SAND2014-2181P

# Hydrogen embrittlement of welds in structural steels

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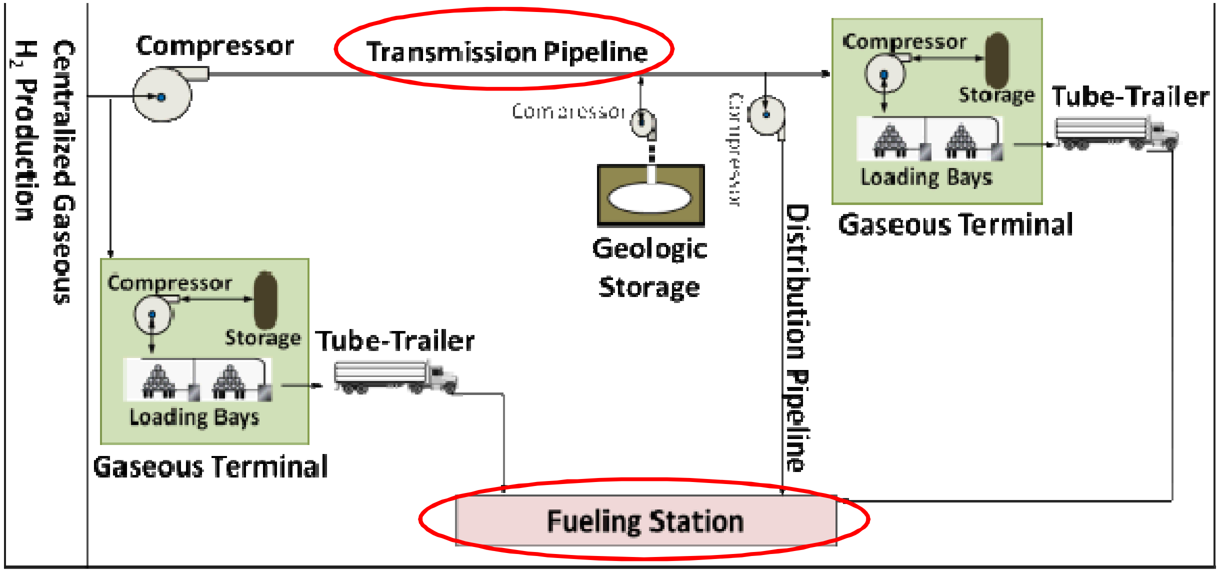
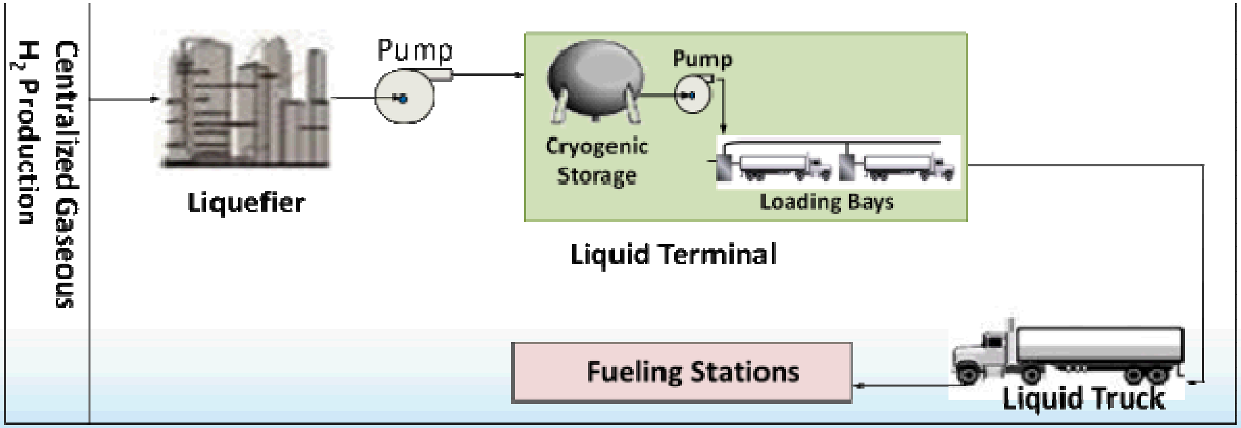
# Materials are central focus for cost reduction and reliability of $H_2$ fuel infrastructure

Gaseous Delivery Pathways




Liquid Delivery Pathway




A. Elgowainy, ANL

***Hydrogen embrittlement recognized as potential reliability issue***

# Motivation

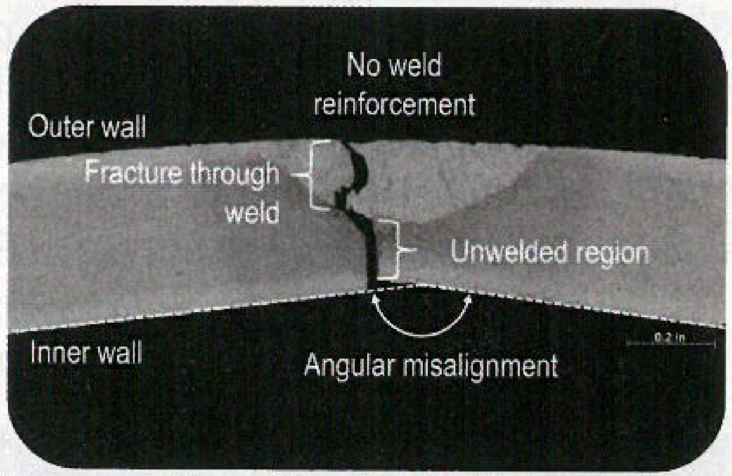
- Steel hydrogen containment components are expected to contain welds
  - Steel transmission pipeline fabrication and installation requires two welds: seam weld and girth weld
  - Orbital tube welding is an effective joining strategy for gas handling and dispensing manifolds
- Hydrogen embrittlement of welds has not been extensively characterized

Conclusion from *Workshop on Hydrogen Compatible Materials* at SNL/CA (November 2010):

- Evaluation of welded structures is critical to deployment of hydrogen infrastructure and technology

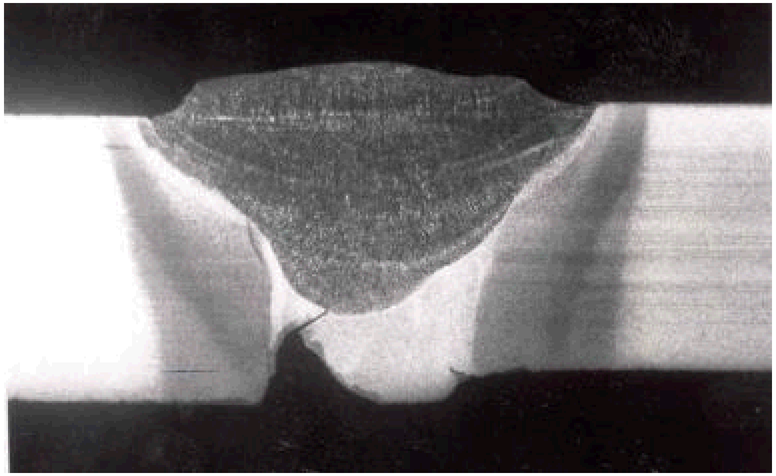
# Reliability of hydrogen containment components may be controlled by welds

F. Richards, *Adv Mat & Processes*, 2013




*Cross section of longitudinal seam in the pup where the rupture initiated.*

I. Alliat, NATURALHY EC project, 2007



- Welding can create defects, increasing probability of crack growth in these regions
- Are weld microstructures (fusion zone, heat-affected zone) more susceptible to hydrogen embrittlement?

# Reliability assessment of steel $H_2$ pipelines must consider hydrogen embrittlement

- Why are steel pipelines candidates for $H_2$ fuel transmission?
  - Steel hydrogen pipelines are safely operated under *static pressure*
- Need: demonstrate reliability of steel hydrogen pipelines for *cyclic pressure* applications
  - Evolving structural integrity management standards employ damage-tolerant approach, requiring fatigue crack growth data in $H_2$ gas

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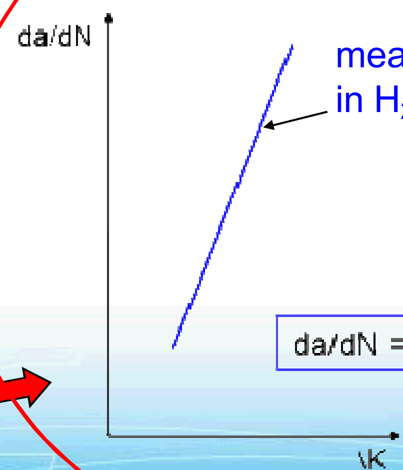
$\Delta K = \Delta p[f(a, t, R_o, R_i)]$




$da/dN = C[\Delta K]^m$

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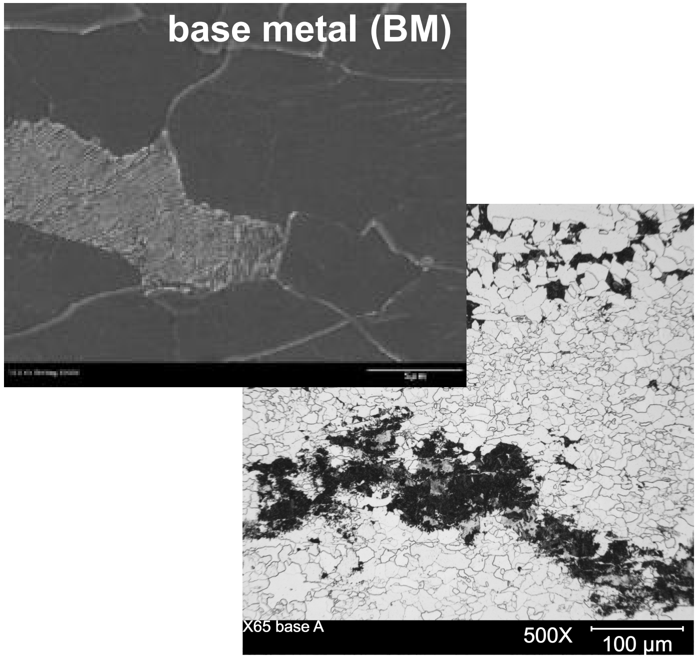
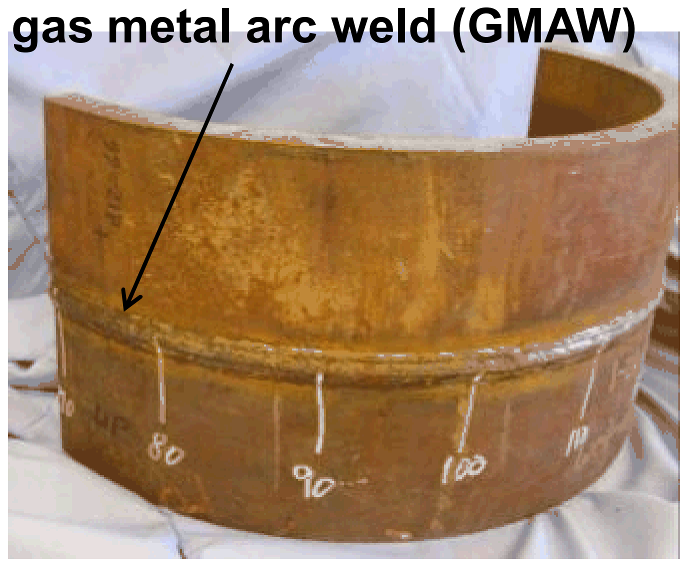
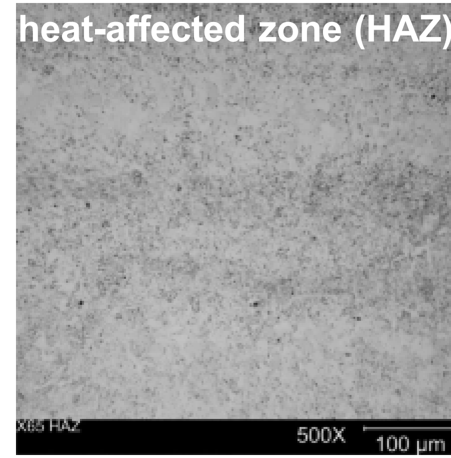
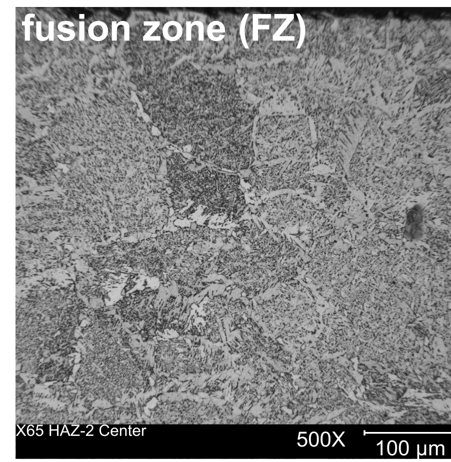
# Fatigue crack growth measurements performed on API 5L X65 pipe with GMAW




508 mm OD x 25.4 mm WT

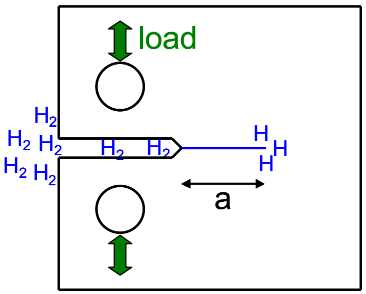


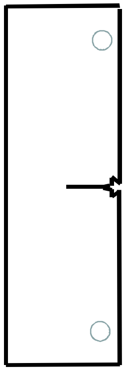



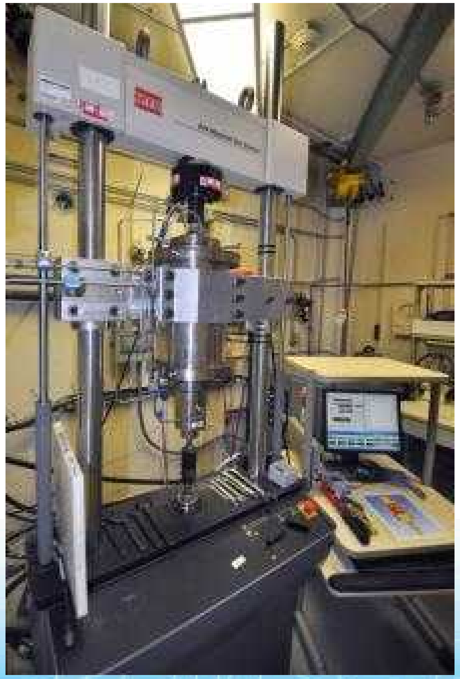

- Tensile properties
  - Yield strength: 591 MPa
  - Ultimate tensile strength: 662 MPa
- Base metal alloy composition

| C | Mn | P | S | B | Si | Cu | Ni | Nb | Ti |
|---|---|---|---|---|---|---|---|---|---|
| 0.08 | 1.53 | 0.01 | 0.001 | 0.002 | 0.32 | 0.024 | 0.038 | 0.039 | 0.002 |

