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National Nuclear Security Administration
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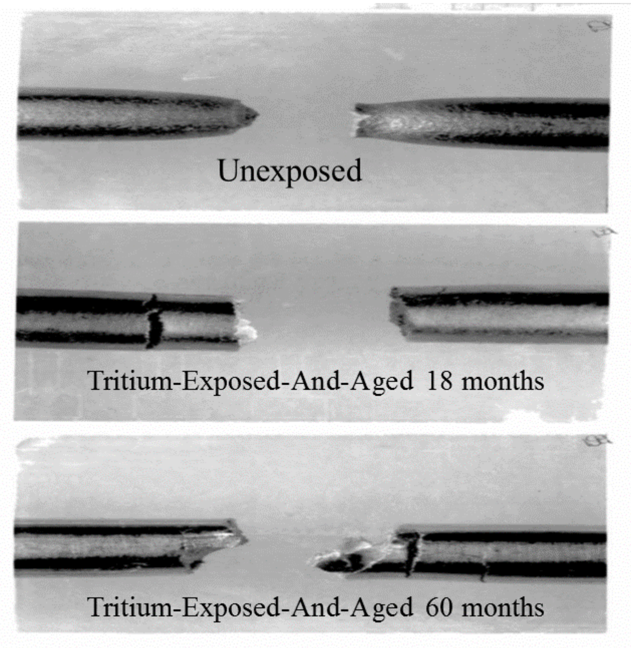
Tritium Effects on the Embrittlement of Stainless Steel Base Metals and Weldments

Michael Morgan, Tim Krentz, and Dale Hitchcock

Tritium Focus Group
Albuquerque, NM – October, 2018

SRNL
SAVANNAH RIVER NATIONAL LABORATORY

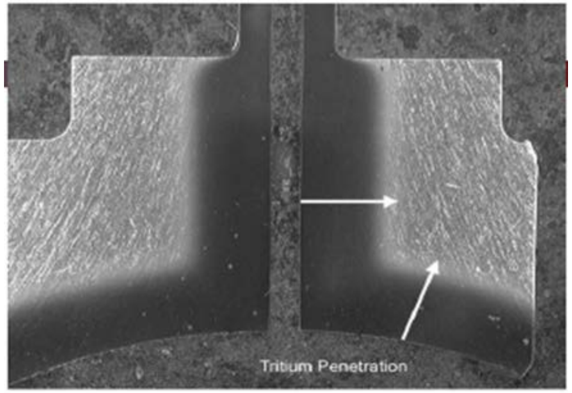
Background

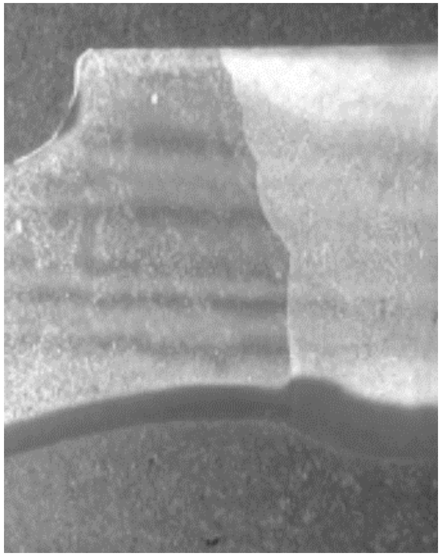


Source: SRL Notebook
Dave Rawl, 11/7/79

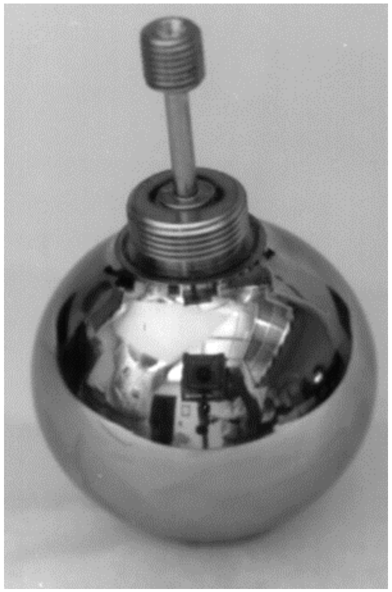
- Tritium embrittlement discovered at SRL in 1979.
- SRNL has been the sole NNSA laboratory conducting experimental R&D on the effects of tritium and decay helium on structural properties for more than 25 years.
- Early studies on permeation and mechanical properties
- Now focused on fracture mechanics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

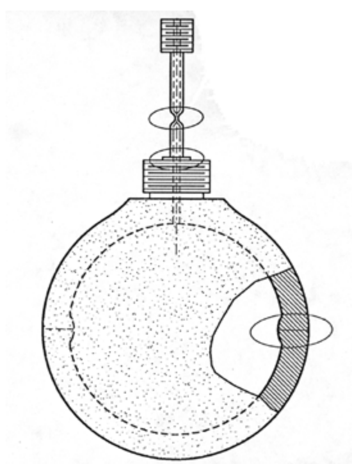
Tritium Autoradiography
Reveals Depth of Penetration





Aging and Lifetimes - Program Goal





Goal:

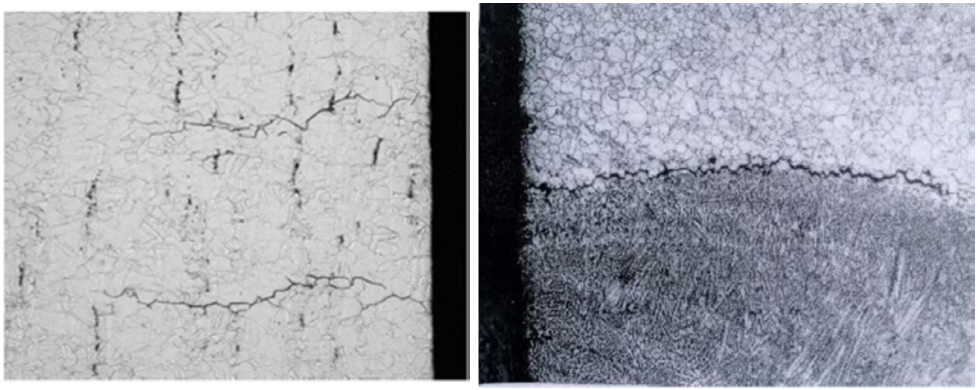
- Develop an understanding of tritium embrittlement and, at the same time, provide required data for establishing and extending tritium reservoir lifetimes.

Current Efforts and Presentation Outline:

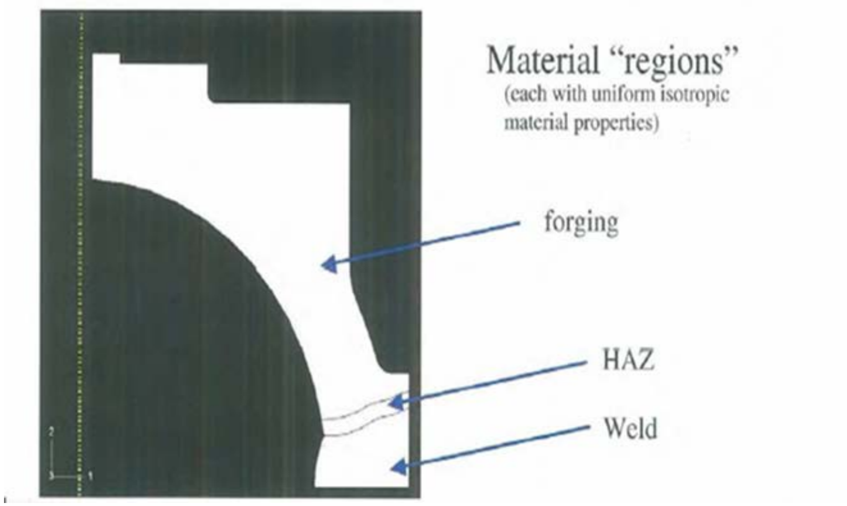
- – Fracture toughness properties of actual reservoir forgings and microstructures
- – Forging process effects on tritium compatibility
- – Weldments and heat-affected zones
- – Additive Manufactured alloys
- – New characterization techniques and modeling

