Fracture Toughness Assessment of Hydrogen Pipelines

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Materials Innovations in an Emerging Hydrogen Economy

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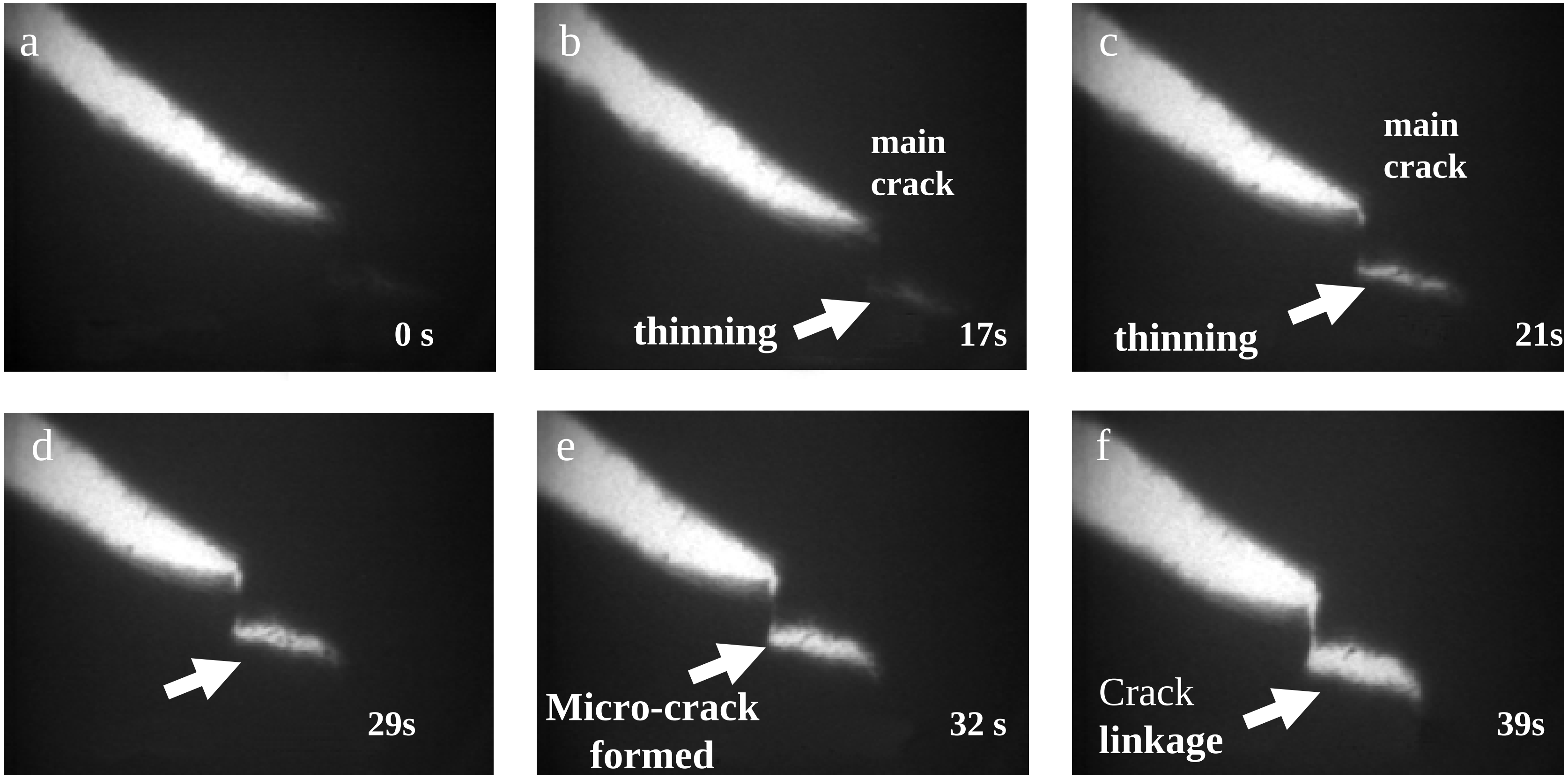
Hydrogen-Induced Crack Propagation