# Fatigue crack growth relationships measured in high-pressure $H_2$ gas





- Material
  - X65 base metal and GMAW
- Instrumentation
  - Internal load cell in feedback loop
  - Crack-opening displacement measured internally using LVDT
  - Crack length calculated from compliance
- Mechanical loading
  - Triangular load-cycle waveform
  - Constant load amplitude
- Environment
  - Supply gas: 99.9999% $H_2$
  - Pressure = 21 MPa (3,000 psi)
  - Room temperature

# Fatigue crack growth rates in base metal depend on specimen orientation

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X65 base metal


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***Does banded ferrite-pearlite microstructure reduce crack growth rates in L-R orientation?***

# Previous results show that $H_2$-assisted crack growth lower in pearlite compared to ferrite

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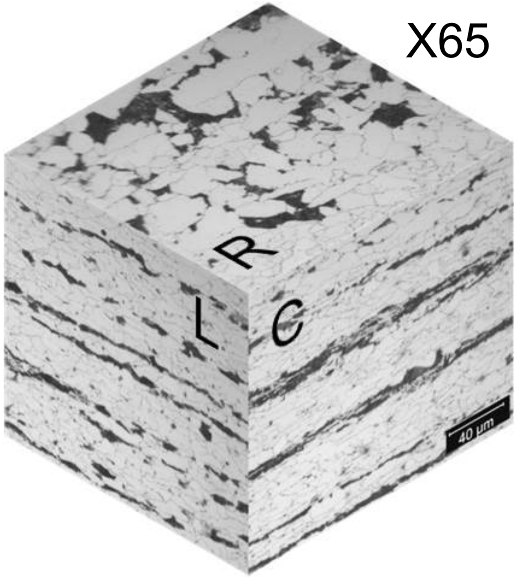
X65




Iron and 1080 steel data:
H. Cialone and J. Holbrook, *Microstructural Science*, 1987

***Banded pearlite may contribute to reduced fatigue crack growth rates in X65 base metal***

# Welds not inherently more susceptible to $H_2$-accelerated cracking compared to base metal



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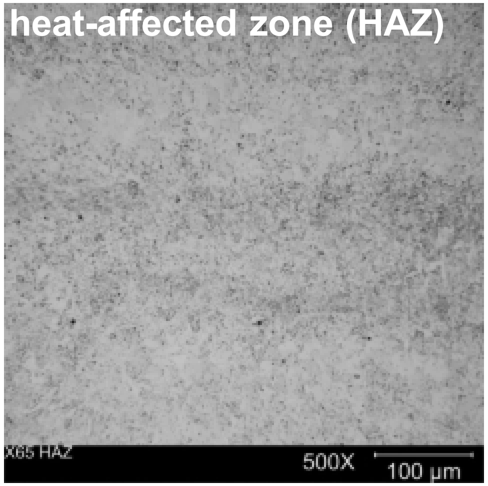
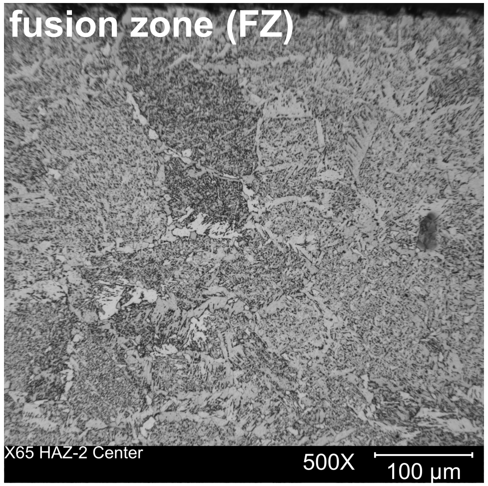
***Unexpectedly, fatigue crack growth rates in HAZ lower than rates in FZ***

# Two features may account for difference in fatigue crack growth rates for HAZ vs. FZ

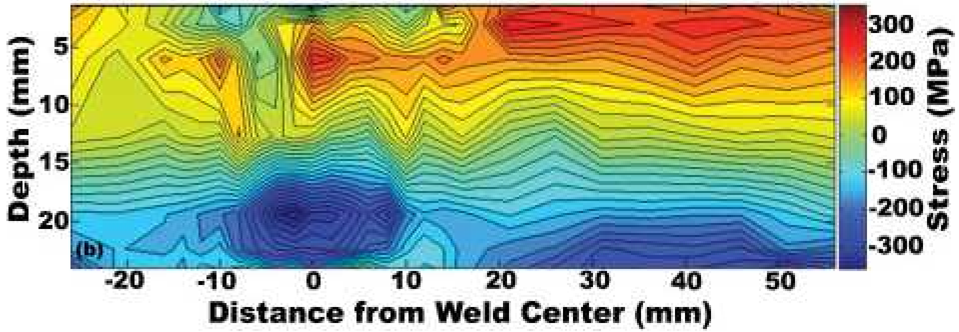
## Microstructure




## Residual Stress

longitudinal stress




**pipe longitudinal direction**

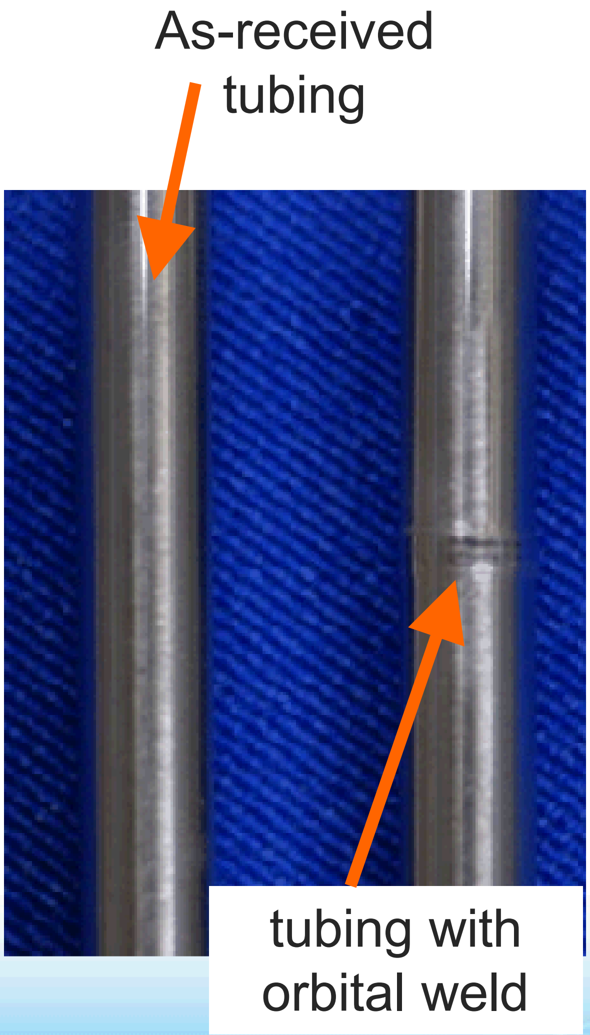
T. Neeraj, *Science and Technology of Welding and Joining*, 2011

***More testing planned to clarify variable controlling fatigue crack growth rates***

# Tensile properties of tubing and orbital tube welds were evaluated

Testing scope: Uniaxial tension

- As-received tubing
- *Internal hydrogen* (~140 wtppm)
  - Produced by thermal precharging (573K in 140 MPa H2)
  - Simulates hydrogen at stress concentrations
- *Orbital tube welds*
  - Automated autogeneous GTA welds
- Effect of subambient *temperature*
  - 293 K (room temperature)
  - 223 K (-50˚C)



# A range of alloy compositions and material strength have been evaluated

| Alloy type | ID | Yield strength (MPa) | Cr (wt%) | Ni (wt%) | C (wt%) | S (wt%) |
|---|---|---|---|---|---|---|
| 316L | 316L | 286 | 16.7 | 12.4 | 0.018 | 0.006 |
| 304L | 1 | n/m | 18.2 | 9.1 | 0.020 | 0.004 |
| 304L | 2 | 656 | 18.6 | 11.7 | 0.021 | 0.0004 |
| 304L | 3 | 296 | 18.7 | 11.6 | 0.015 | 0.026 |
| 304 | 4 | 627 | 18.3 | 10.2 | 0.04 | 0.002 |
| 304L | 5 | 359 | 18.4 | 10.2 | 0.01 | 0.007 |
| 304L | 6 | 763 | 18.4 | 8.2 | 0.024 | 0.003 |

Low nickel

High carbon

High sulfur

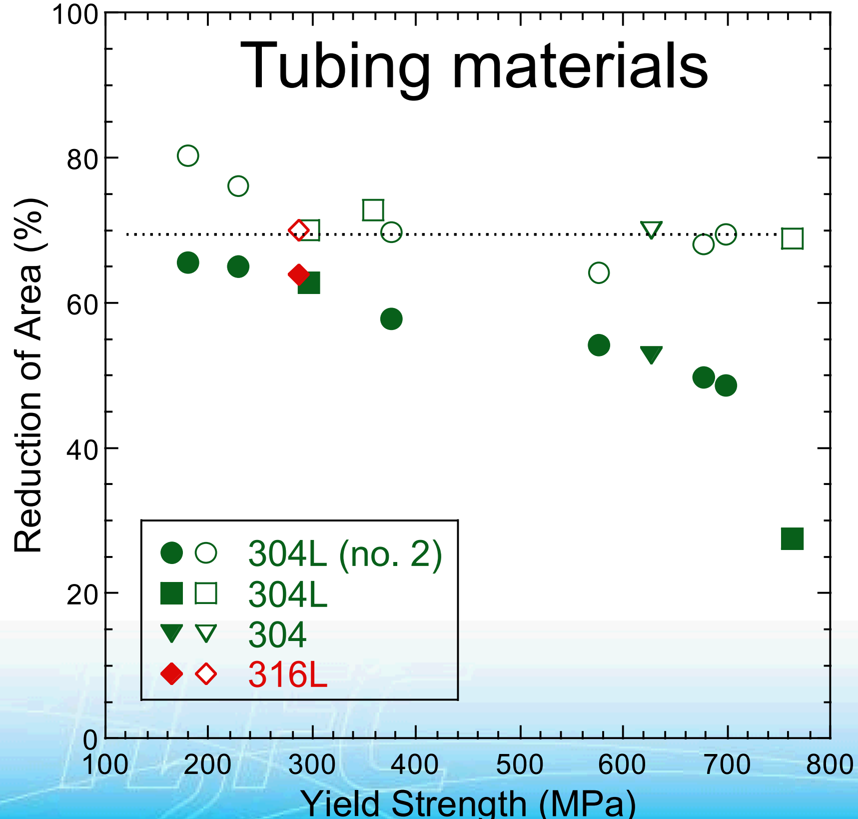
# Thermal treatment was used to vary microstructure

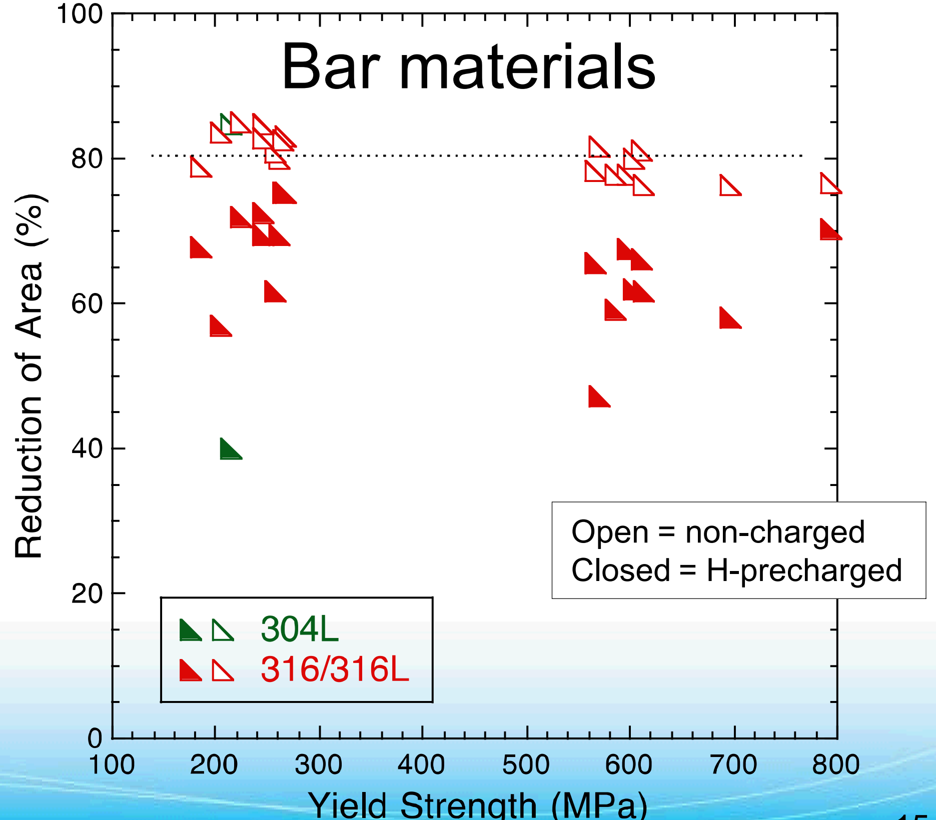
| Material/ Condition | Temperature (K) | Time (min) | Yield strength (MPa) | Target microstructure |
|---|---|---|---|---|
| 2A | n/a | n/a | 697 | Strain-hardened (as-received) |
| 2B | 866 | 60 | 678 | Partially recovered |
| 2C | 1000 | 30 | 576 | Full recovery |
| 2D | 1033 | 30 | 377 | Partially recrystallized |
| 2E | 1116 | 60 | 228 | Fully recrystallized |
| 2F | 1311 | 60 | 179 | Annealed |
| *x*S | 998 | 240 | varies | Sensitized |