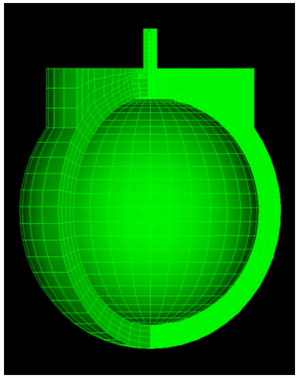
Fracture Mechanics Approach

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

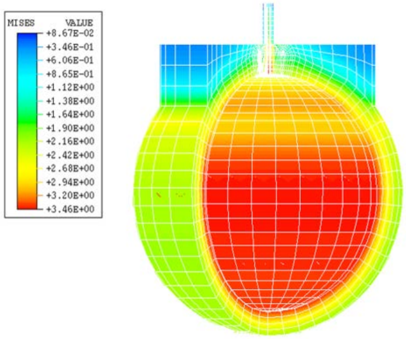


Tritium Induced Cracking in Base Metal and Weld Heat Affected Zone





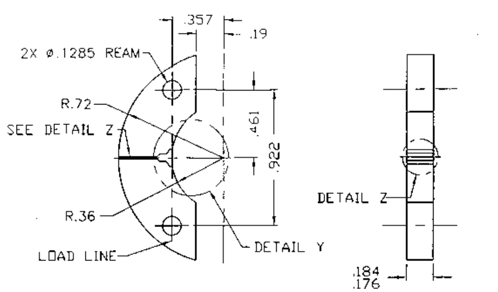
Reservoir Model



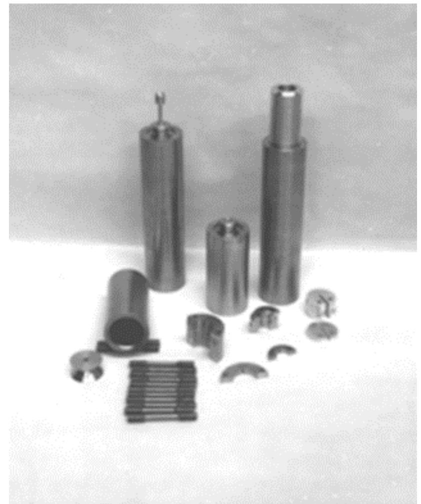
Stress Distribution

Experimental Procedure

Fabricate samples
From Forgings



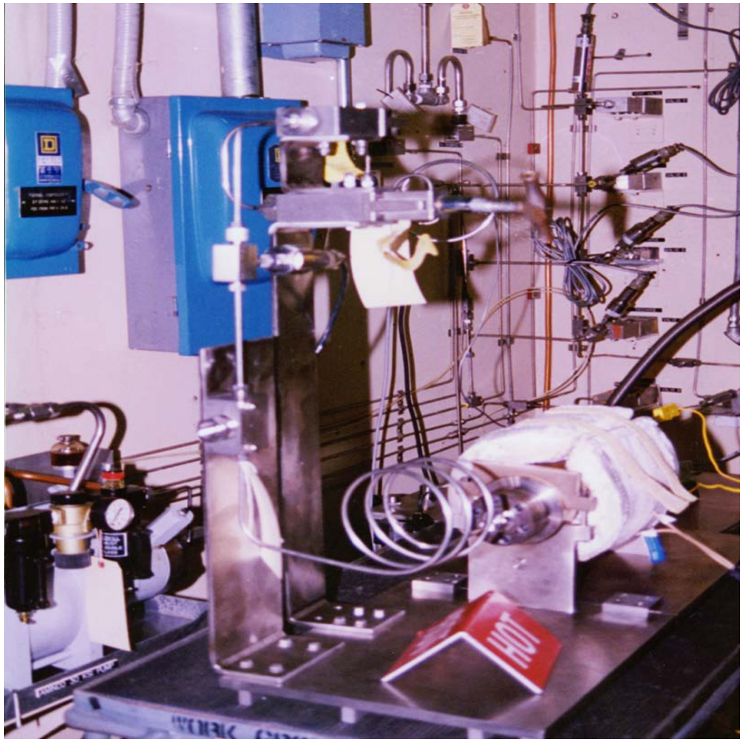
ARC–SHAPE FRACTURE TOUGHNESS SAMPLE

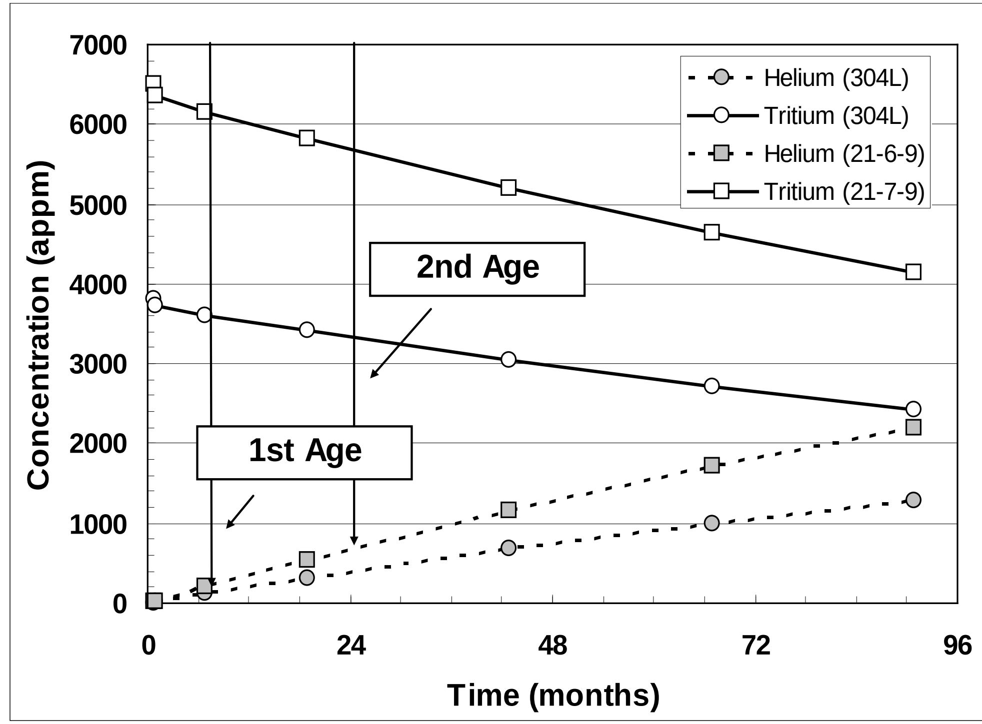


Hydrogen/Tritium
Exposure at 350C
and 5000 psi – Age
at -50 C to Build-in
Helium W/O Losing
Tritium

Test In Hood For
Containment of
Tritium Off-gassing

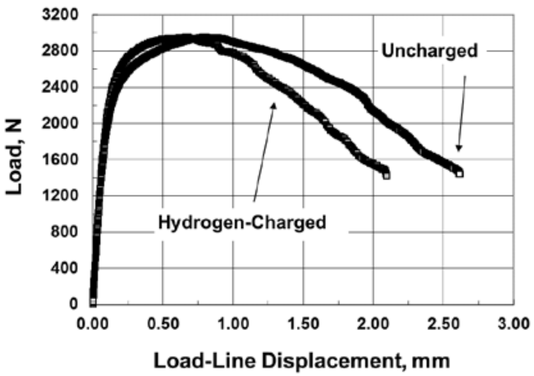
Tritium Exposures and Aging

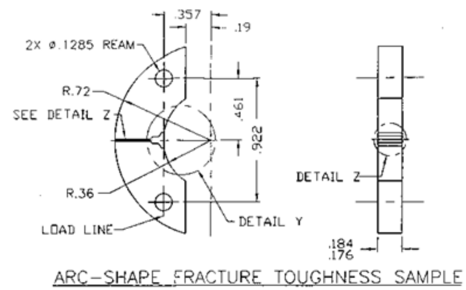


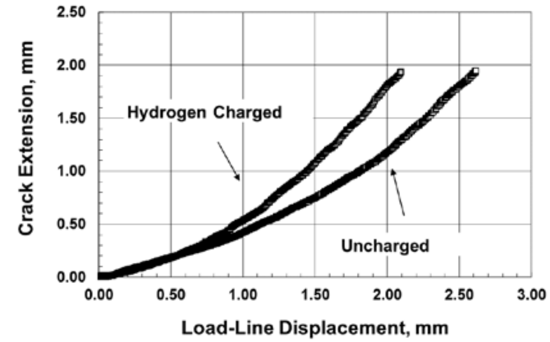


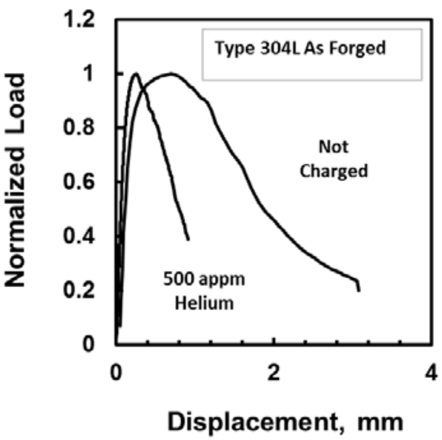
- Expose to tritium gas at 35 MPa and 350°C for two weeks.
- Age to ~1000 appm He.
- 350°C is high enough for to saturate samples with tritium but low enough to minimize any change in microstructure.
- Aging at low temperature minimizes off-gassing losses.

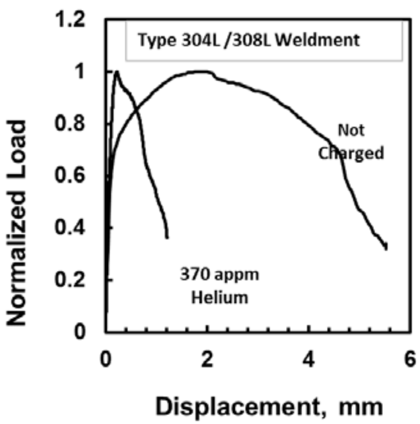
Load, Displacement and Crack Length Monitored During Mechanical Testing