We do not understand the relationship between macroscopic parameters (e.g. applied load and pressure) and the operating microscopic degradation mechanism

Static crack in vacuum. Hydrogen gas introduced →



No load increase is needed for the crack to grow

Hydrogen Embrittlement Mechanisms

- **Several candidate mechanisms have evolved over the years each of which is supported by a set of experimental observations and <u>strong personal views</u>**

- **Viable mechanisms of embrittlement**
  - **Stress induced hydride formation and cleavage**
    - **Metals with stable hydrides (Group Vb metals, Ti, Mg, Zr and their alloys)**
    - **Supported by experimental observations**
  - **Hydrogen enhanced localized plasticity (HELP)**
    - **Increased dislocation mobility, failure by plastic deformation mechanisms**
    - **Supported by experimental observations**
  - **Hydrogen induced decohesion**
    - **Direct evidence is lacking**
    - **Supported by First Principles Calculations (DFT)**

- **Degradation is often due to the synergistic action of mechanisms**

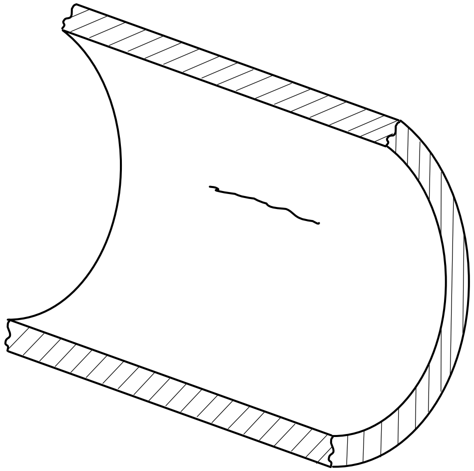
Embrittlement and Phenomenology

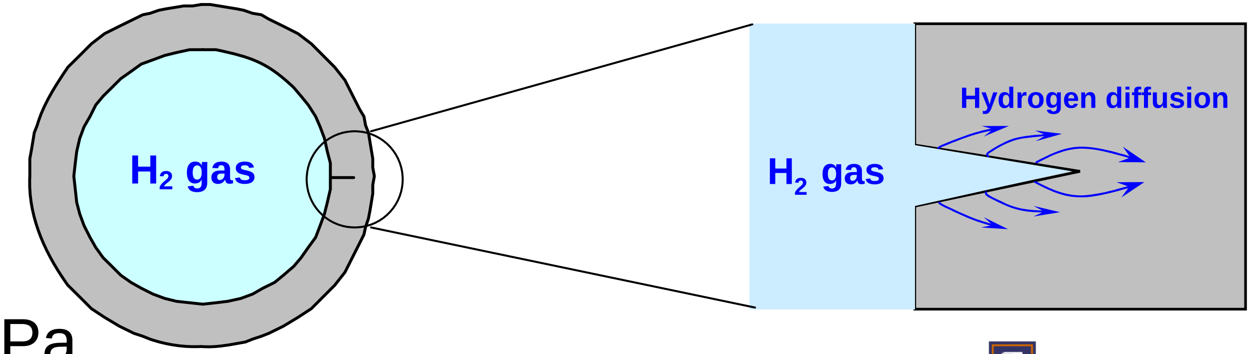
- Fractographic evidence suggests that low strength steels under static loading fail by
  - Hydrogen-assisted transgranular fracture induced by void or microcrack initiation through decohesion at internal interface (precipitate/inclusion or phase boundaries) ahead of a crack or notch accompanied by shear localization (HELP) leading to the linking of the void/microcrack with the tip of the crack
  - Fracture is controlled by yield strength level and microstructure
- Our contention, which needs to be verified through experiment, is that embrittlement
  - Under static load is a result of the synergistic action of the HELP and decohesion mechanisms
  - Under cyclic load can be intergranular (extremely dangerous mode of failure)