- Heat treatments applied to only alloy no. 2 (304L)
- Sensitization is used to evaluate susceptibility to stress corrosion cracking (not evaluated for the 316L alloy)

# Austenitic stainless steel tubing behaves similarly to bar materials

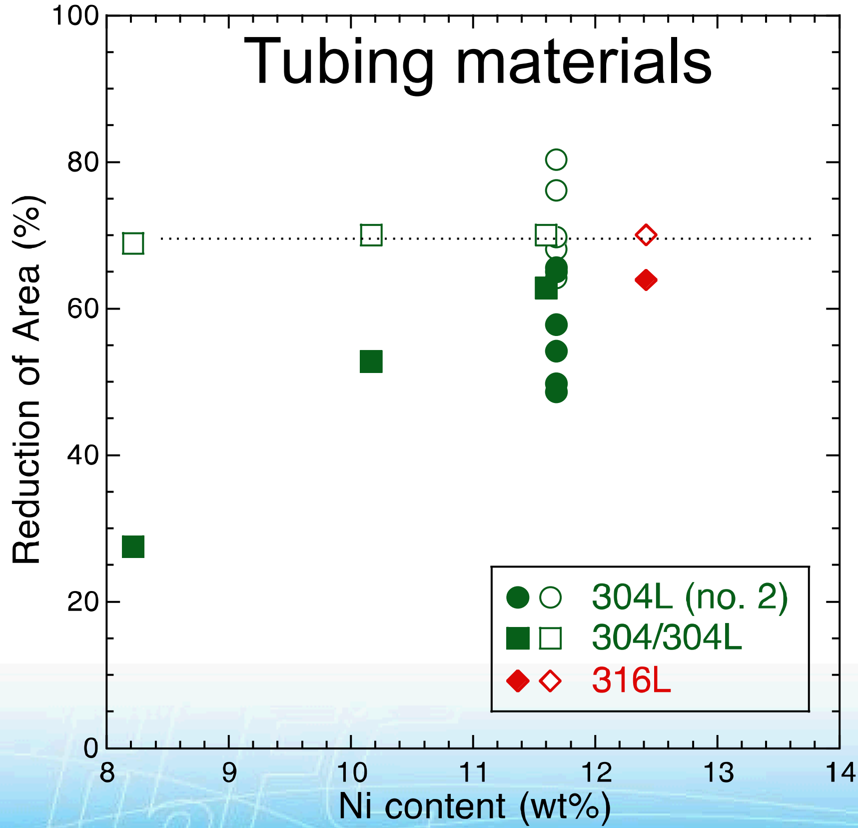
- Higher yield strength results in lower tensile ductility in the presence of hydrogen for both tubing and bar
- Other material variables can dominate effects

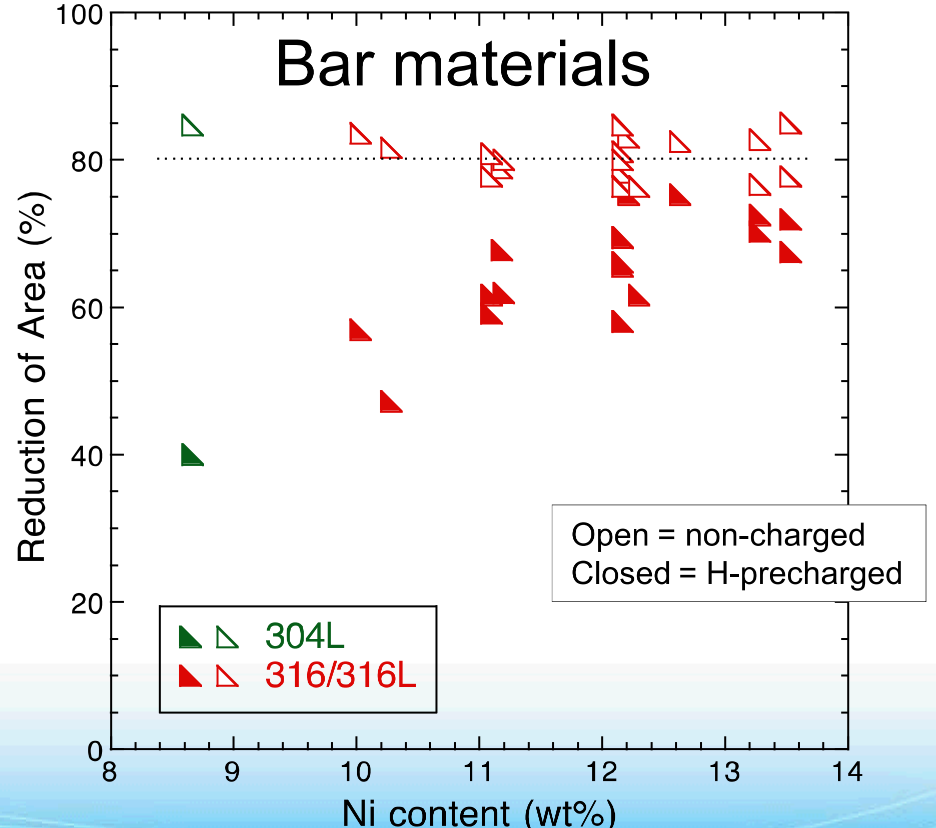


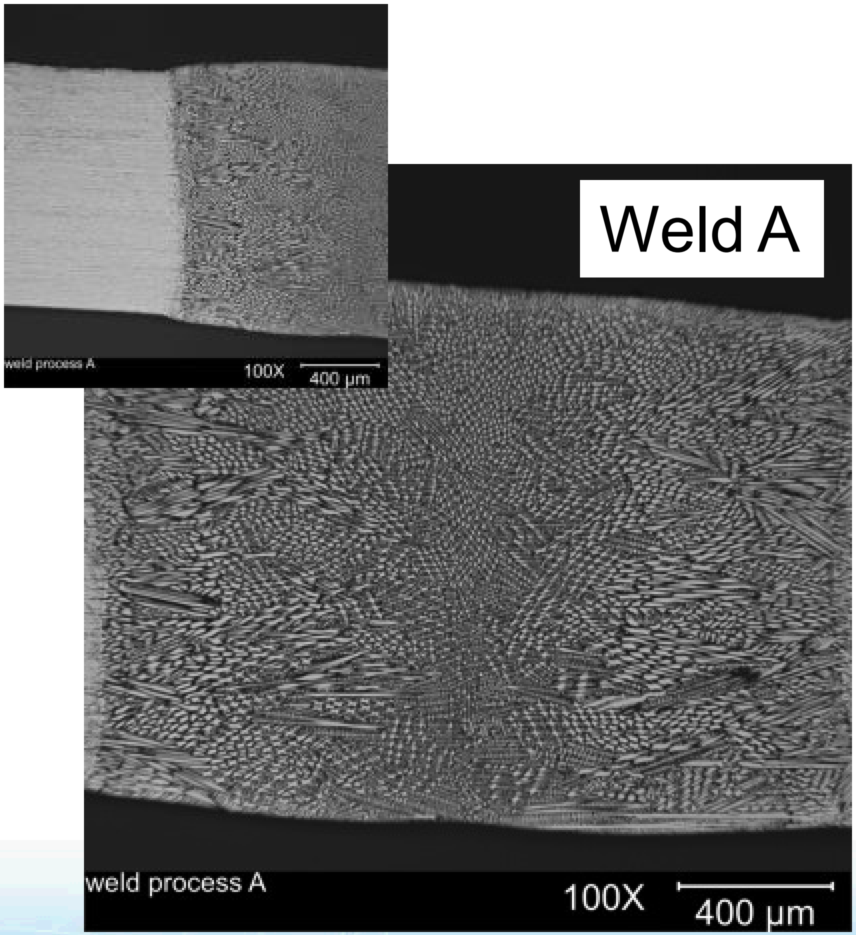
# Compositional trends are also similar for tubing and bar

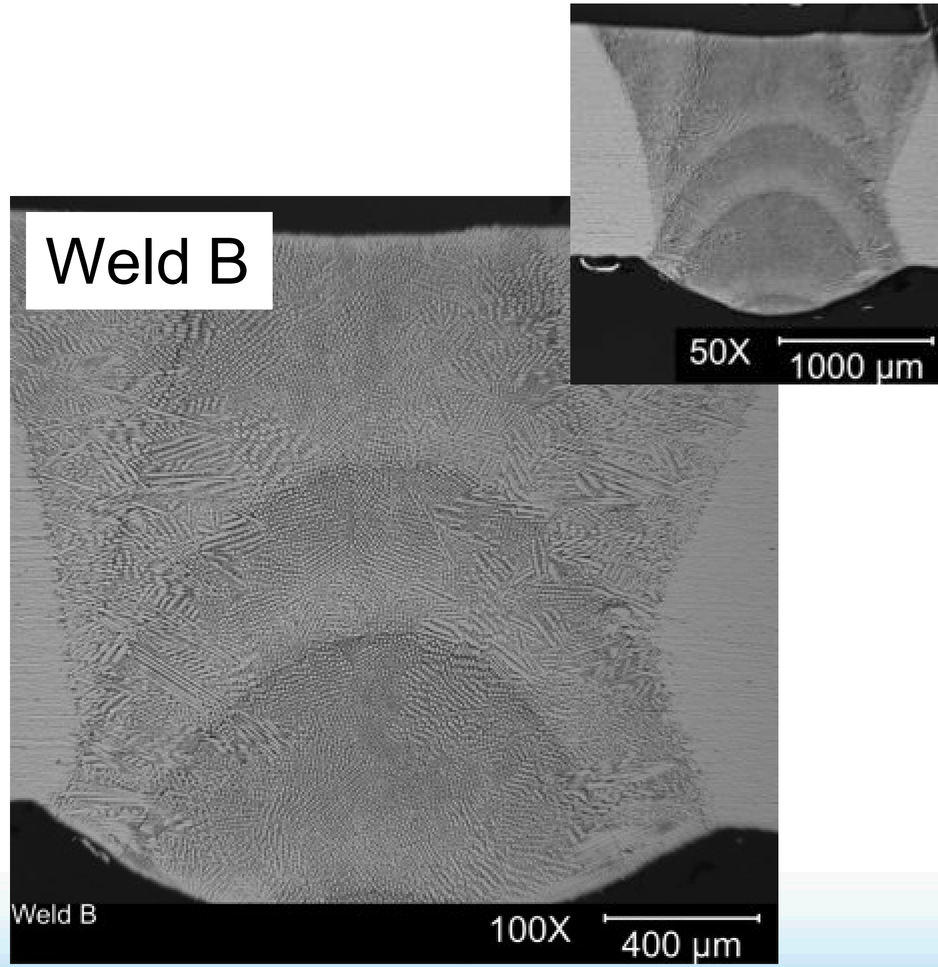
- Nickel composition is an important variable in hydrogen-assisted fracture of austenitic stainless steels