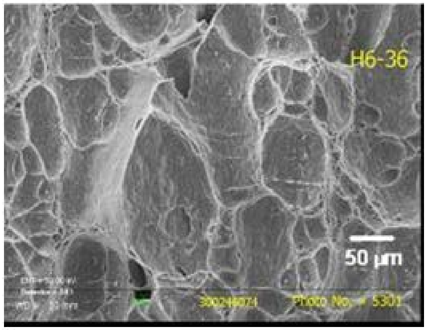




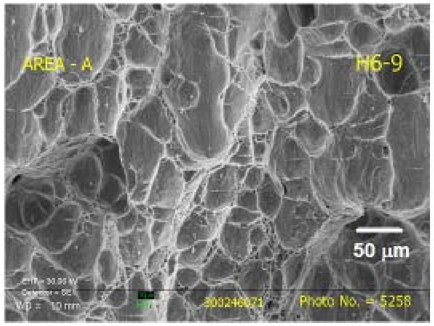




Fracture Appearance for Hydrogen-Charged Samples

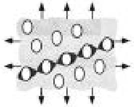


Non-Charged

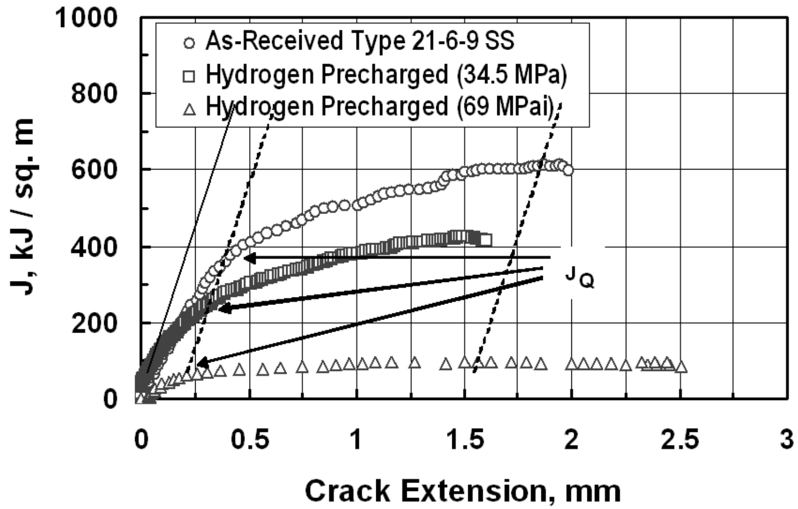


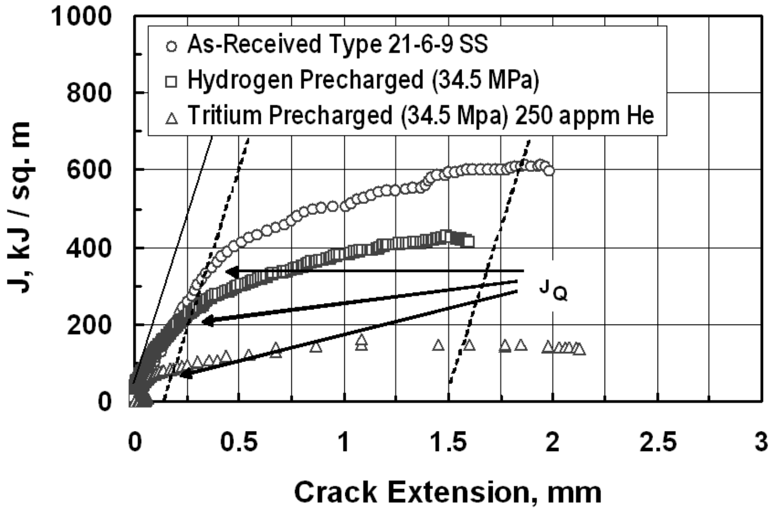
Hydrogen-Charged

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.

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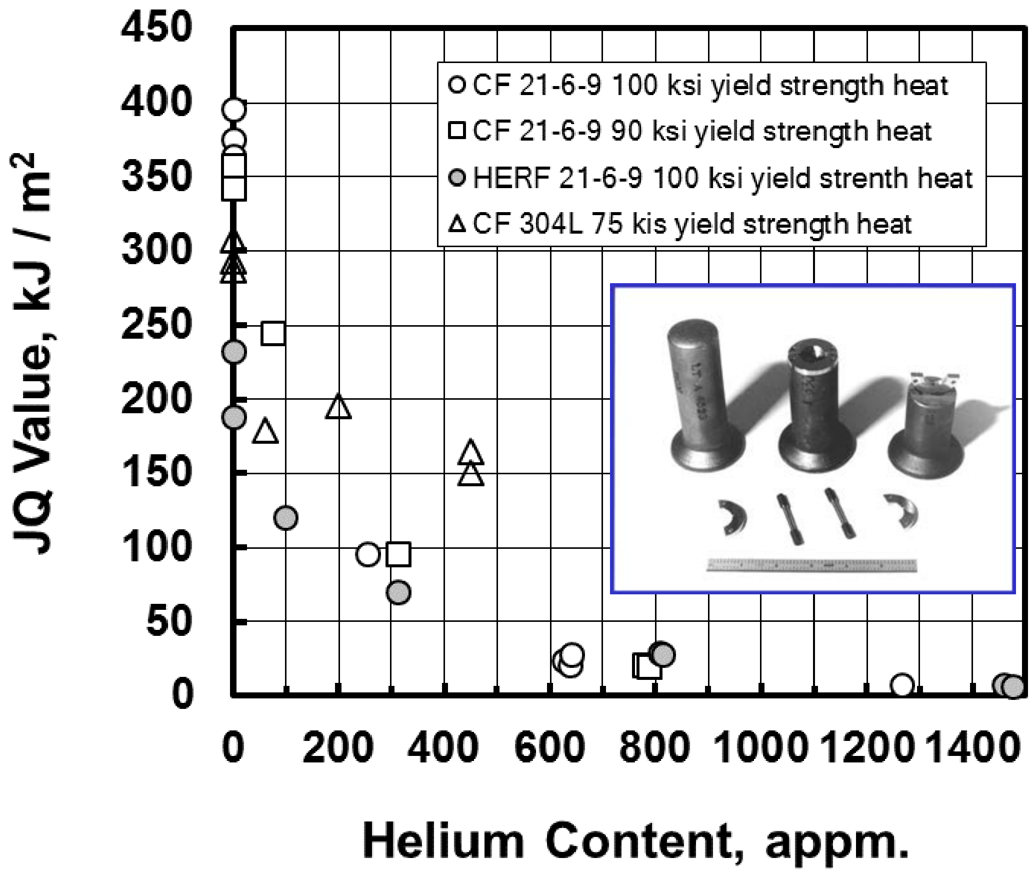
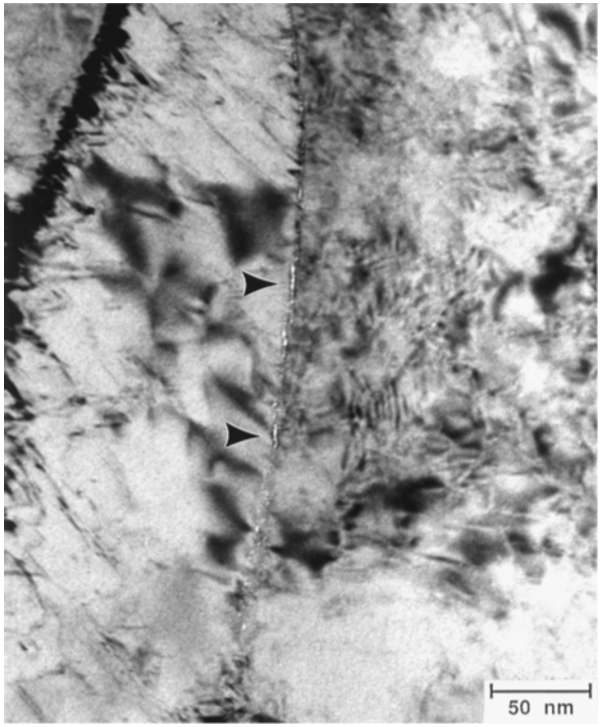
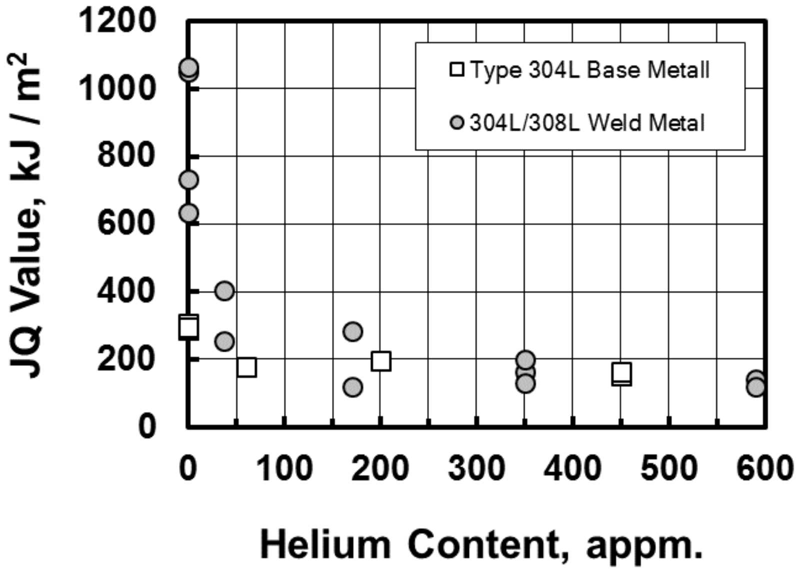
Effect of Decay Helium on Base Metal Toughness (up to 18 years of Aging)