Fracture Mechanics Approach to Design of Steel Pipelines Transporting Hydrogen

To characterize embrittlement we need to understand the interaction of hydrogen with the elastoplastic deformation of the material at a crack tip

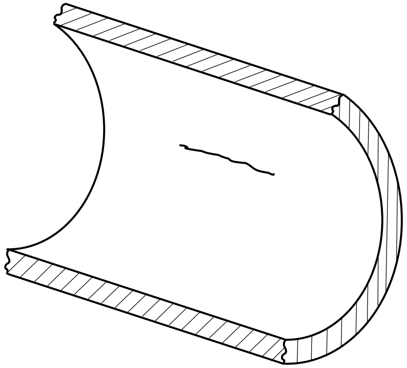
Objective: Determine stress, deformation, and hydrogen concentration fields
in the neighborhood of
an axial crack in a steel pipeline



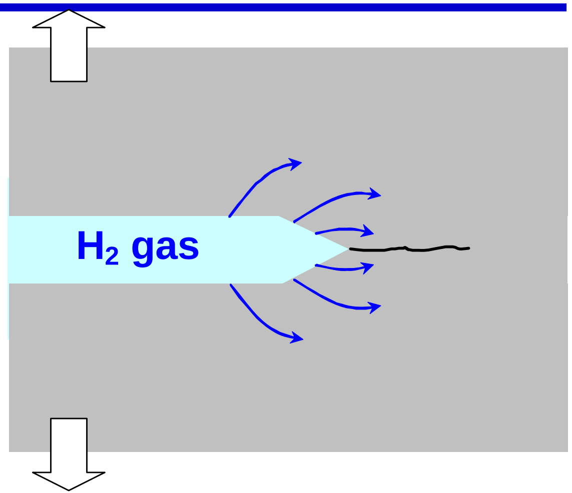


H_2-Pressure of 15MPa

Fracture Mechanics Approach to Design of Pipelines Actual-Pipeline Solution vs Laboratory-Specimen Solution

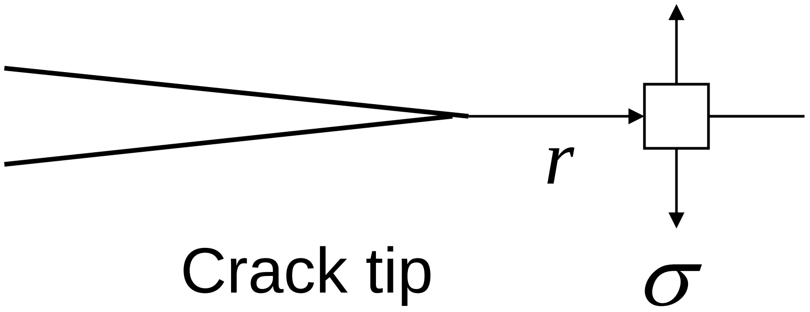


Is there a similarity between the full-field (pipeline) solution and that at laboratory specimens?



If yes, we conjecture that parameters which characterize fracture in the laboratory specimen can be used to characterize fracture in the pipeline

Subcritical crack growth experiments with WOL specimen carried out at Sandia



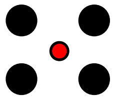
$$\sigma = \frac{K_I}{\sqrt{2\pi r}}$$

If K_I characterizes fracture in the specimen, can it be used to characterize fracture in the pipeline in the presence of hydrogen?

Tranferrability

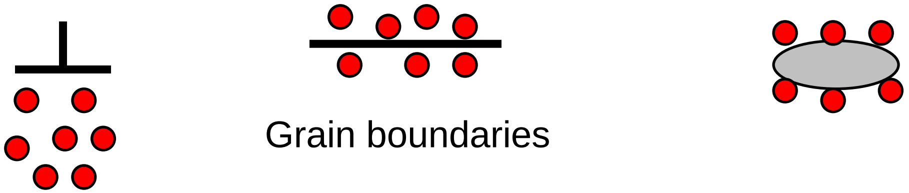
Hydrogen Transport Analysis

- **Diffusing hydrogen resides at**
  - **Normal Interstitial Lattice Sites (NILS)**