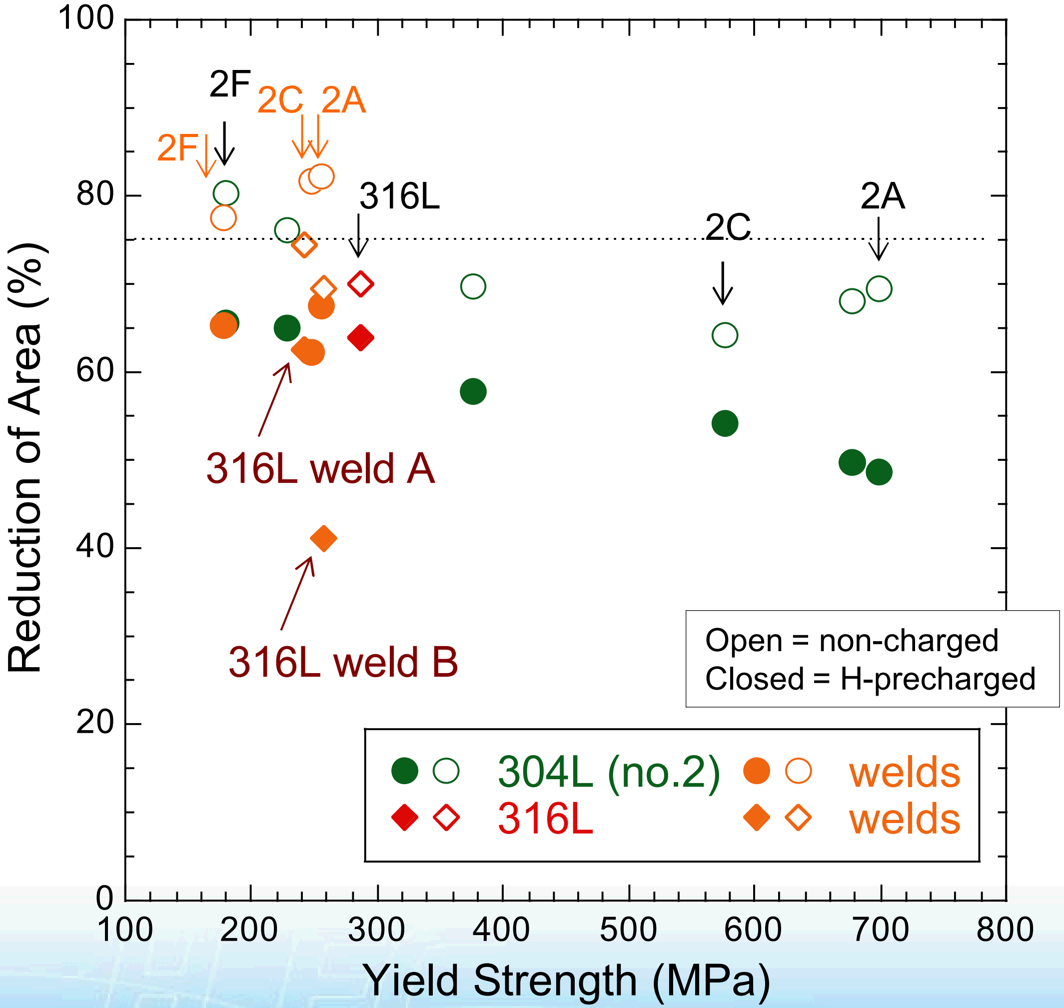



# Optical microscopy of type 316L orbital tube welds



# Orbital tube welds behave similarly to the tubing materials

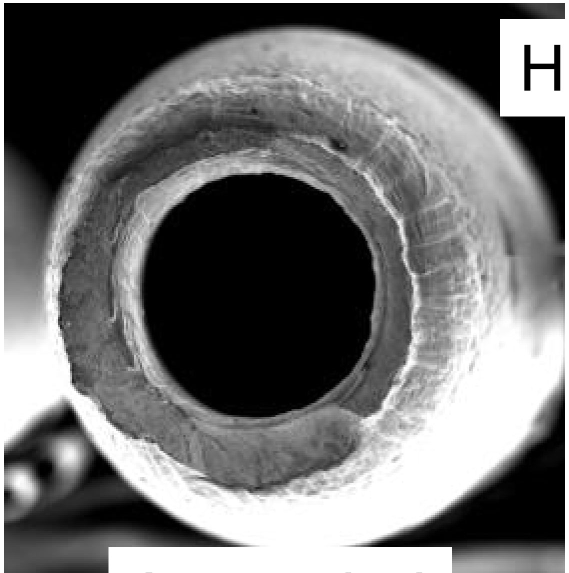


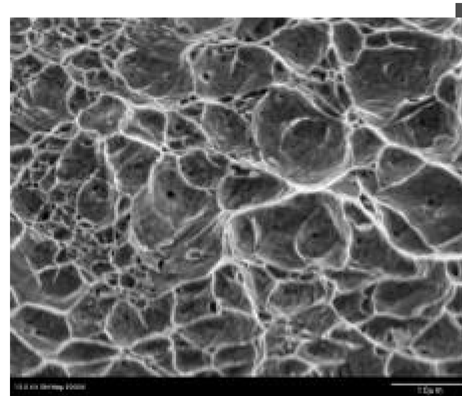

- The yield strength of the welded specimens are similar to annealed tubing
- Tensile ductility H-precharged welded specimens can be similar to tubing

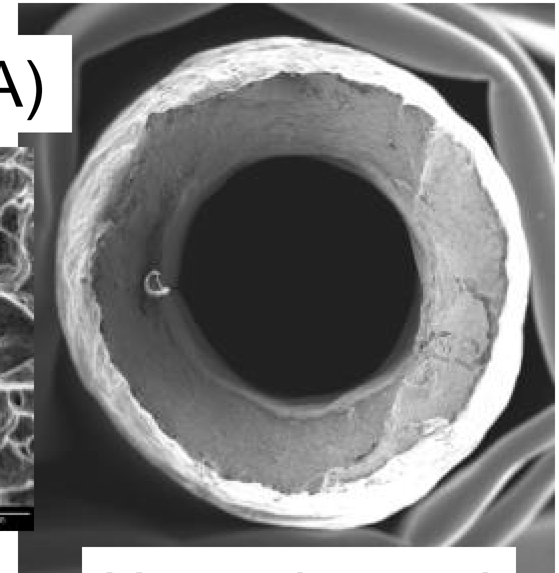
- Two sets of 316L welds (A and B) produced independently, resulted in different effects of H
  - Weld B is still very ductile

# Fractography of type 304L orbital tube welds

High strength tubing (2A)

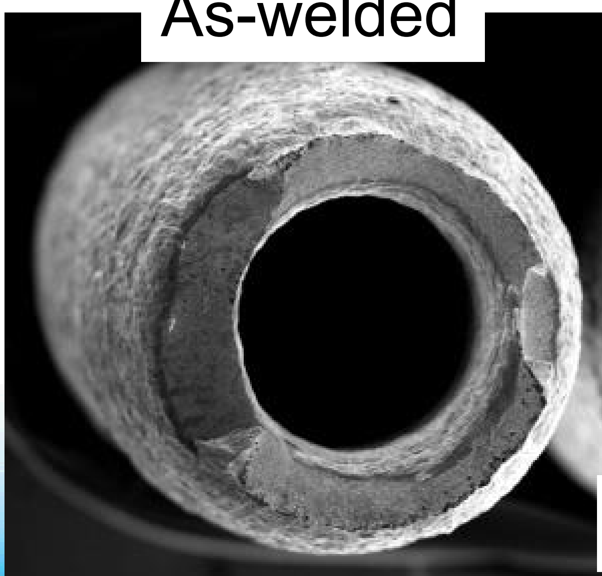


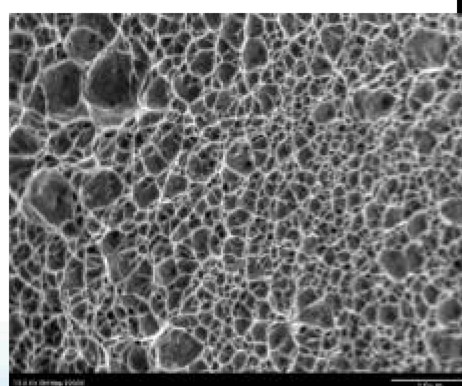


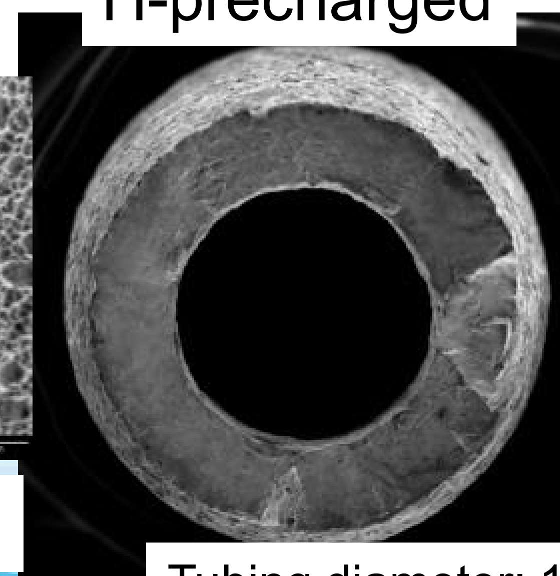


As-welded

H-precharged





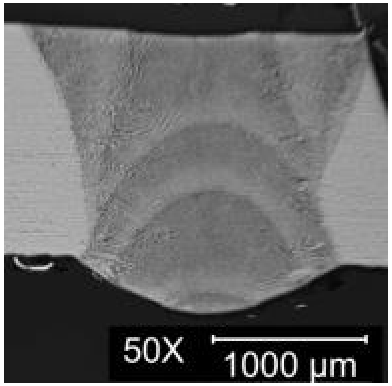


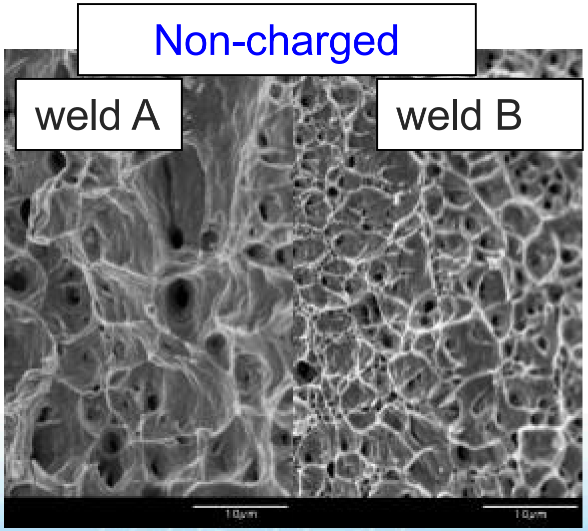
Annealed tubing (2F)

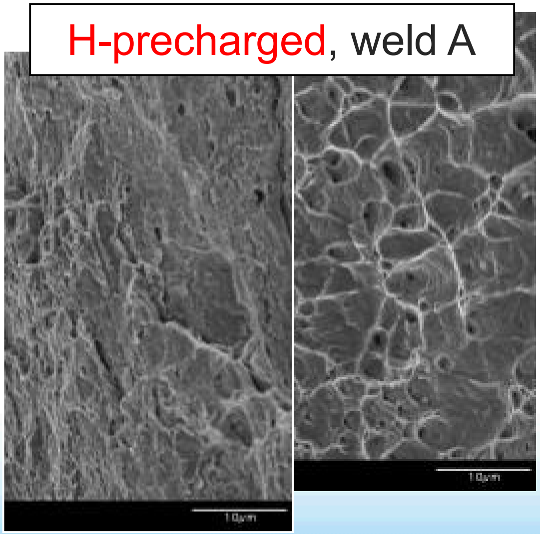
Tubing diameter: 1/8”

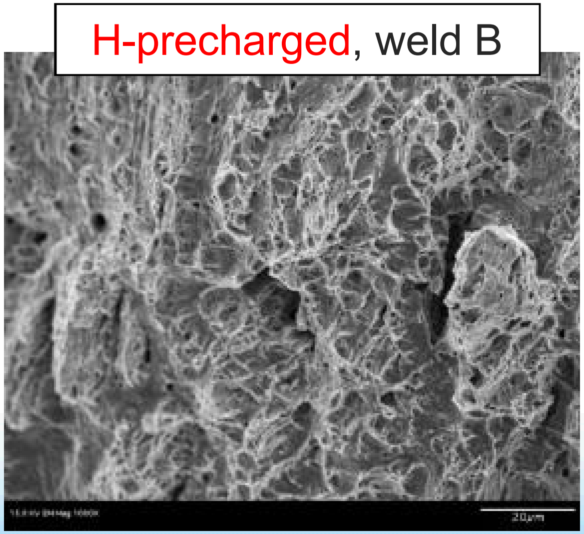
# Relationship between welded microstructure and hydrogen-assisted fracture of welds is still emerging

- Evaluation of microstructure and fractography shows hydrogen effects are consistent with experience from base materials (type 316L bar)
- Fracture features from weld A show no clear relationship to weld microstructure
- Fracture features from weld B suggest local regions with intrinsic relationship to weld microstructure



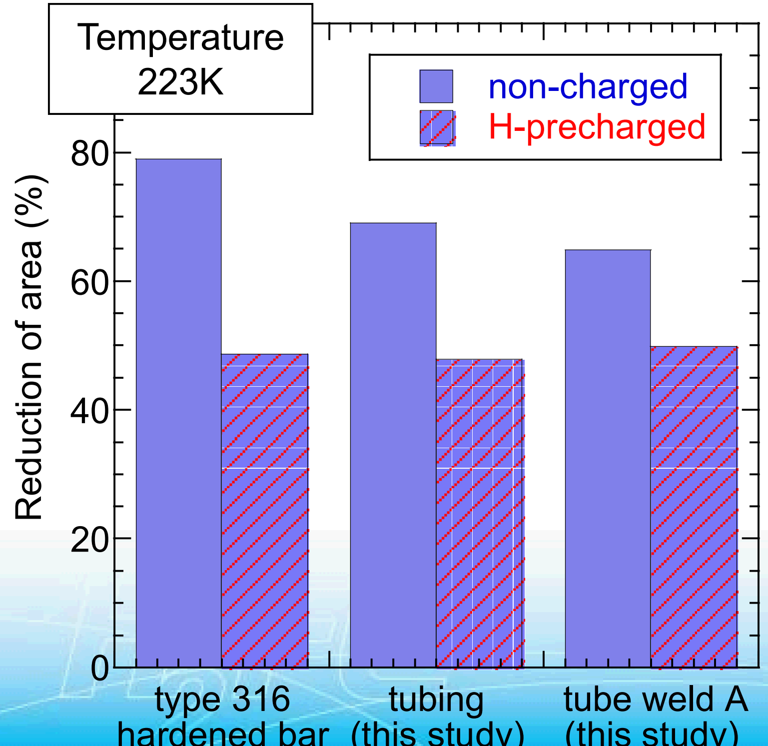



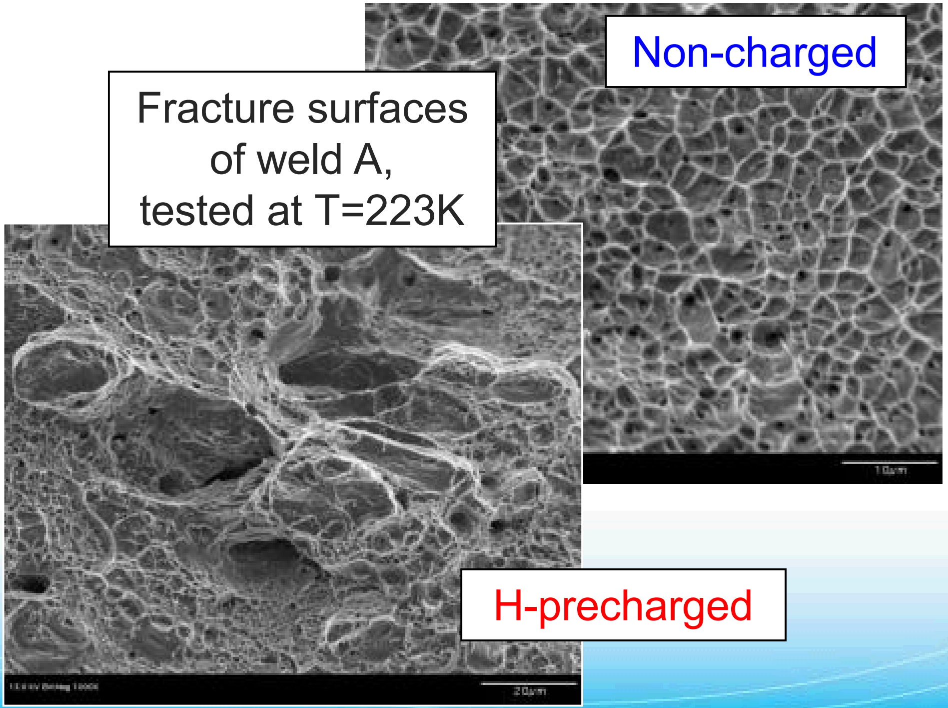




Room temperature fracture surfaces: 316L tubing

# Effect of low temperature evaluated for orbital tube welds

- Hydrogen-assisted fracture in austenitic stainless steel generally enhanced at low temperature
- At temperature of 223K, like at room temperature, hydrogen effects on ductility similar for as-received tubing and weld A
- Fracture surfaces show ductile features with the involvement of boundaries when H-precharged, similar to bar materials