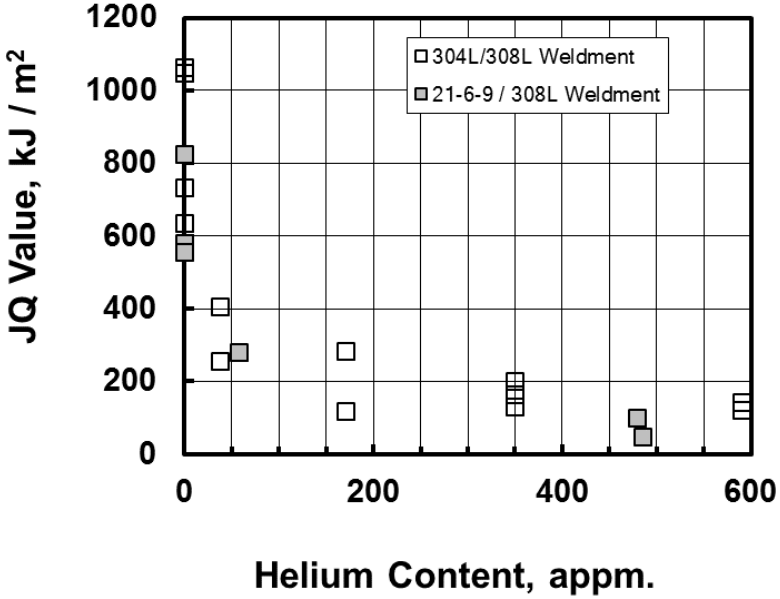




Decay Helium Bubbles Observed on Grain Boundary after Plastic Deformation

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

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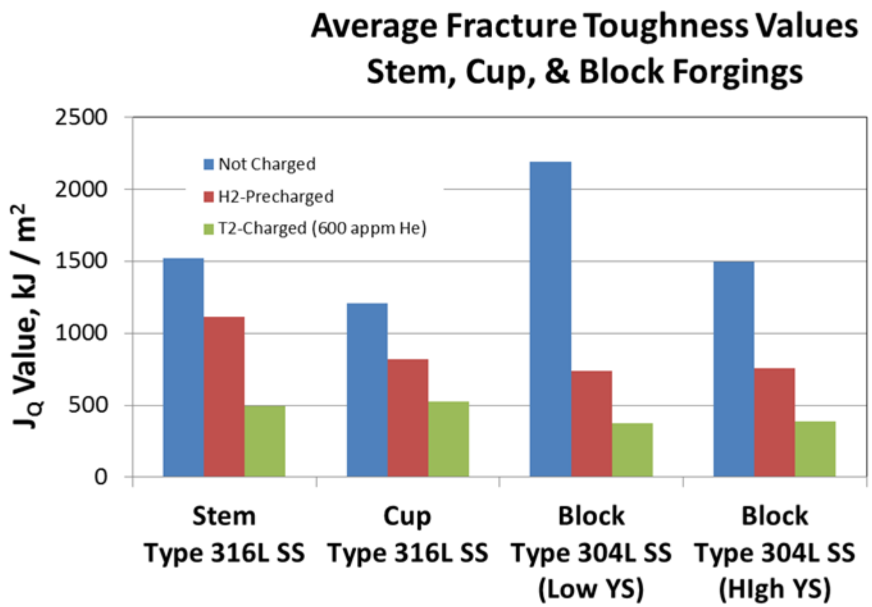
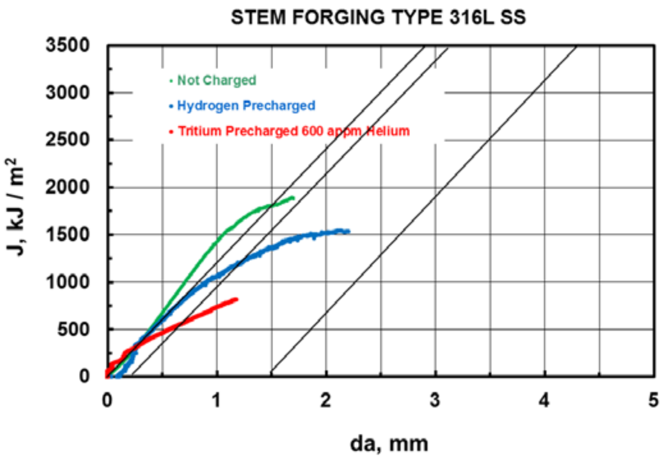
Fracture Toughness Properties of Actual Forgings

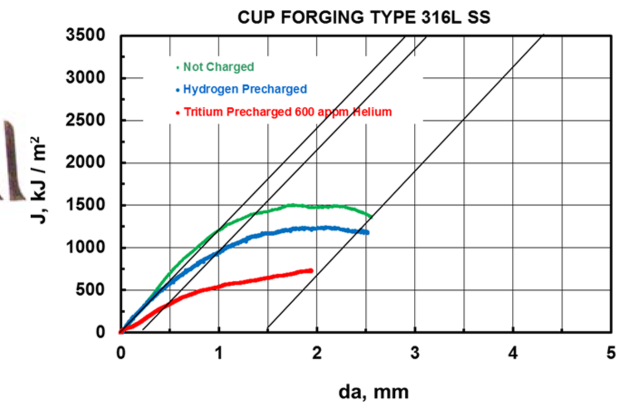
Stem Forging



Block Forging



Cup Forging



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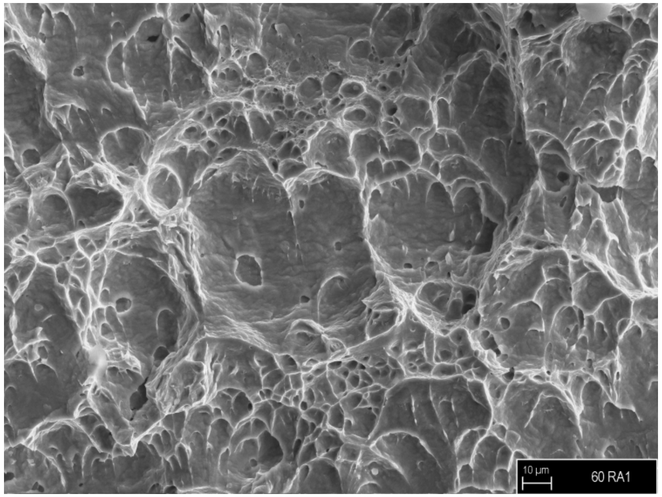
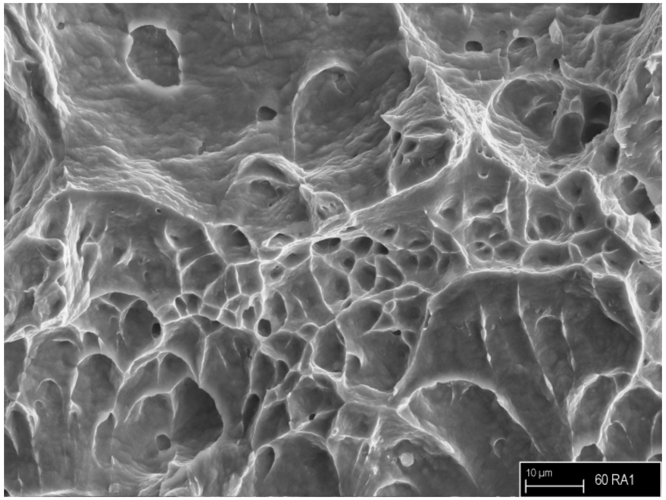
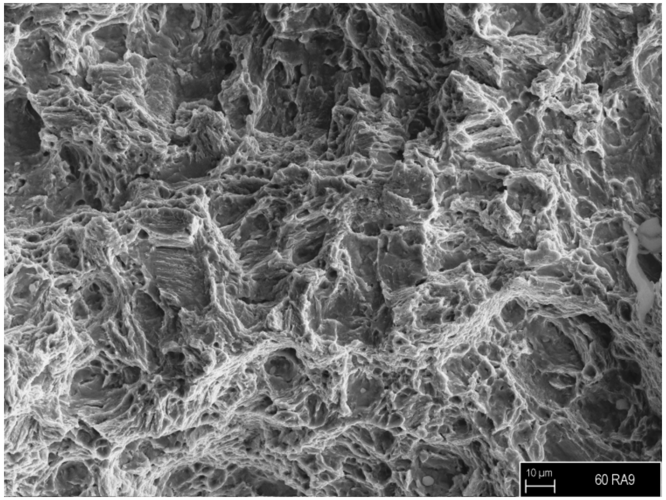
Fracture Appearance - Type 304L SS Block Forging