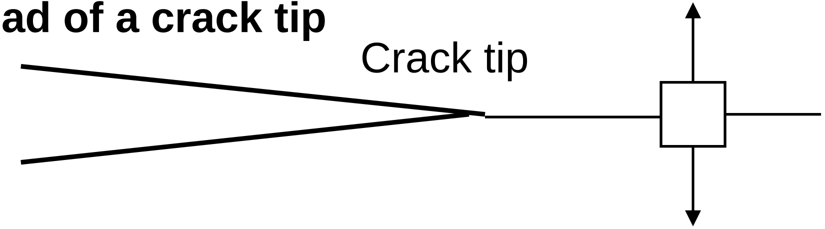


C_L

  - **Trapping Sites** C_T
    - **Microstructural heterogeneities such as dislocations, grain boundaries, inclusions, voids, interfaces, impurity atom clusters**



- **Diffusing hydrogen interacts with stresses and strains**
  - **Hydrogen dilates the lattice and thus interacts with hydrostatic stress**
    - **Moves from regions under compression toward regions under tension, e.g ahead of a crack tip**



$$\sigma > 0$$

  - **Hydrogen enhances dislocation mobility, thus it facilitates plastic flow**

- **As hydrogen diffuses stresses and strains change. At the same time local stresses and strains affect the diffusion paths. So the problem is coupled**

