Tritium Effects on the Embrittlement of Stainless Steel Base Metals and Weldments

Michael Morgan, Tim Krentz, and Dale Hitchcock

Tritium Focus Group
Albuquerque, NM – October, 2018
Background

- Tritium embrittlement discovered at SRL in 1979.
- SRNL has been the sole NNSA laboratory conducting experimental R&D on the effects of tritium and decay helium on structural properties for more than 25 years.
- Early studies on permeation and mechanical properties
- Now focused on fracture mechanics behavior of alloys and fabrication processes.
- Supported by Enhanced Surveillance (Aging and Lifetimes) (~$800K/yr).

Tritium Autoradiography Reveals Depth of Penetration

Source: SRL Notebook
Dave Rawl, 11/7/79
**Aging and Lifetimes - Program Goal**

**Goal:**
- Develop an understanding of tritium embrittlement and, at the same time, provide required data for establishing and extending tritium reservoir lifetimes.

**Current Efforts and Presentation Outline:**
- Fracture toughness properties of actual reservoir forgings and microstructures
- Forging process effects on tritium compatibility
- Weldments and heat-affected zones
- Additive Manufactured alloys
- New characterization techniques and modeling
Fracture Mechanics Approach

Measure fracture toughness properties of base metals, weldments and heat-affected zones after tritium exposure.

Tritium Induced Cracking in Base Metal and Weld Heat Affected Zone

Reservoir Model Stress Distribution
Experimental Procedure

Fabricate samples From Forgings

Hydrogen/Tritium Exposure at 350C and 5000 psi – Age at -50 C to Build-in Helium W/O Losing Tritium

Test In Hood For Containment of Tritium Off-gassing
Tritium Exposures and Aging

- Expose to tritium gas at 35 MPa and 350°C for two weeks.
- Age to ~1000 appm He.
- 350°C is high enough for to saturate samples with tritium but low enough to minimize any change in microstructure.
- Aging at low temperature minimizes off-gassing losses.
Load, Displacement and Crack Length Monitored During Mechanical Testing

Fracture Appearance for Hydrogen-Charged Samples

Note - There is a higher density of microvoids on the fracture surface for hydrogen-charged samples than for non-charged samples suggesting that hydrogen makes void nucleation easier.
Typical Results – Rising Load Tests

- J-Integral fracture toughness properties are decreased with increasing hydrogen exposure pressures.
- Properties decrease further with aging from build in of decay helium.
Effect of Decay Helium on Base Metal Toughness (up to 18 years of Aging)

Decay Helium Bubbles Observed on Grain Boundary after Plastic Deformation
Effect of Decay Helium on Weldment Toughness (up to 18 years of Aging)

Welds are Tougher than Base Metals Before Exposure; Similar Toughness After Exposure

Weldments of the Two Steels Had Similar Low Toughness Properties After Aging
Fracture Toughness Properties of Actual Forgings

Average Fracture Toughness Values
Stem, Cup, & Block Forgings

<table>
<thead>
<tr>
<th>Type</th>
<th>Value, kJ/m²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stem Type 316L SS</td>
<td>Not Charged 1300, H₂ Precharged 1000, T₂-Charged (600 ppm He) 700</td>
</tr>
<tr>
<td>Cup Type 316L SS</td>
<td>Not Charged 1100, H₂ Precharged 900, T₂-Charged (600 ppm He) 500</td>
</tr>
<tr>
<td>Block Type 304L SS (Low YS)</td>
<td>Not Charged 2200, H₂ Precharged 1800, T₂-Charged (600 ppm He) 1400</td>
</tr>
<tr>
<td>Block Type 304L SS (High YS)</td>
<td>Not Charged 2500, H₂ Precharged 2100, T₂-Charged (600 ppm He) 1700</td>
</tr>
</tbody>
</table>

Stem Forging
Cup Forging
Block Forging
Fracture Appearance - Type 304L SS Block Forging

Not-Charged
60RA1

Not-Charged
60RA1

H2-Charged
60RA9

H2-Charged
60RA9
Forging Process Effects: Strain Rate and Temperatures

Specimens Fabricated from Remnants of Multi-Stage Forging Processes

<table>
<thead>
<tr>
<th>Final Forging Process</th>
<th>Approximate Forging Velocity (mm/s)</th>
<th>Deformation Time (s)</th>
<th>Engineering Strain Rate (s^-1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydraulic Press</td>
<td>60</td>
<td>0.4</td>
<td>1</td>
</tr>
<tr>
<td>Mech. Press</td>
<td>300</td>
<td>0.08</td>
<td>5</td>
</tr>
<tr>
<td>Screw Press</td>
<td>575</td>
<td>0.04</td>
<td>10</td>
</tr>
<tr>
<td>Dynapak HERF</td>
<td>6500-7500</td>
<td>0.004</td>
<td>100</td>
</tr>
</tbody>
</table>

Screw Press—Type 304L Stainless Steel
Forging Process Effect

Tritium-precharging produced a similar reduction in the fracture toughness values for all of the forging conditions.
Tritium Effects on Welds and Heat Affected Zones

• Joint effort with Sandia National Laboratory (Joe Ronevich)
• Two steels – Types 304L and 21-6-9 SS
• Cracks in Fusion Zone and HAZ
• SNL – vessel fabrication; specimen machining, control tests.
• SRNL – specimen pre-cracking, tritium charging & testing.