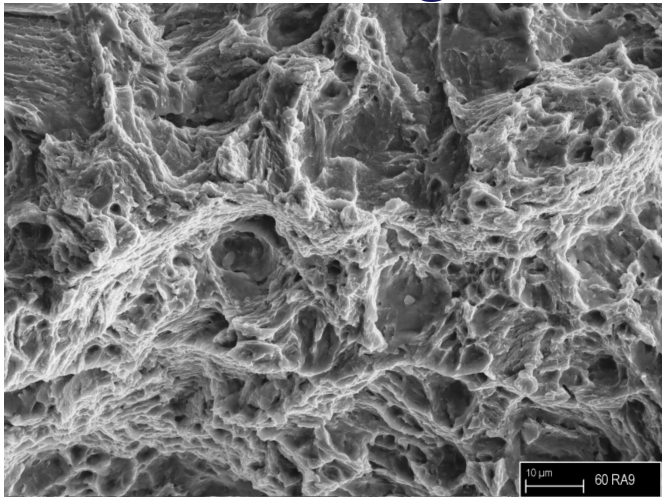




Not-Charged

Not-Charged

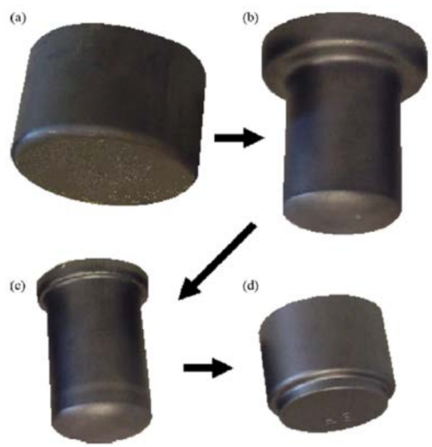




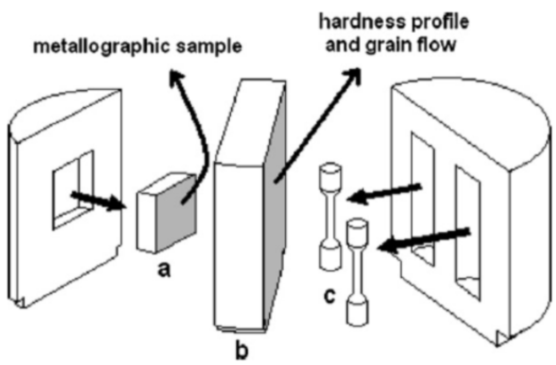
H2-Charged
60RA9

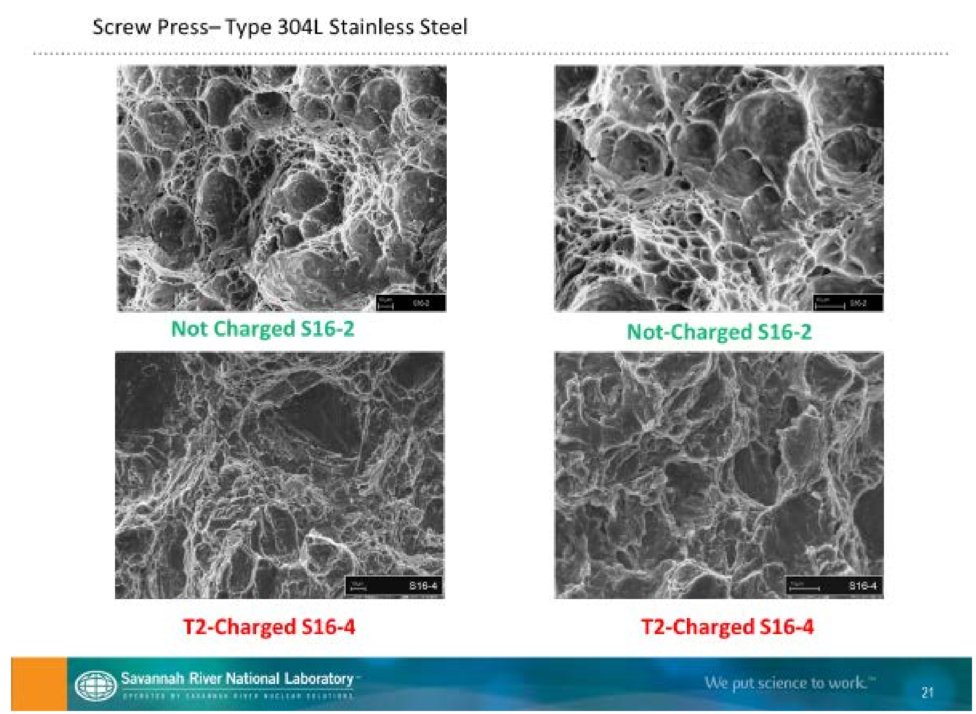
H2-Charged
60RA9

Forging Process Effects: Strain Rate and Temperatures



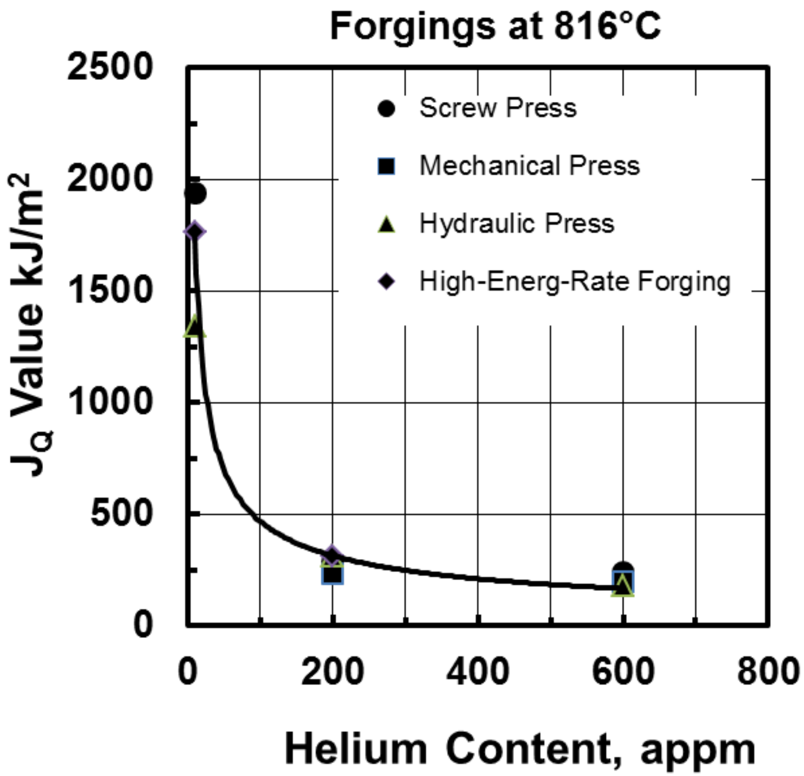
Final Forging Process	Approximate Forging Velocity (mm/s)	Deformation Time (s)	Engineering Strain Rate (s-1)
Hydraulic Press	60	0.4	1
Mech. Press	300	0.08	5
Screw Press	575	0.04	10
Dynapak HERF	6500-7500	0.004	100

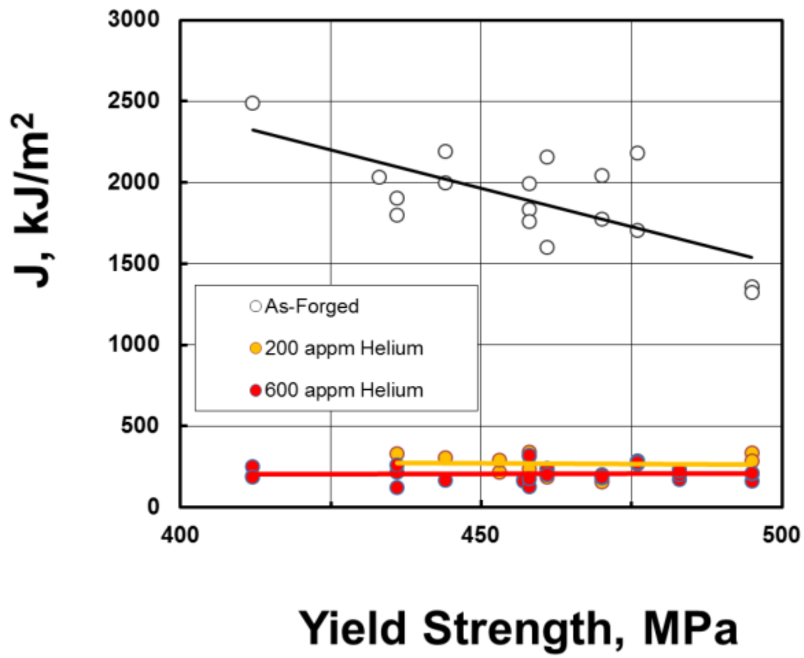




Specimens Fabricated from Remnants of Multi-Stage Forging Processes

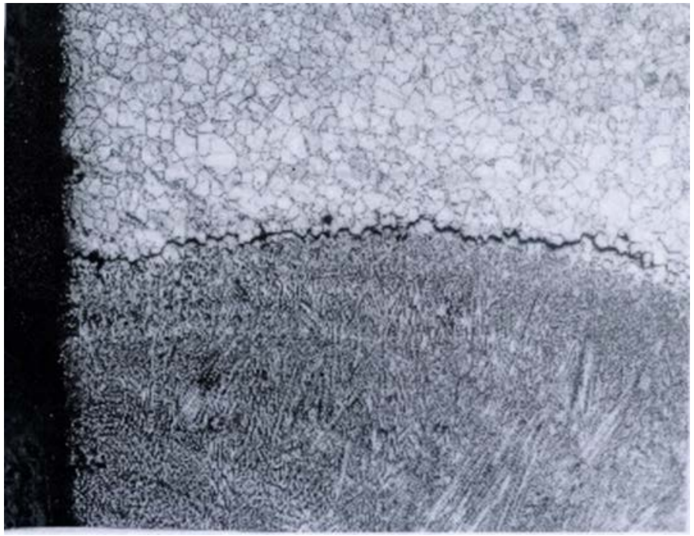
Forging Process Effect



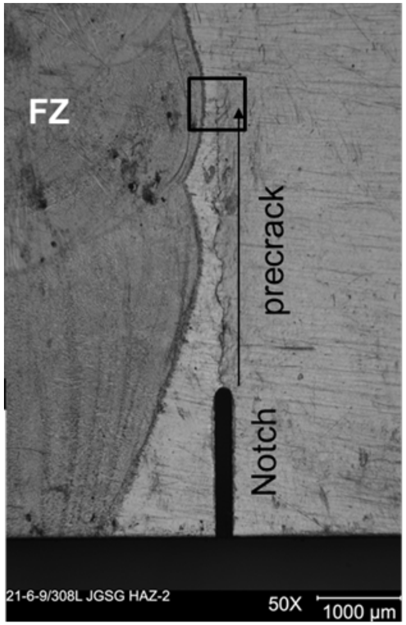


Tritium-precharging produced a similar reduction in the fracture toughness values for all of the forging conditions.

Tritium Effects on Welds and Heat Affected Zones



HAZ Cracking After Long-Term Exposure of Test Reservoir



- 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.