Cracked Pipeline: Problem Statement

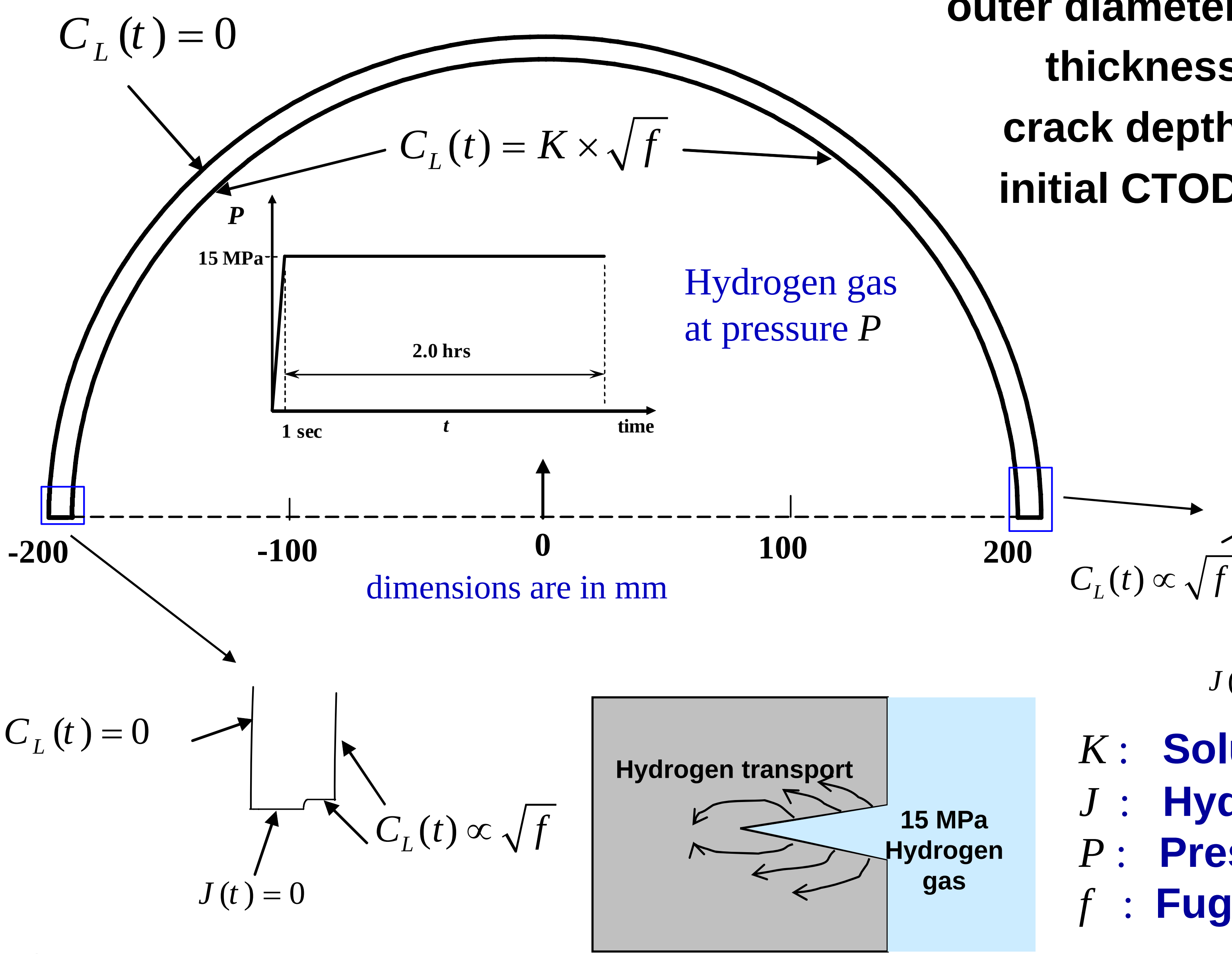
outer diameter: 40.64 cm

thickness: $h = 9.52$ mm

crack depth: $a = 1.9$ mm

initial CTOD: $b_0 = 1.5$ μm

$a / h = 0.2$



K : **Solubility**

J : **Hydrogen flux**

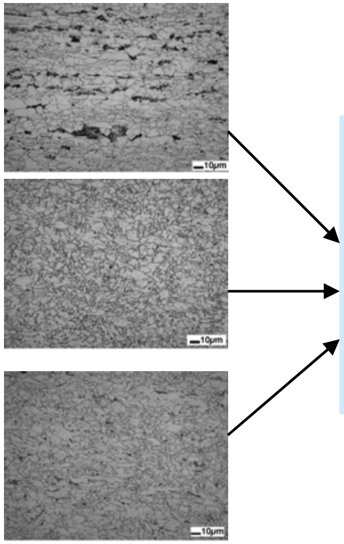
P : **Pressure**

f : **Fugacity**

Materials Characterization

- **Microstructural characterization: Optical, SEM, and TEM studies**
  - Existing pipeline steel samples provided by Air Liquide and Air Products.
  - New micro-alloyed steels (new microstructures) provided by Oregon Steel Mills through DGS Metallurgical Solutions, Inc.

SECAT



	API/ Grade	C	Mn	Si	Cu	Ni	V	Nb	Cr	Ti
A	X70	0.08	1.53	0.28	0.01	0.00	0.050	0.061	0.01	0.014
B	X70/80	0.05	1.52	0.12	0.23	0.14	0.001	0.092	0.25	0.012
C	X70/80	0.04	1.61	0.14	0.22	0.12	0.000	0.096	0.42	0.015
D	X52/60	0.03	1.14	0.18	0.24	0.14	0.001	0.084	0.16	0.014

Typical natural gas pipeline steel
Ferrite/acicular ferrite
Ferrite/acicular ferrite
Ferrite/low level of pearlite

- **Establish the diffusion characteristics of existing and new pipeline steel microstructures**