HAZ Cracking After Long-Term Exposure of Test Reservoir
Hydrogen Effects on Additive Manufactured Stainless Steel

Hydrogen Effect on Toughness Properties

Specimens Exposed to Hydrogen Gas at 350°C and 17 Mpa and Tested in 3-Pt Bending
Characterization Techniques – TEM Used for Revealing Helium Bubbles

TEM Discs Cut, Punched, and Thinned From Near Fracture Region

Nanometer sized bubbles randomly distributed and clustered on defects
Decay Helium Bubbles Difficult to Resolve in Forged Steels

Helium Bubbles Resolved in Austenite of Weldment But Not in Ferrite

Deformation Twinning in Tritium-Aged Forged Steel Helium Bubbles Not Resolved
Small Angle Neutron Scattering (Dale Hitchcock, Tim Krentz, & Ken Imrich)

- Small angle neutron scattering (SANS) is being used to probe tritium exposed steels.
- The purpose is to learn as much as possible about decay helium bubble size, spacing and distribution.
- Complement TEM observations for forged microstructure since nanometer-sized bubbles are not easily be resolved.

Neutron scattering measurements were performed on tritium exposed stainless steel samples on beamline CG-2 at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Lab (ORNL).
Small Angle Neutron Scattering

**Samples**

<table>
<thead>
<tr>
<th>Samples</th>
<th>Charging</th>
<th>Age</th>
</tr>
</thead>
<tbody>
<tr>
<td>304L/308 filled steel weldment</td>
<td>T2</td>
<td>17-year</td>
</tr>
<tr>
<td>high energy rate forged 304L</td>
<td>T2</td>
<td>3-year</td>
</tr>
<tr>
<td>steel</td>
<td></td>
<td></td>
</tr>
<tr>
<td>304L/308 filled steel weldment</td>
<td>H2 (5000 psi)</td>
<td>N/A</td>
</tr>
<tr>
<td>304L/308 filled steel weldment</td>
<td>H2 (10000 psi)</td>
<td>N/A</td>
</tr>
<tr>
<td>304L/308 filled steel weldment</td>
<td>Uncharged</td>
<td>N/A</td>
</tr>
<tr>
<td>high energy rate forged 304L</td>
<td>uncharged</td>
<td>N/A</td>
</tr>
<tr>
<td>steel</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Neutron scattering measurements were performed on tritium exposed stainless steel samples on beamline CG-2 at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Lab (ORNL).

Small angle neutron scattering (SANS) measurements were performed to elucidate the nature of Helium bubble formation in the matrix, and small angle incoherent neutron scattering (SAINS) measurements were performed to measure the hydrogen content in the samples.
Hydrogen charged weld – fit with Gunier-Porod model (incoherent background 1 order of magnitude higher than tritium – matches decay + T vs H cross sections)

Tritium charged weld – fit with hard sphere model (Rg (ave. bubble size) =2.3nm, bubble pressures ~ 3GPa)
Can models be built using existing data and/or short-term tests that predict performance after many years in tritium service?
Summary and Conclusions

- Fracture mechanics properties of stainless steels are being measured after hydrogen and tritium exposures.
- Included are new fracture toughness results after 18 years of aging.
- Type 21-6-9 steels and weldments showed complete brittle fracture and greatly reduced toughness values to just 2-5% of the original values.
- Results and program plans are transmitted to design agencies in annual technical reports.
- Programs are underway to measure toughness in actual reservoir forgings, weldments and HAZs, as well as Additive Manufactured alloys.
- New characterization techniques are being explored and fracture models developed to gain better understanding and predictive capabilities for hydrogen and helium embrittlement.
In addition to the investigators mentioned above and others I failed to mention, I am grateful to the following key contributors:

- **Jim Wilderman, Stephen Crossland, and Glenn Chapman** for safely conducting mechanical testing, high-pressure hydrogen charging, and radioactive tritium testing.
- **Ken Imrich** for electric discharge machining of clean and tritium-contaminated specimens and for transferring tritium-exposed specimens from the loading line to SRNL.
- **Calvin Clamp and Chad Sweeney** – tritium charging engineering and charging vessel tests, etc.
- **John McIntosh** for data acquisition and potential drop system
- **Greg Creech & Jim Wilderman** – Material Control and Accountability of tritium specimens.
- **Henry Ajo** for fractography.