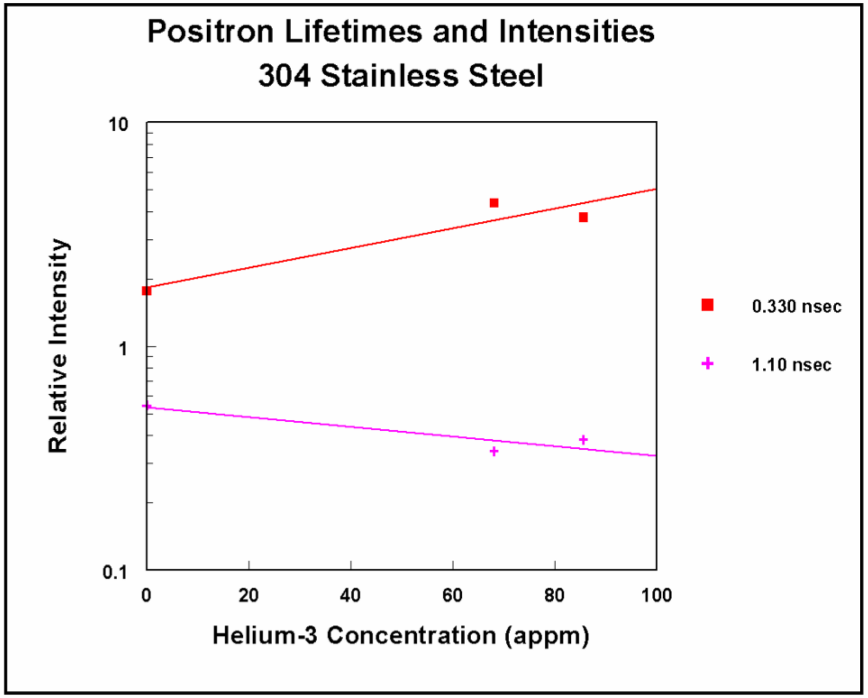
PALS Results



- Trendlines again show changes
  - 0.33 and 1.10 nsec components
  - Intensities vary with ^{3}He concentration
  - 0.33 component consistent with microvoids of ~15 vacancies
- However:
  - Relatively few samples
  - Low concentration range

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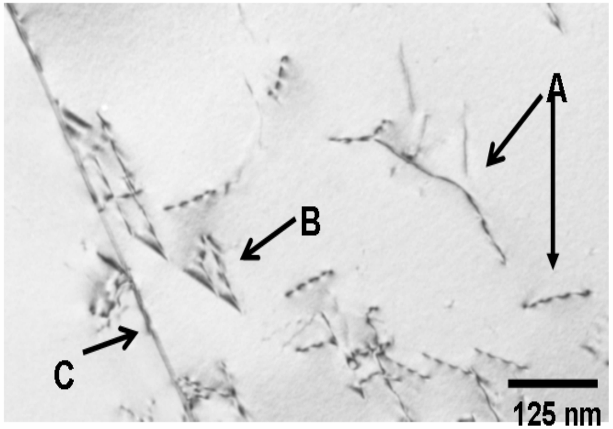
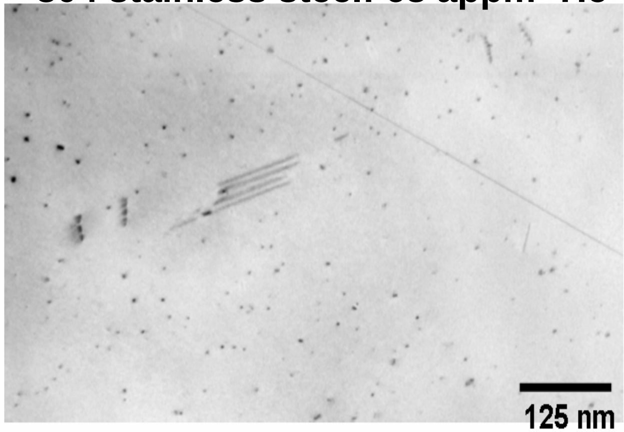
TEM Results

304 stainless steel: As received



304 stainless steel: 68 appm ^{3}He



- Analyzed helium bubble distribution
- 304 stainless steel
  - Helium bubbles observed
    - In the matrix (grain interiors)
    - On matrix dislocations: Associated with 10-20 nm diameter dislocation loops
- 316LN stainless steel
  - Surprisingly, no discernable helium bubbles were observed
  - Slight contrast variations at some dislocations may indicate possible bubble formation
  - No actual bubbles were observed

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CONCLUSIONS AND RECOMMENDATIONS

- PALS Method
  - Applied to 304 and 316LN Stainless Steels
  - Scoping study shows changes with increasing ^{3}He concentration
- Recommend
  - Larger number of samples to confirm results
  - Higher ^{3}He concentrations than those available for this study
  - Determine a "true" number density of bubbles/loops in the 304 specimens for comparison with PALS data
  - Investigate differences between PALS and TEM results on 316LN

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