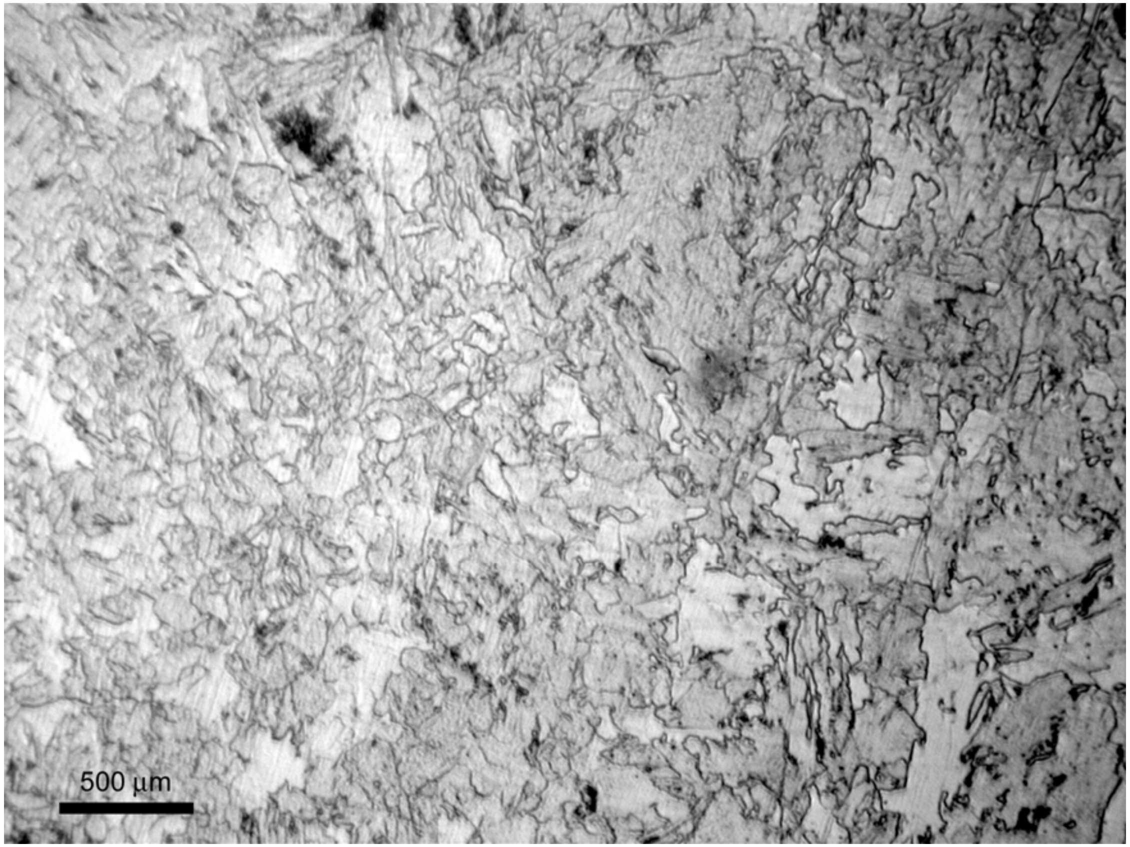
- **Determine uniaxial tension macroscopic flow characteristics in the presence of hydrogen**

- **Carry out fracture testing: Collaboration with Sandia, Livermore**
  - Fracture surfaces, particle, dislocation, and grain boundary characterization

Optical Analysis of New “Steel C” Microstructure

API Grade	C	Mn	Si	Cu	Ni	V	Nb	Cr	Ti
X70/80	0.04	1.61	0.14	0.22	0.12	0.000	0.096	0.42	0.015