# Summary

- Fatigue crack growth relationships measured for pipeline steel base metal and girth weld in $H_2$ gas
  - Orientation-dependent fatigue crack growth rates in base metal attributed to pearlite banding
  - Weld microstructures not inherently more susceptible to $H_2$-accelerated crack growth compared to base metal
- Hydrogen effects evaluated in austenitic stainless steel tubing and orbital tube welds
  - Effect of internal hydrogen on tensile ductility in tubing is similar to previous work on bar materials
  - Welded specimens generally display similar tensile ductility as the tubing of the same strength
  - Welded specimens remain very ductile after hydrogen precharging
- b
- Conclusion: These limited results for pipeline steels and stainless steel tubes indicate hydrogen embrittlement may not be more severe in welds compared to base metal

# Acknowledgments

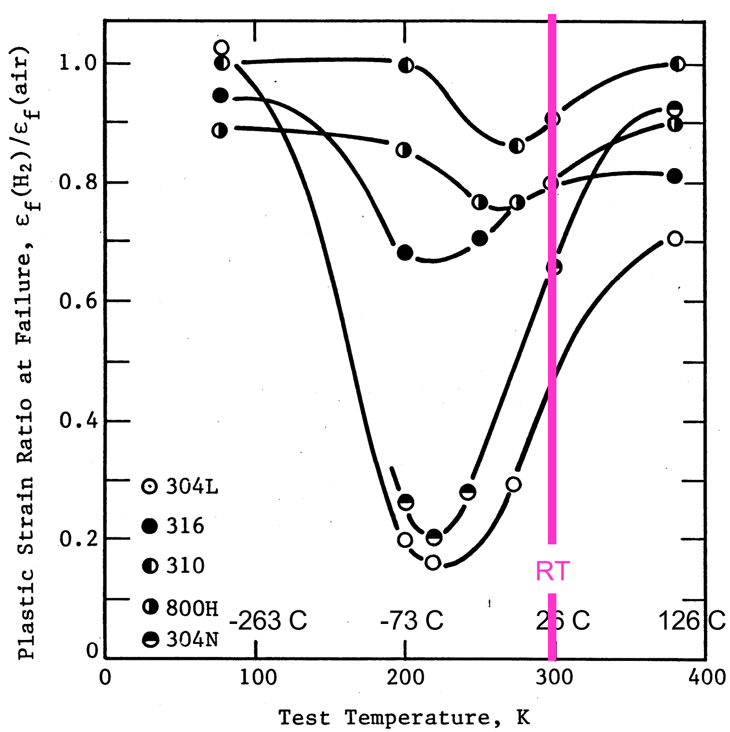

- The ongoing support from the US Department of Energy, Office of Energy Efficiency and Renewable Energy, Fuel Cell Technologies Office is gratefully acknowledged

- Hydrogen Effects on Materials Laboratory team at Sandia National Laboratories in Livermore CA:

Brian Somerday
Chris San Marchi
Ken Lee
Jeff Campbell
Mark Zimmerman
Joe Ronevich
Tom Reynolds
Zach Harris

# Stainless steel susceptible to hydrogen embrittlement at lower temperatures

*Austentic stainless steel ductility. Tensile Data*

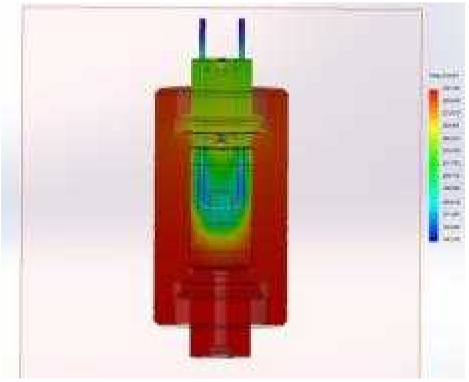



- Stainless steels are essential to national defense and hydrogen energy missions
  - Many applications require operation down to -50˚C
- Materials such as stainless steels show greater hydrogen embrittlement at low temperature
- Materials qualification testing must include testing over the full range of operating temperatures

*US institutions currently lack the ability to conduct dynamic fracture tests at -50˚C in high pressure hydrogen*

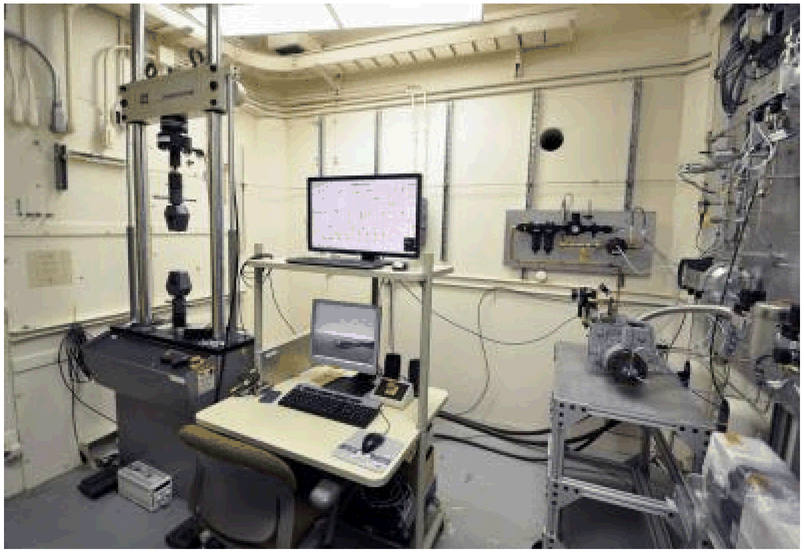
# We plan to fill the need with the appropriate test capability

| | Tensile/fracture | Fatigue |
|---|---|---|
| Pressure | 3-138 MPa (200-20,000 psi) | 3-138 MPa (200-20,000 psi) |
| Temperature | -100° C to 200° C (148° F to 392° F) | -100° C to 200° C (148° F to 392° F) |
| Force | 44 kN (10,000 lbs) | 22 kN (5,000 lbs) |
| Displacement | 25 mm ( 1 inch) | 5 mm (0.2 inch) |
| Test control | .025-25 mm/s: tensile | 0.001-10 Hz |



*This capability is also important to our stakeholders*

# 2 of 3 subsystems installed

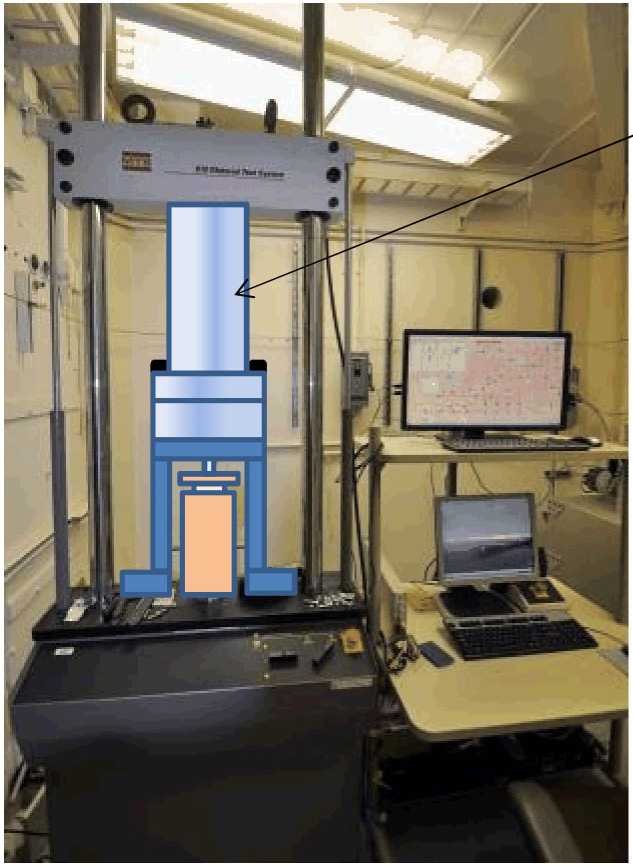


- Procured test frame, test controller, hydraulic pump, testing software.

- Gas handling manifold designed and installed
- Automated gas management software developed and exercised

*Significant progress toward final system*

# Significant progress but not there yet



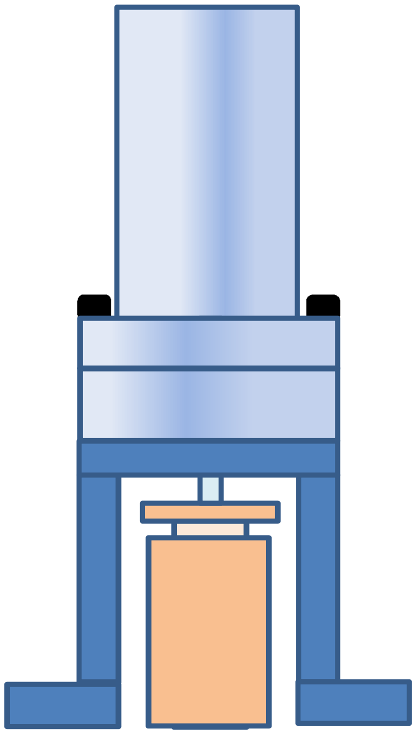
- Missing subsystem– Variable temperature pressure vessel and chiller.

*What are remaining needs for procuring vessel?*

# Two final needs for procuring variable-temperature pressure vessel

- Specify critical design details
  - Internal or external cooling mechanism
  - Thermal analysis of vessel

- Need final installment of funding
  - Qualified vessel design and fabrication
  - Chiller procurement



*Partnerships enabled design specifications*

# Relationships with partners to provide tools and insights

Sandia National Laboratories

HYDROGENIUS

VTT Technical Research Centre of Finland

cea

Powertech

BOISE STATE

TWI

NIST National Institute of Standards and Technology U.S. Department of Commerce

NIPPON STEEL & SUMITOMO METAL

cnrs

Université de Poitiers

isae Institut Supérieur de l'Aéronautique et de l'Espace ENSMA

KRISS Korea Research Institute of Standards and Science

# Material testing workshop provided confidence for internal cooling mechanism

Sandia National Laboratories

- Advancing Materials Testing in Hydrogen Gas, 2013
- International Workshop
- Representatives from 10 institutions, 7 countries


*Japan*

*France*

*Korea*

*Finland*

*Canada*

*UK*

*USA*

*World leaders in material testing*

# How did we benefit most from the workshop?

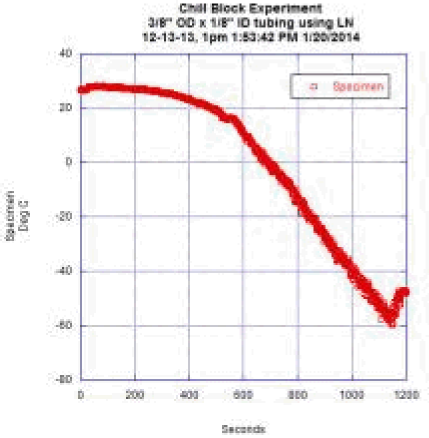
- Workshop provided specific design concepts for internal cooling mechanism

*We needed to flesh out the details of this concept*

# Boise State student worked as summer intern in our lab

Sandia National Laboratories

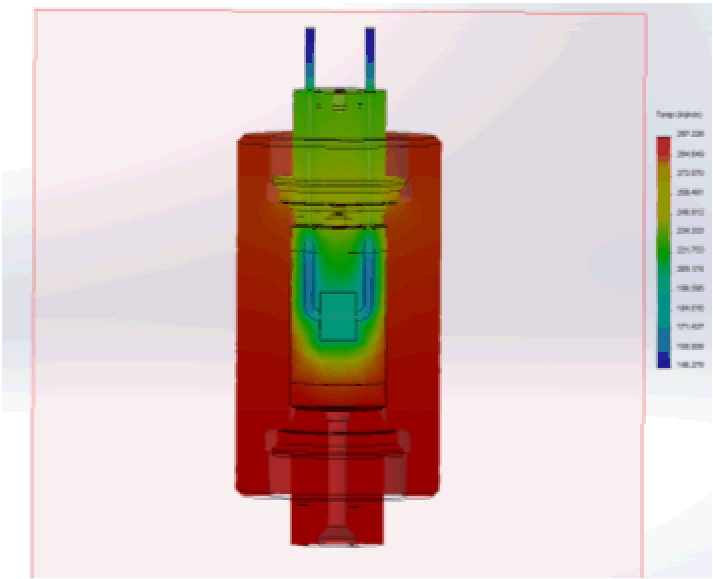
- Chill block cooling prototypes
  - Optimized tube ID for cooling
  - Optimized tube wall thickness to react against external high pressure gas
- ANSYS for thermal analysis
- SolidWorks from mechanical modeling