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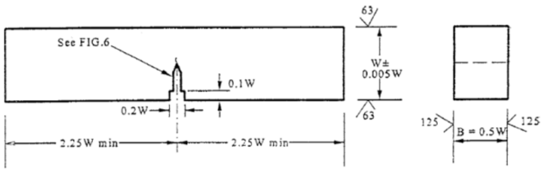
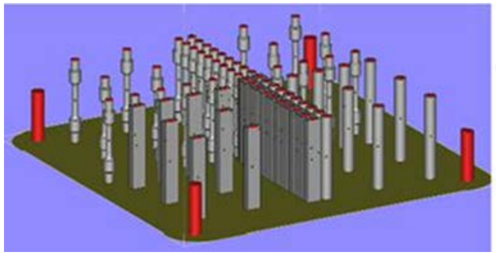
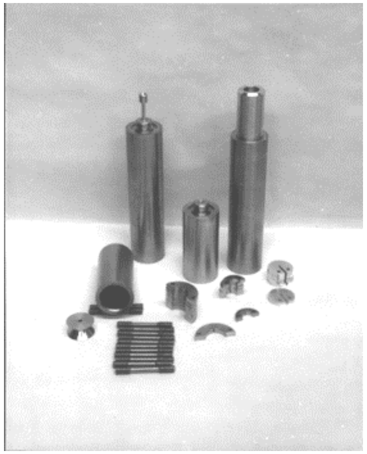
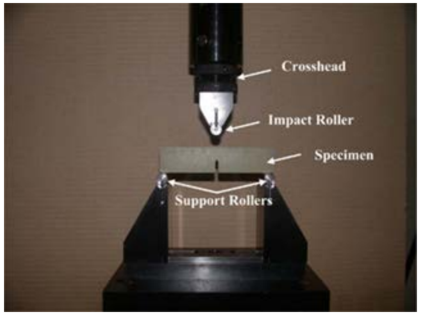
Hydrogen Effects on Additive Manufactured Stainless Steel

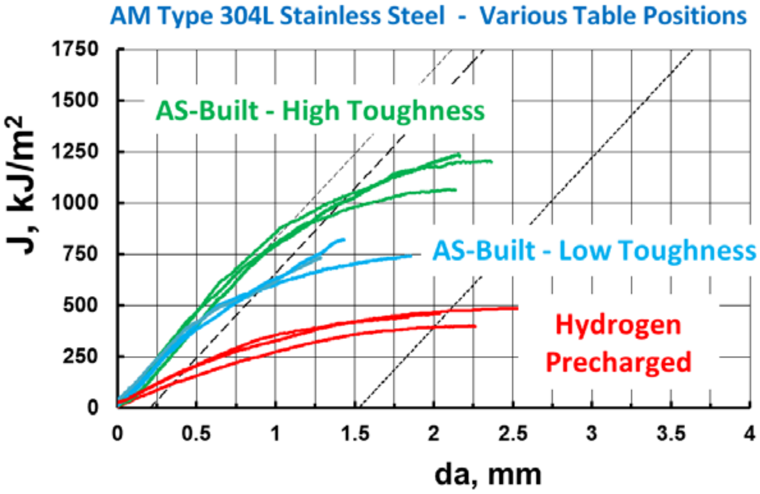






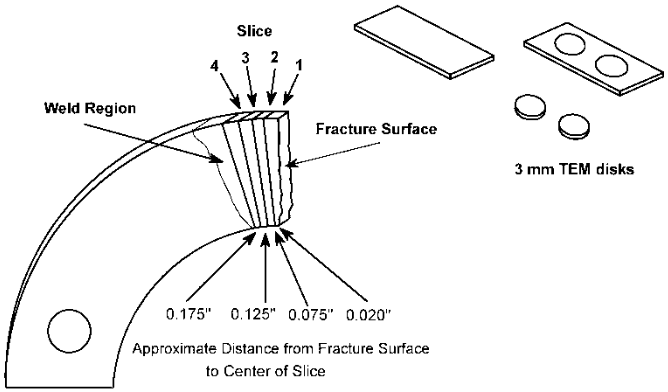
Specimens Exposed to Hydrogen Gas at 350C and 17 Mpa and Tested in 3-Pt Bending



