Positron Method For Detection And Measurement Of Helium-3 Bubbles

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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
PALS Technique

- Expose material to positrons ($\beta^+$) 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
PALS Technique

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

- Plastic Scintillators

![Lifetime Spectra](image)
SAMPLES

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

- Gain instability
  - Multiple spectra
  - Centroid shifts

- Corrected
  - Rebinned data
  - Constant centroid
  - Improved FWHM
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 (T₀)
- Maximum Entropy Lifetime (MELT) Method
  - Yields more consistent lifetime estimates with fewer counts
  - Input to LT9 to reduce free parameters
PALS Results

- **PALS**
  - Best for lifetimes $>\sim 0.050$ 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
### 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
TEM Results

- 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
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