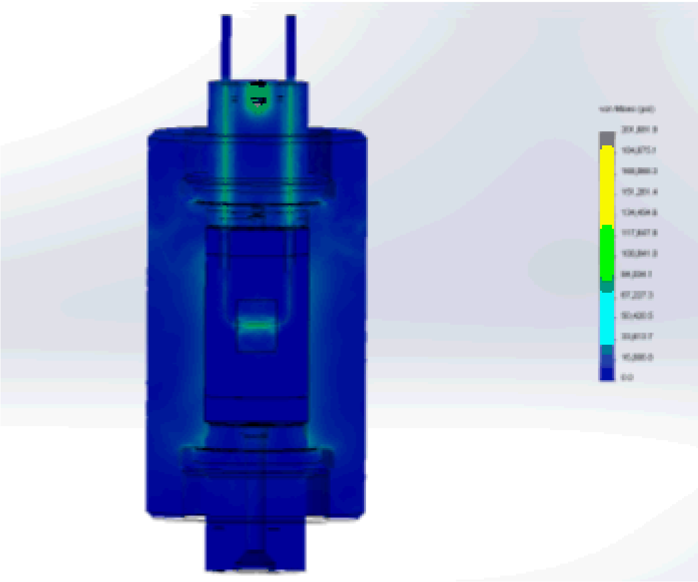

*Prototyping validated cooling mechanism and model inputs*

# Final model constructed

- Empirical data from chill block test used to calibrate thermal model



Pressure vessel
thermal gradient at -50º C



Combined thermal and
pressure stress
-50º C at 138 MPa (20,000 psi)

*Design specifications for vessel finalized*

# Start engaging manufacturers

- Partnerships valuable for development and procurement of pressure vessel
- Pressure vessel design
  - Communications started with Autoclave Engineering
  - Autoclave has provided a cost estimate
- Chiller
  - Received cost estimate

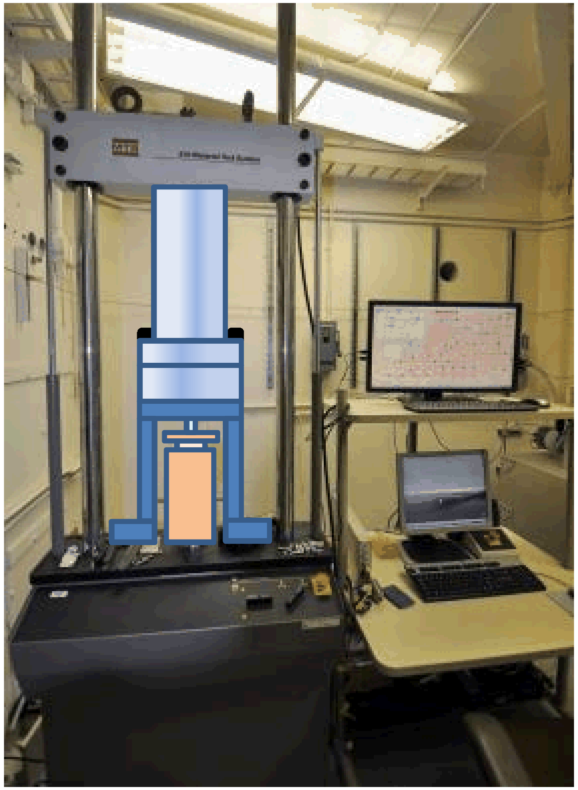
*Obtaining estimate was enabled by design tools, workshop information, prototyping and modeling*

# Conclusion

- Most hardware is in place
  - Test cell
  - Test frame
  - Test controller and software
  - Manifold hardware
  - Manifold control software operational

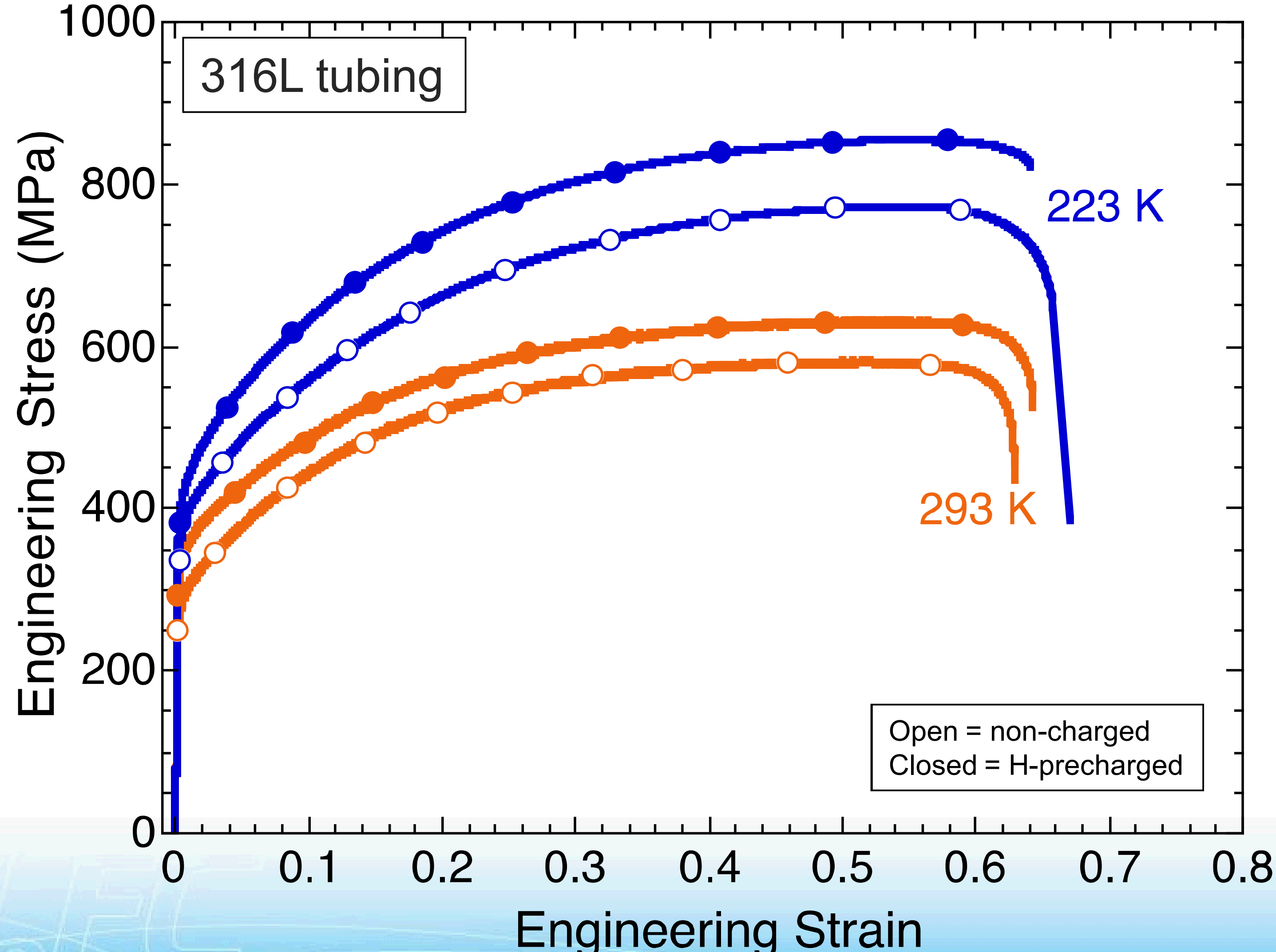
- Vision: dynamic material testing
  - At -50 °C (-58 °F), 138 MPa (20,000 psi)
  - At 200 °C (392 °F), 138 MPa (20,000 psi)

- Remaining hardware needed
  - Variable Temperature Pressure Vessel: $120 K
  - Industrial Chiller : $20K-$180K

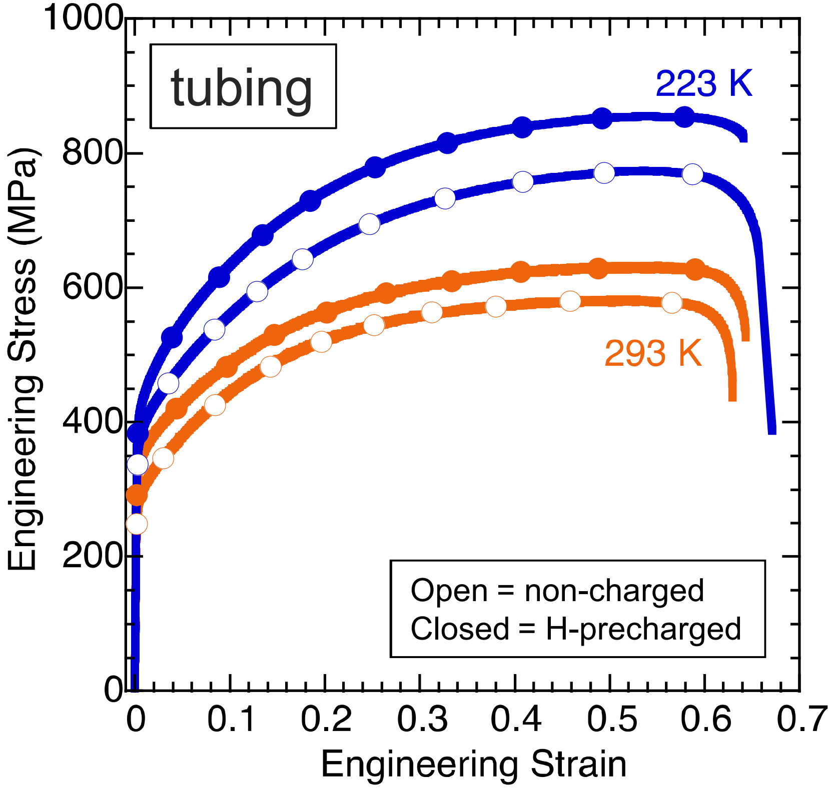


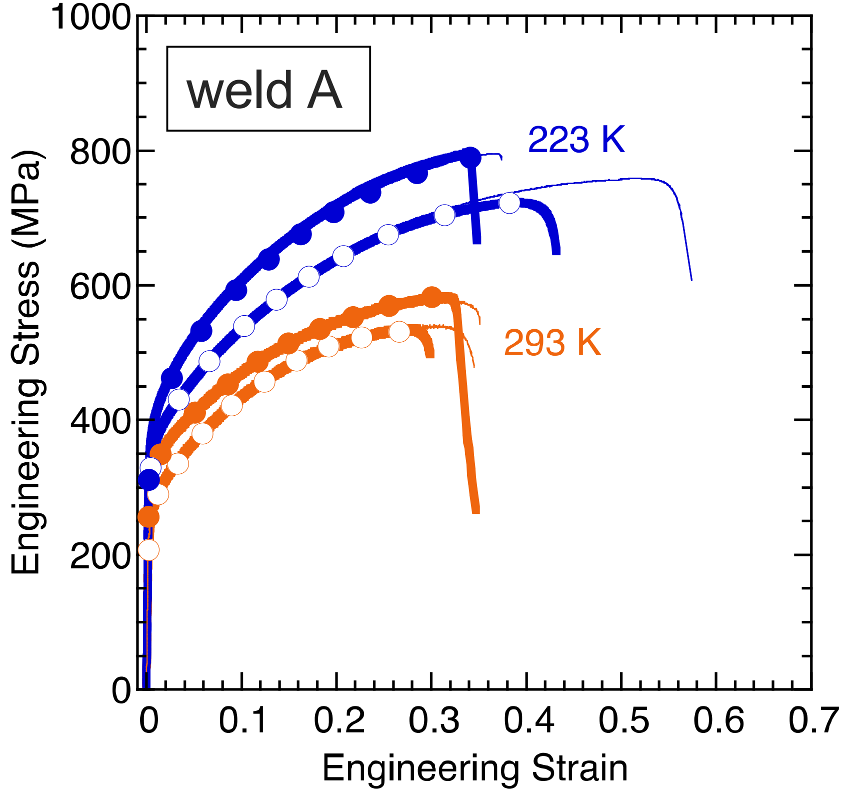
*Now ready for fabrication of qualified vessel*

# Type 316L tubing show similar tensile properties with and without hydrogen



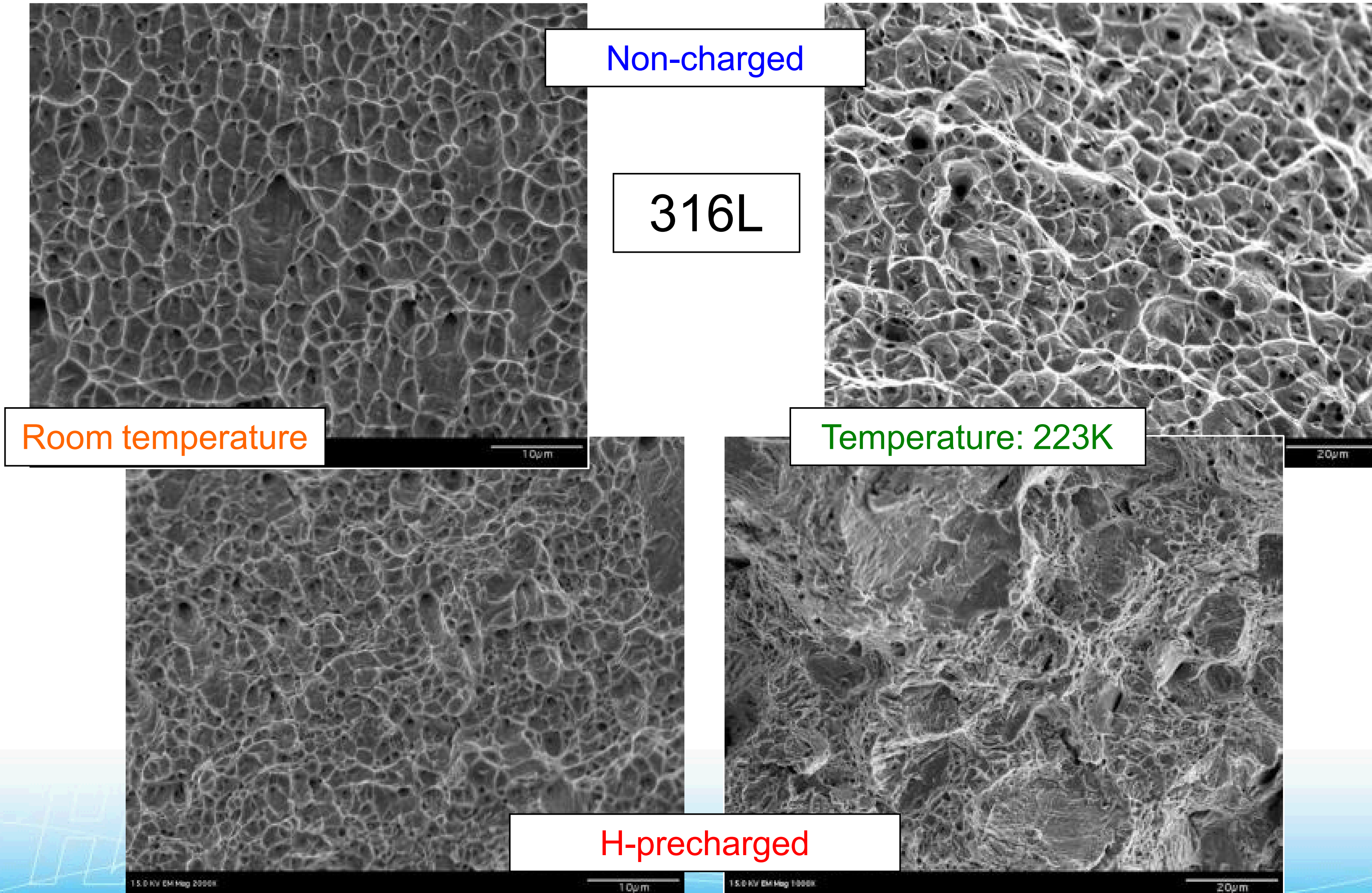
# Type 316L tubing and welds show similar tensile properties with and without hydrogen