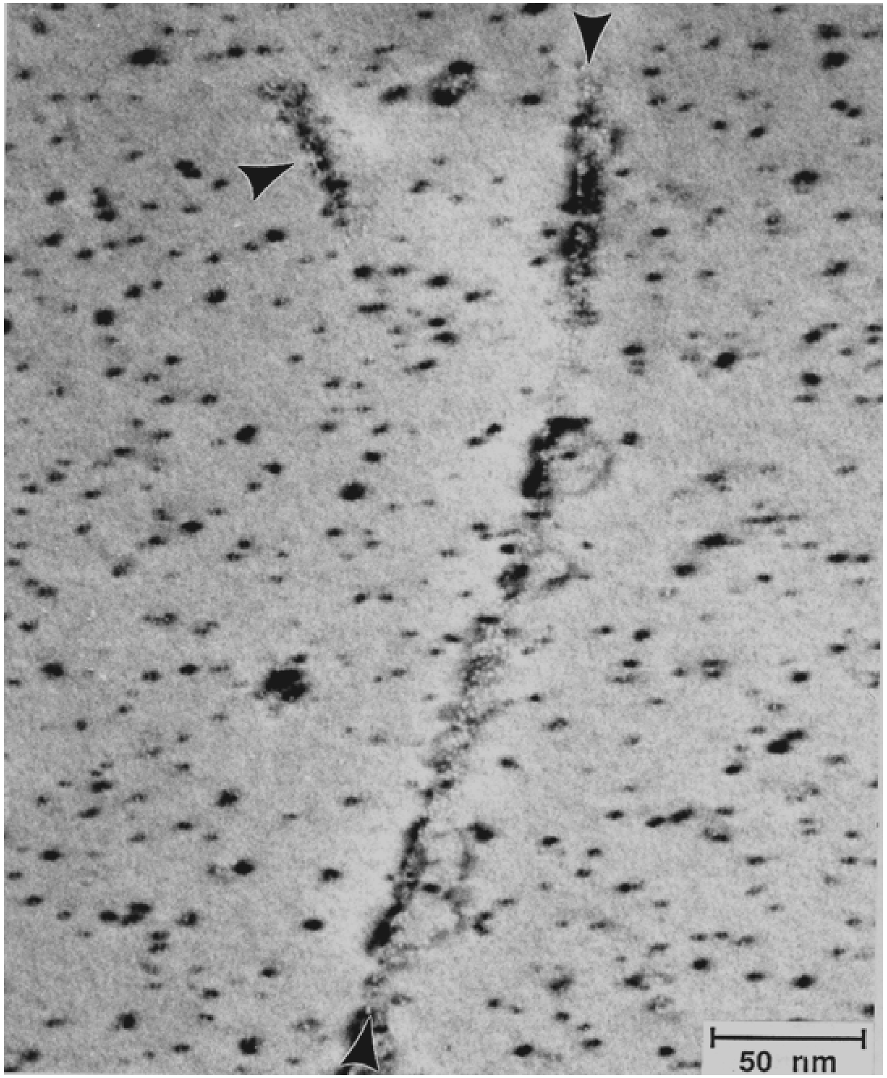


Hydrogen Effect on Toughness Properties

Characterization Techniques – TEM Used for Revealing Helium Bubbles

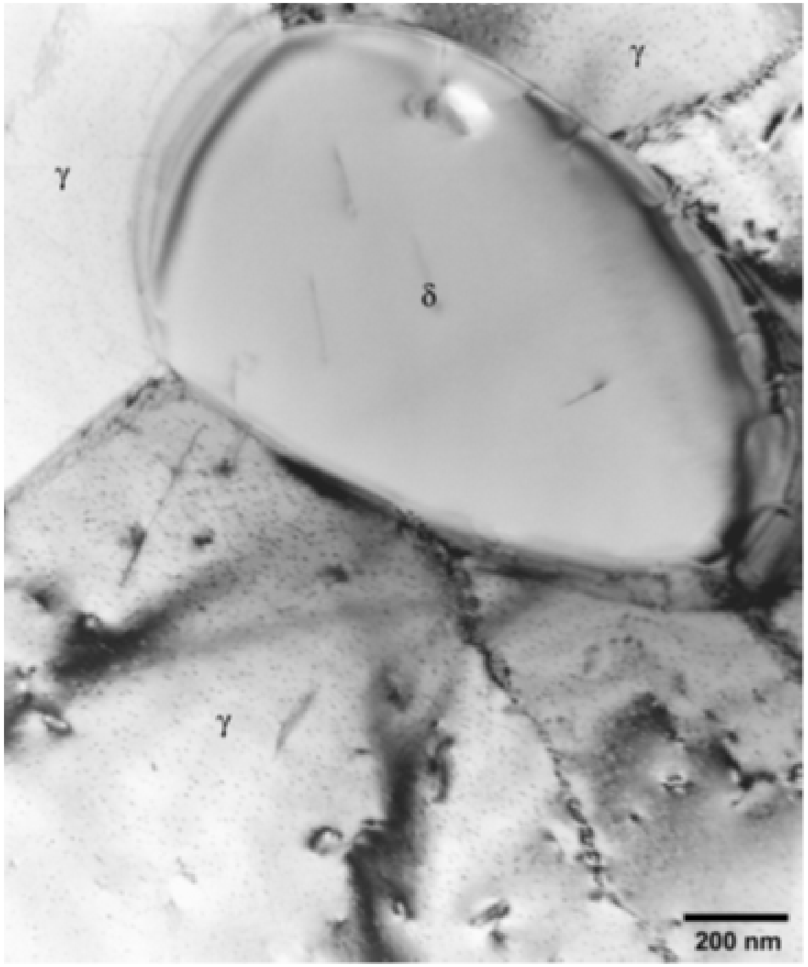


TEM Discs Cut, Punched, and Thinned From Near Fracture Region

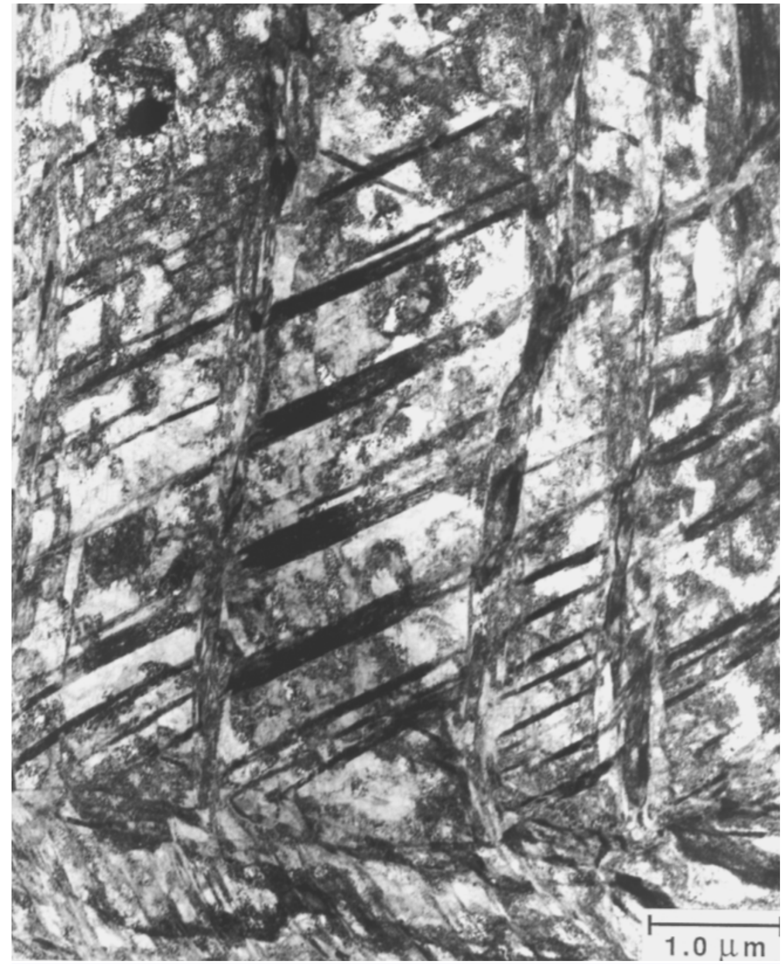


Nanometer sized bubbles randomly distributed and clustered on defects

Decay Helium Bubbles Difficult to Resolve in Forged Steels

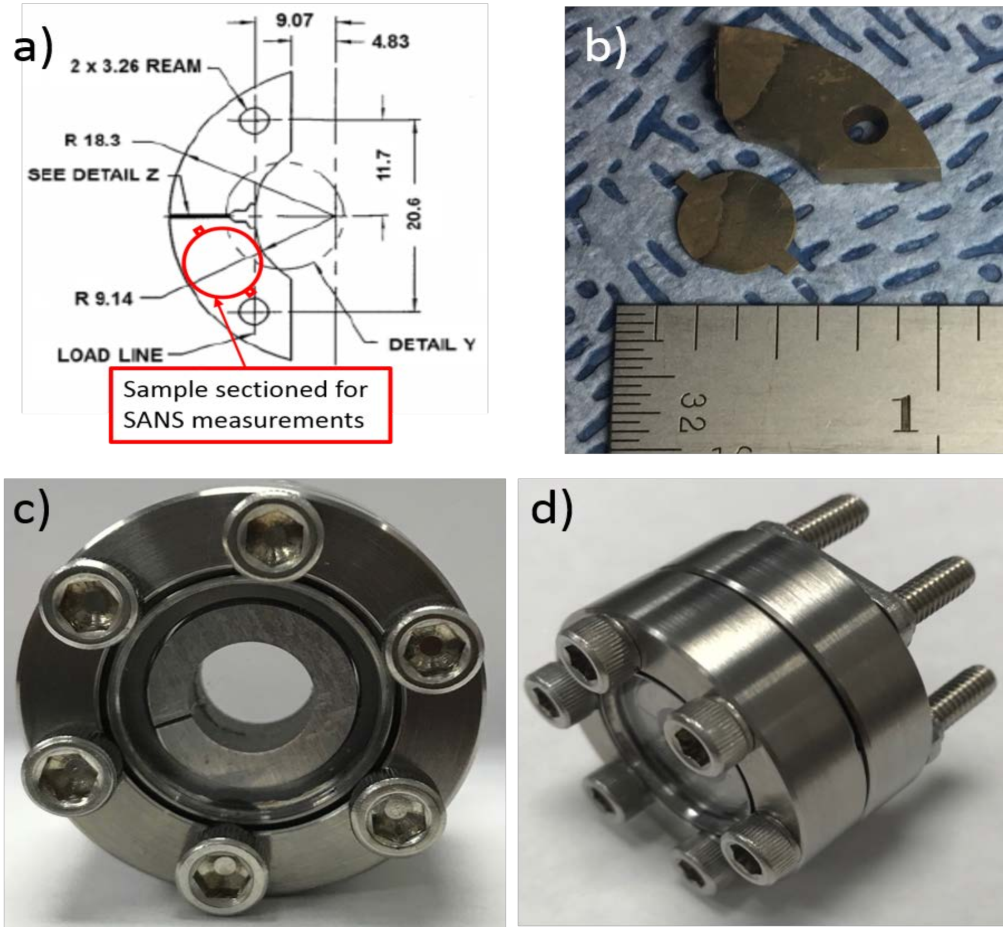


Helium Bubbles Resolved in Austenite of Weldment But Not in Ferrite



Deformation Twinning in Tritium-Aged Forged Steel
Helium Bubbles Not Resolved

Small Angle Neutron Scattering
(Dale Hitchcock, Tim Krentz, & Ken Imrich)



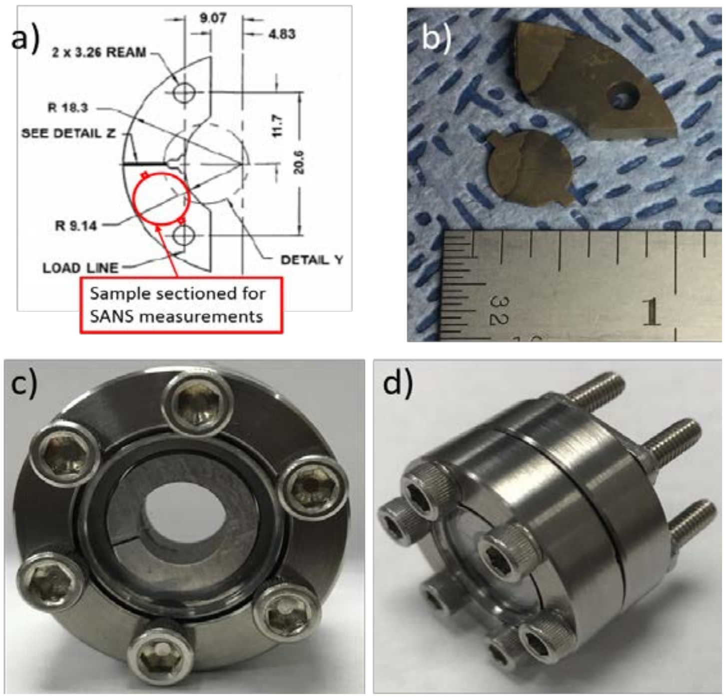
- Small angel neutron scattering (SANS) is being used to probe tritium exposed steels.

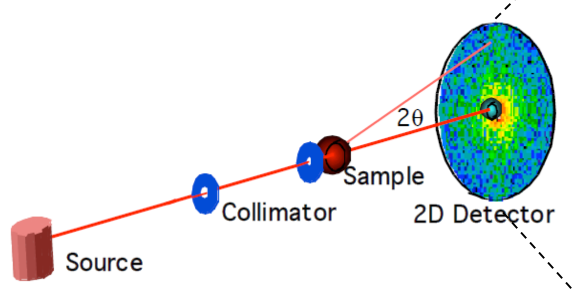
- The purpose is to learn as much as possible about decay helium bubble size, spacing and distribution.

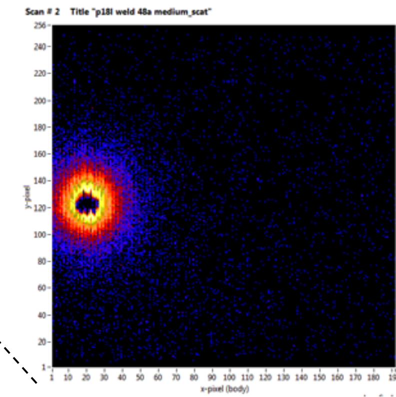
- Complement TEM observations for forged microstructure since nanometer-sized bubbles are not easily be resolved.

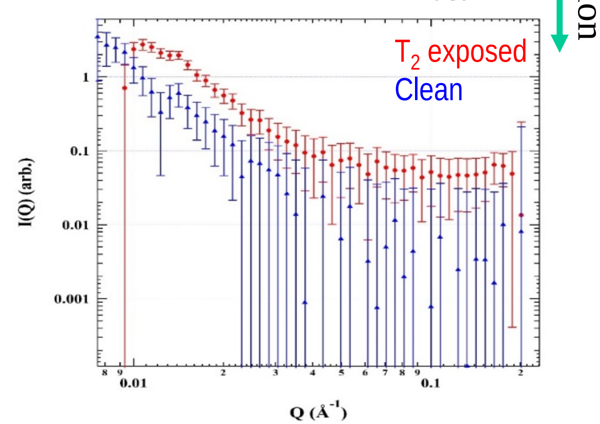
Neutron scattering measurements were performed on tritium exposed stainless steel samples on beamline CG-2 at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Lab (ORNL).

Small Angle Neutron Scattering







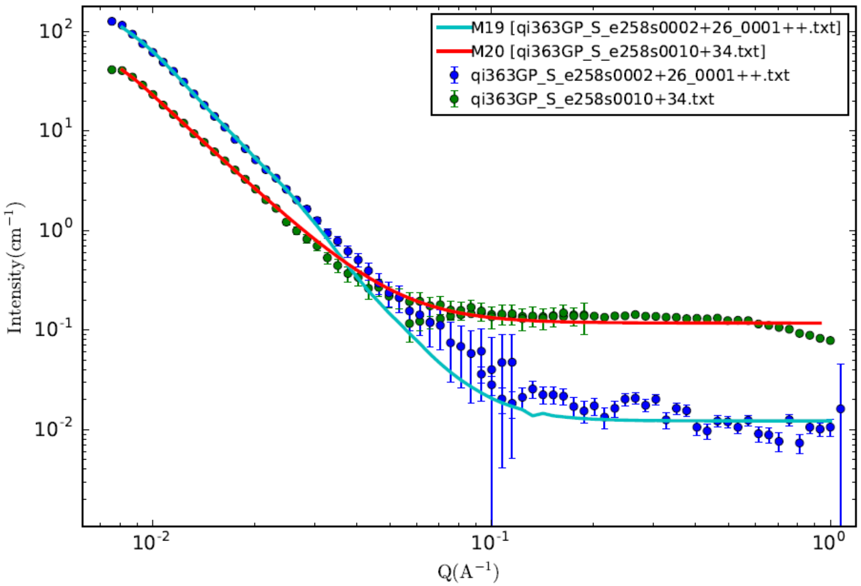


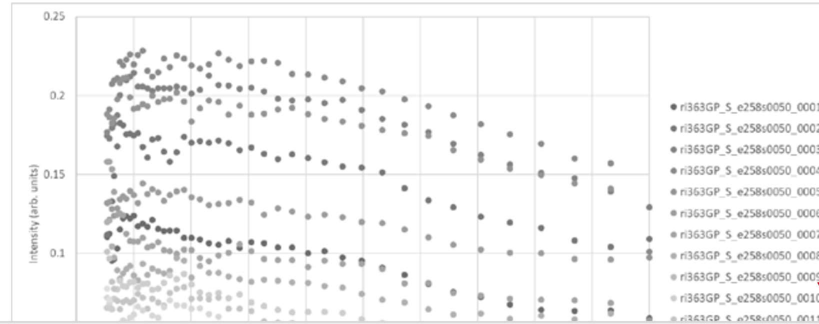
Samples	Charging	Age
304L/308 filled steel weldment	T2	17-year
high energy rate forged 304L steel	T2	3-year
304L/308 filled steel weldment	H2 (5000 psi)	N/A
304L/308 filled steel weldment	H2 (10000 psi)	N/A
304L/308 filled steel weldment	Uncharged	N/A
high energy rate forged 304L steel	uncharged	N/A

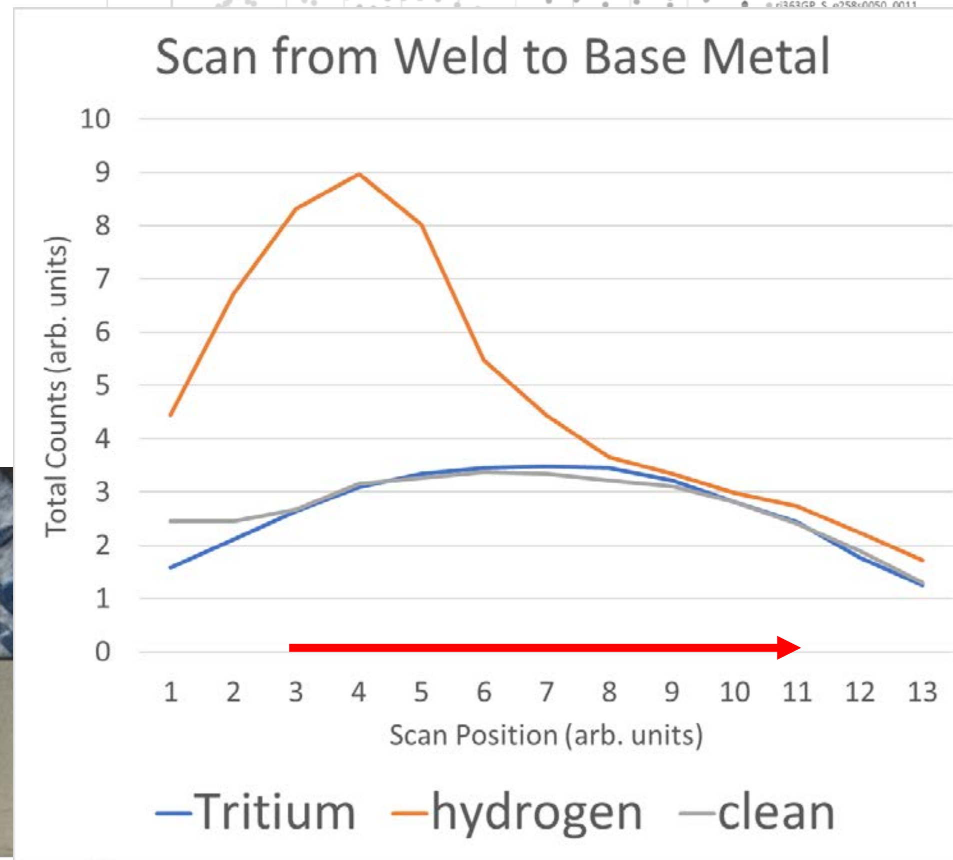
Neutron scattering measurements were performed on tritium exposed stainless steel samples on beamline CG-2 at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Lab (ORNL).

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.

1 Small Angle Neutron Scattering

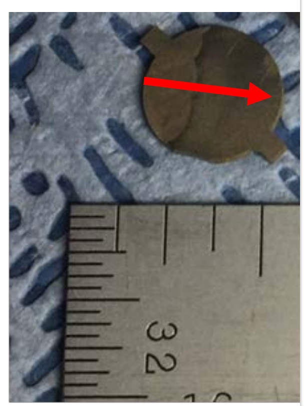




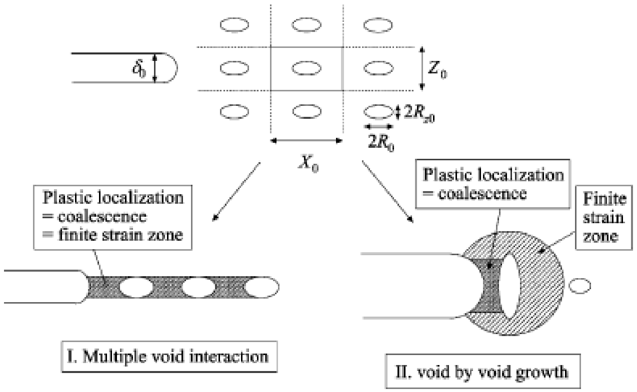


Hydrogen charged weld – fit with Gunier-Porod model (incoherent background 1 order of magnitude higher than tritium – matches decay + T vs H cross sections)

Tritium charged weld – fit with hard sphere model (Rg (ave. bubble size) =2.3nm, bubble pressures ~ 3GPa)



Modeling for Simulation and Prediction (P. S. Lam and U of Mich)



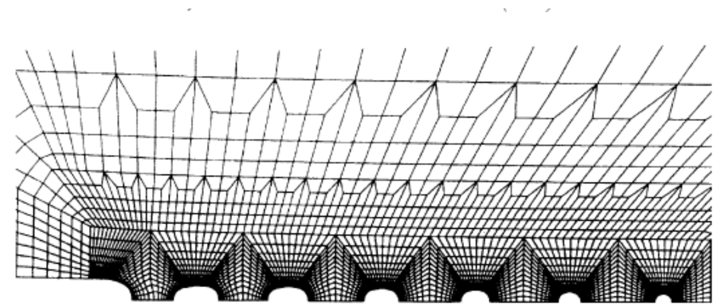


Fig. 4. Finite element 2D small scale yielding simulations of ductile crack growth with discrete voids [9].

Can models be built using existing data and/or short-term tests that predict performance after many years in tritium service?

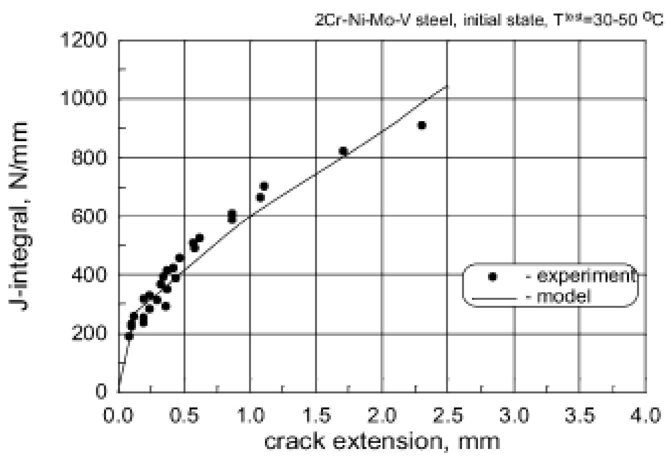


Fig. 13. Comparison between the predicted and the experimental J_R-curves.

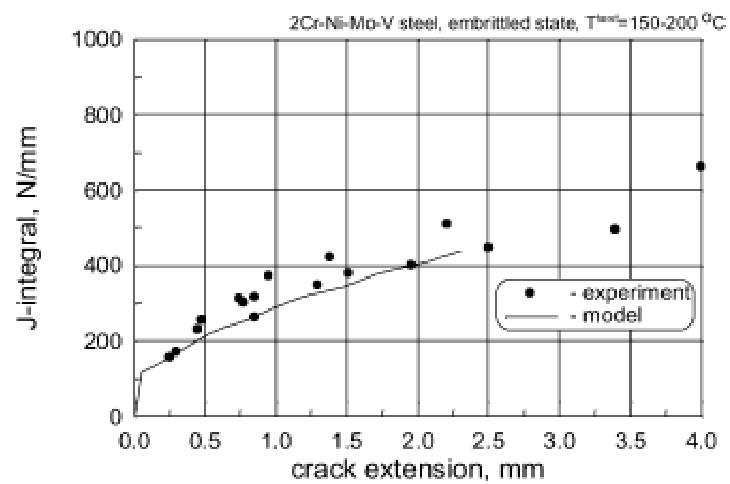


Fig. 14. Comparison between the predicted and the experimental J_R-curves.

SRNL

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.

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- **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.