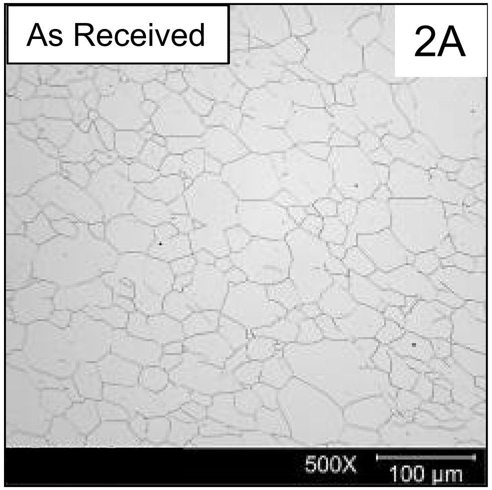



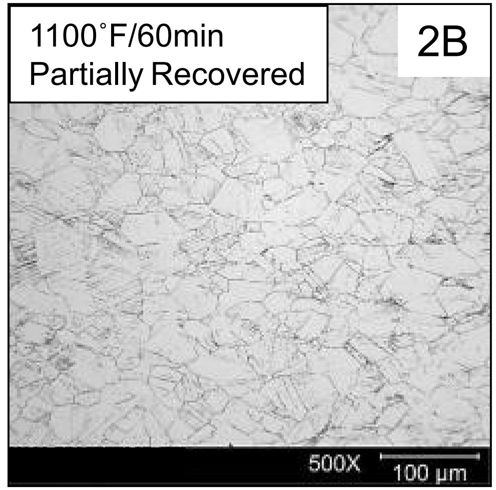

- Welded specimens show less elongation because deformation is restricted to the weld zone (RA is similar for tubing and welds)
- Welded specimens show more variability in elongation than tubing without welds

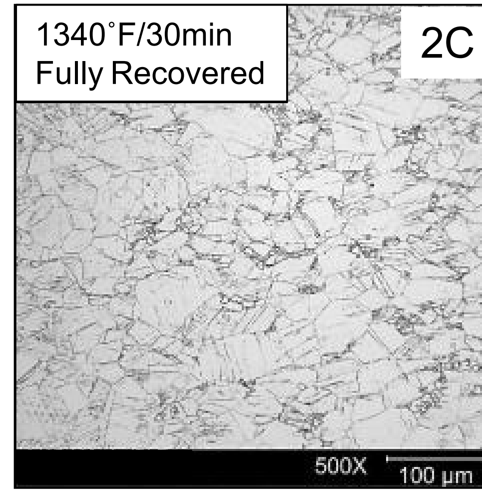
# Fractography of tubing is similar to base materials

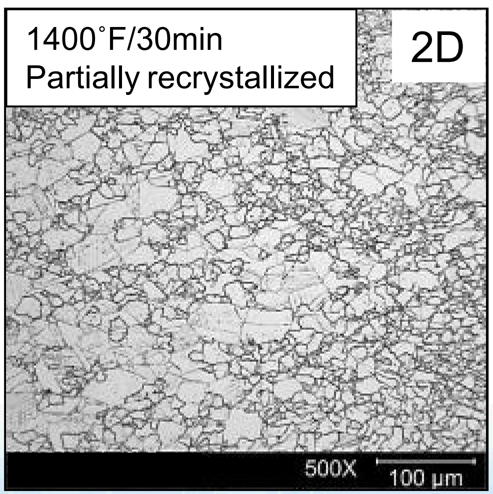


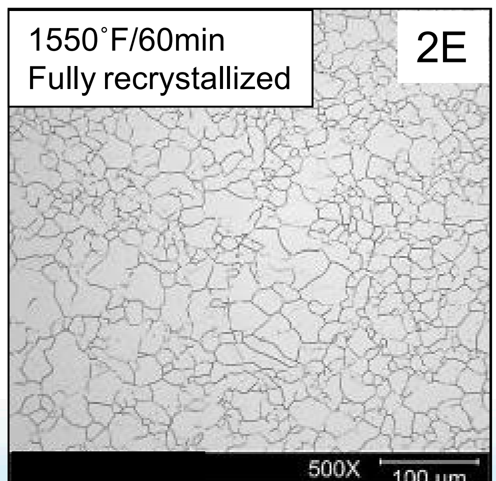
# Microstructures of thermally treated tubing (304L no.2)

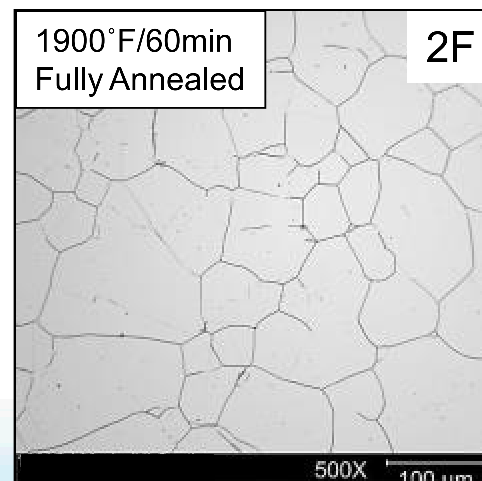


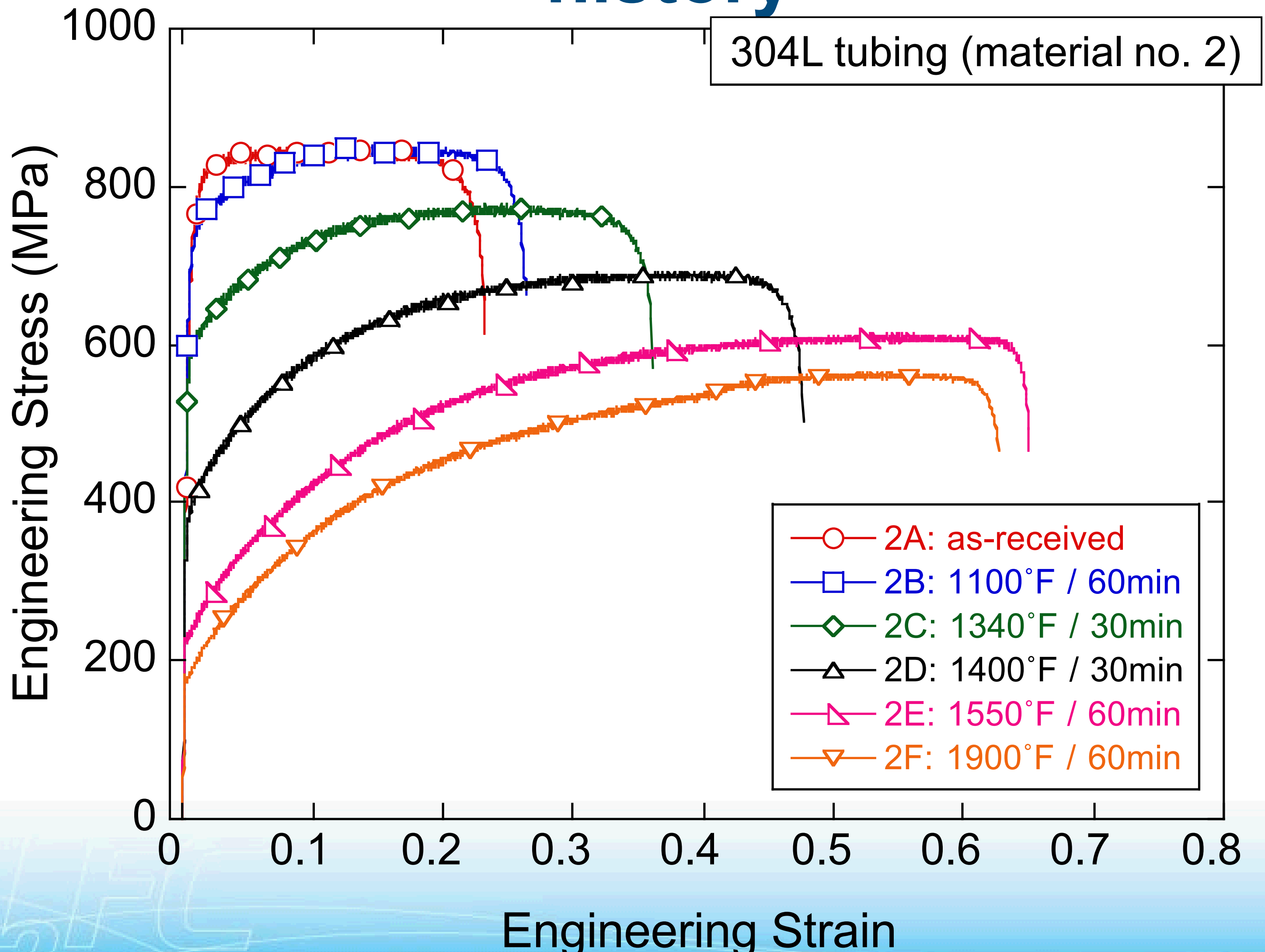



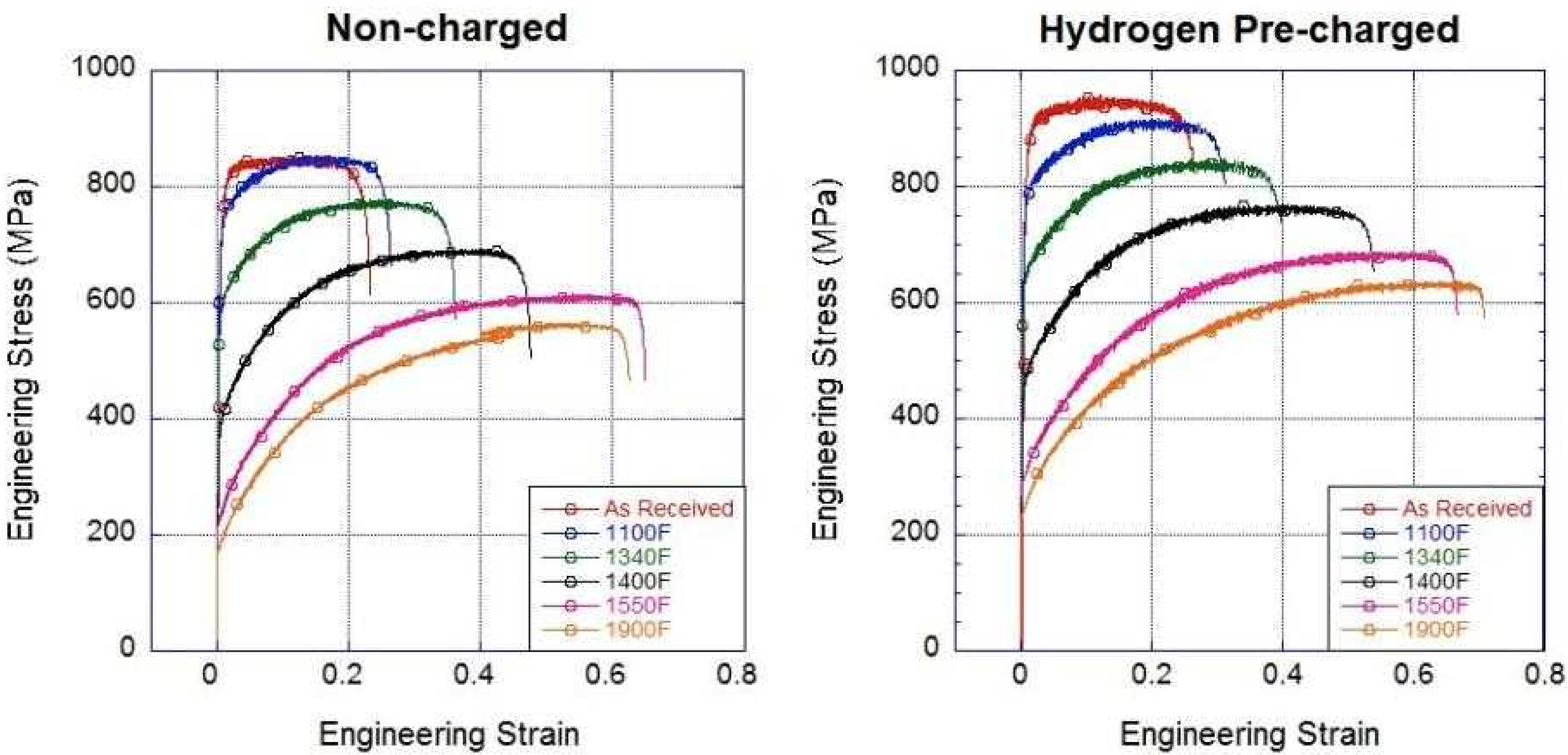




Significant microstructural change is evident after annealing

# Tensile flow curves depend on thermal history

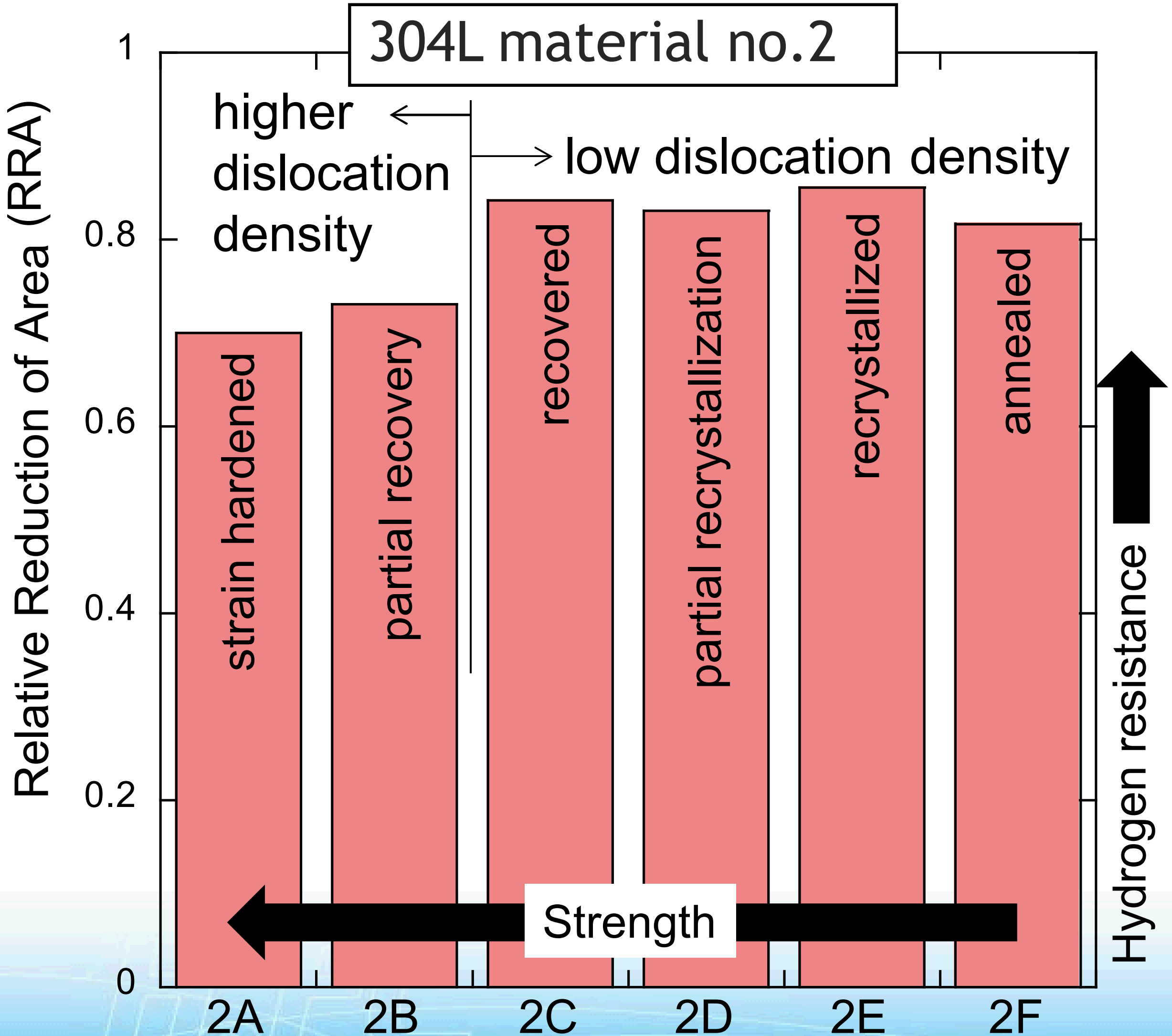


# Hydrogen has little influence on tensile flow curves




304L tubing (material no. 2)

# Strength and microstructure show only modest effect on hydrogen resistance

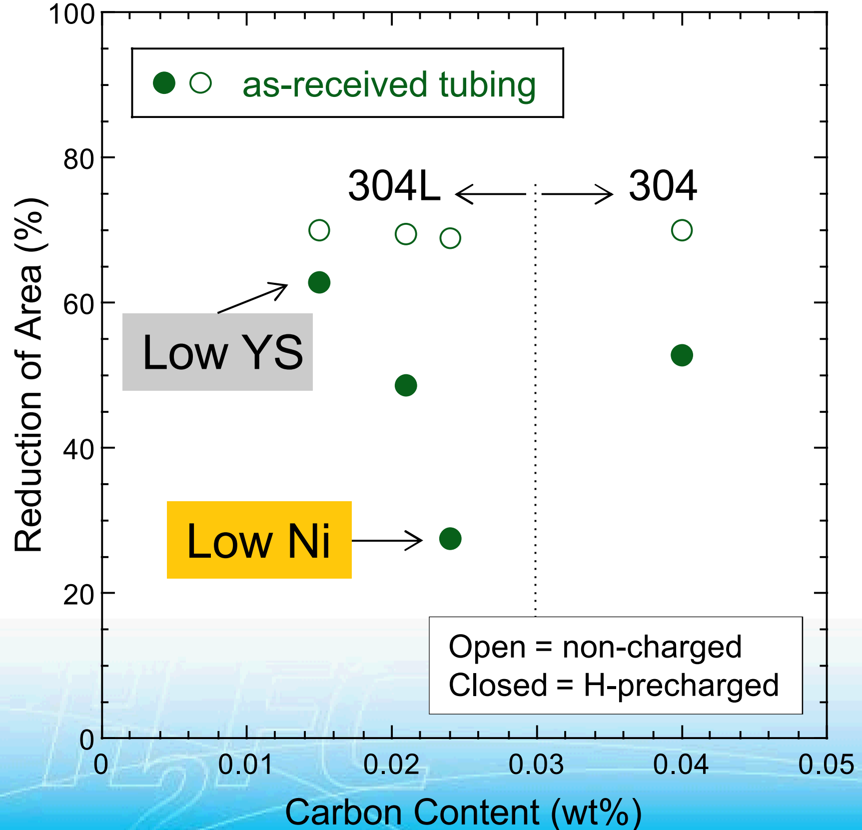



- Hydrogen resistance dependence on yield strength is relatively small for strength in range of 180 to 700 MPa
- RRA > 0.8 for low dislocation microstructures
- RRA ~ 0.7 for high dislocation microstructures

RRA = RA(H)/RA(air)

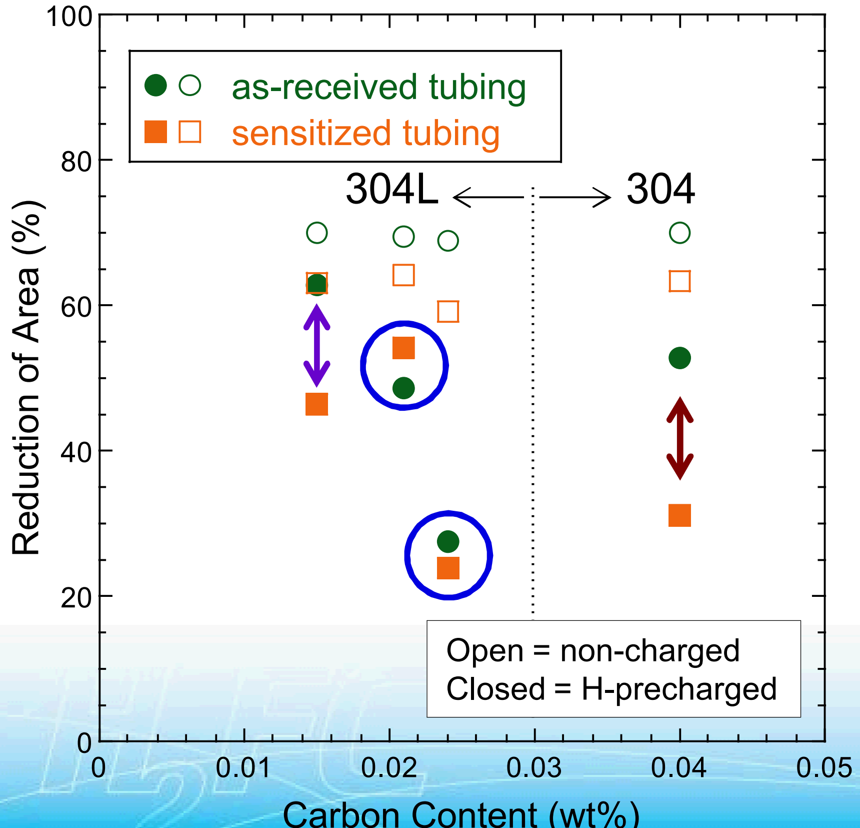
# Carbon content has indirect consequences for hydrogen resistance

- There is no direct trend of hydrogen ductility loss with carbon content
- However, carbon content can indirectly influence the effects of H



# Carbon content has indirect consequences for hydrogen resistance

- There is no direct trend of hydrogen ductility loss with carbon content
- However, carbon content can indirectly influence the effects of H
- Sensitization evaluates the sensitivity of austenitic stainless steels to microstructural segregation, which affects resistance to H

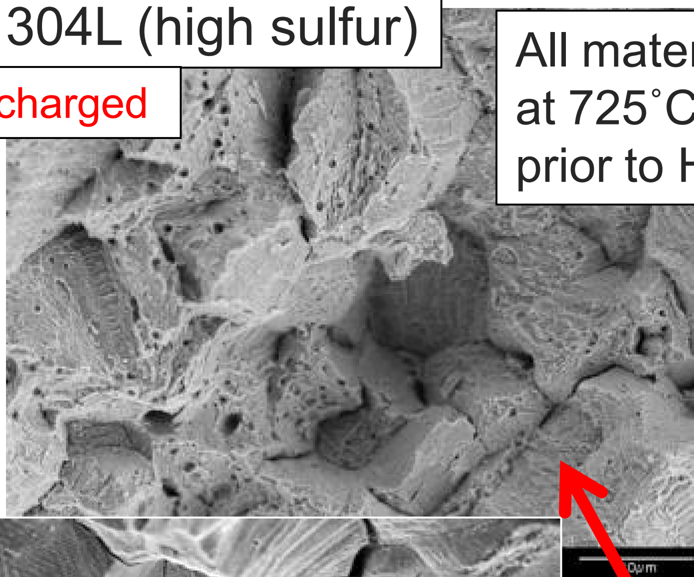



- For type 304L: sensitization does not strongly impact hydrogen effects
- For type 304 (high carbon): sensitization enhances effects of hydrogen
- Effect of high sulfur?
  - Material no. 3 has ~5x more sulfur than the other alloys

# Compositional segregation (S & C) appear to enhance the effects of H

Type 304L (high sulfur)

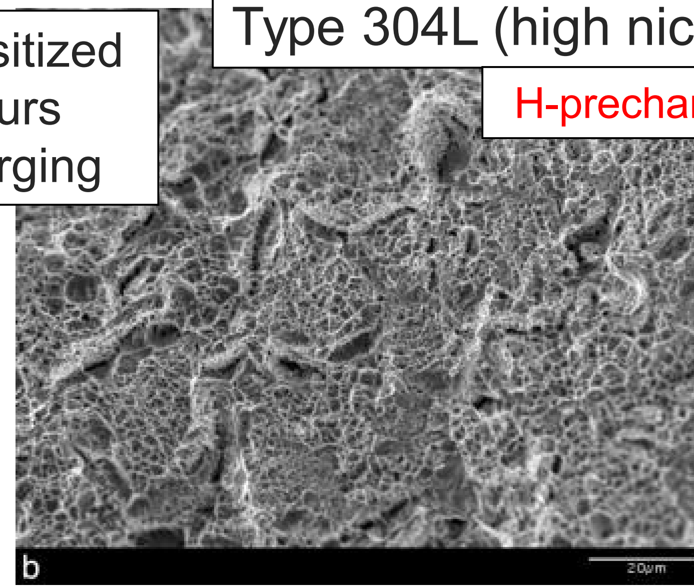
H-precharged

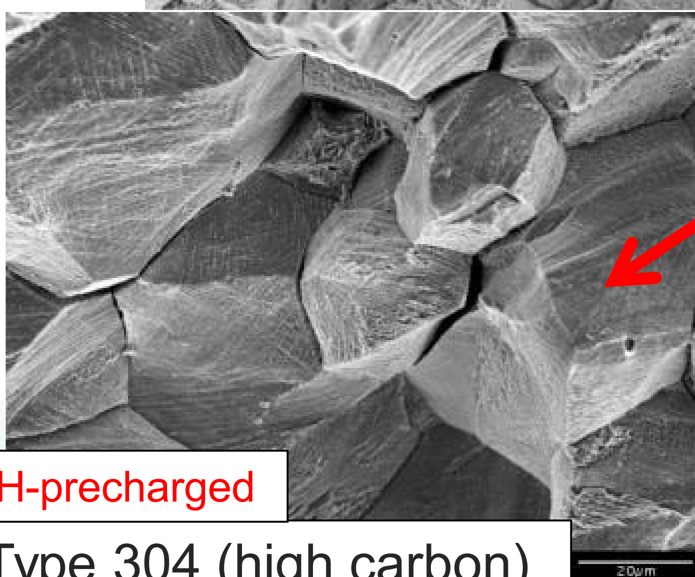


All materials sensitized at 725˚C for 4 hours prior to H-precharging

Type 304L (high nickel)

H-precharged



H-precharged

Type 304 (high carbon)

Undesirable “embrittled” fracture features

- Alloys with high carbon and sulfur show large reductions of ductility
    - Presumably due to the combined effects of segregation (sensitization) and hydrogen