Ferrite/acicular ferrite

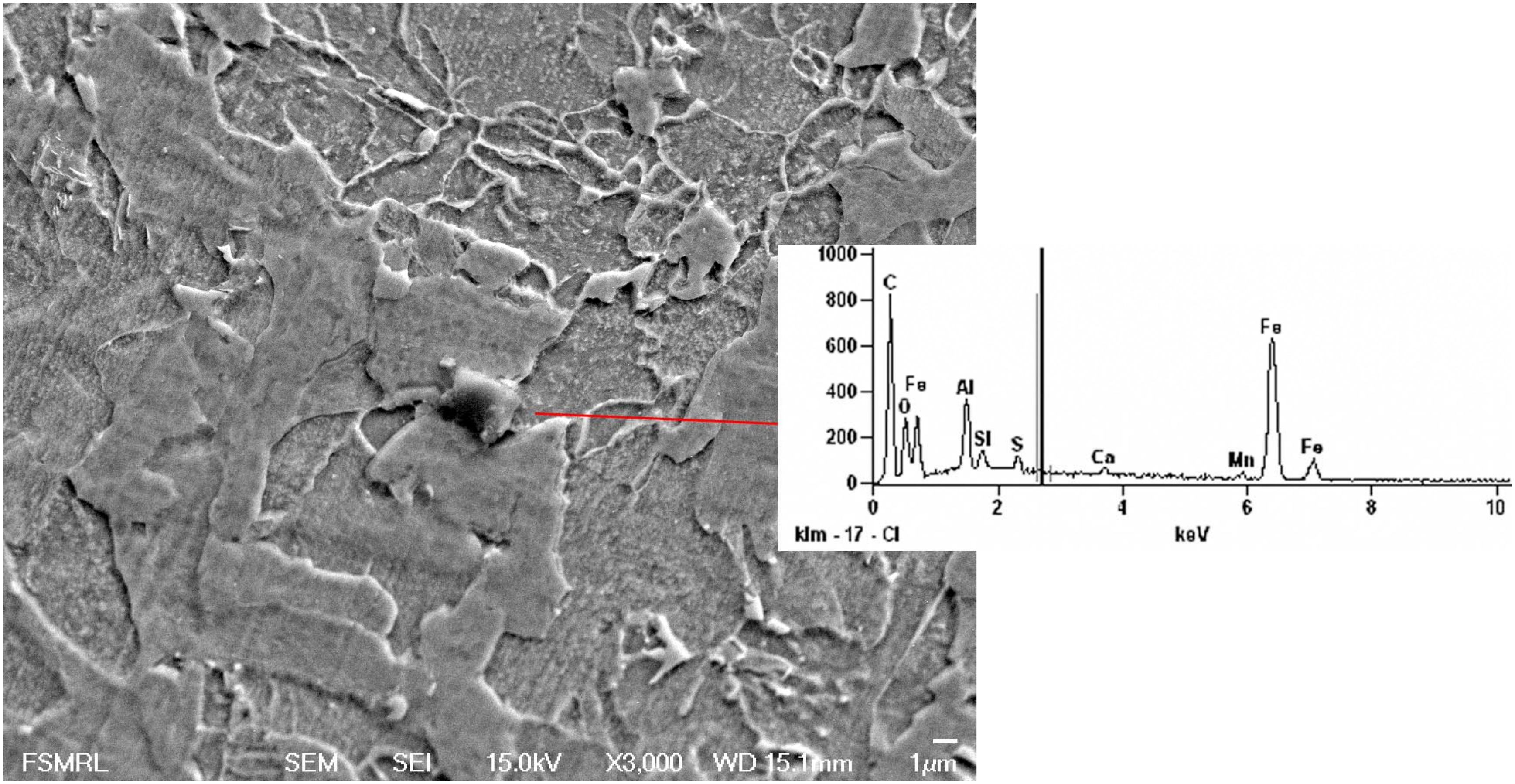


Average grain size :35 µm

3% pearlite

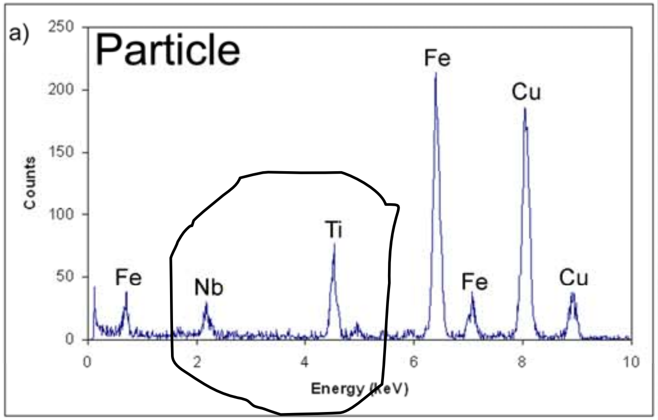
Demonstrated to be good in the presence of H_2S sour service natural gas applications

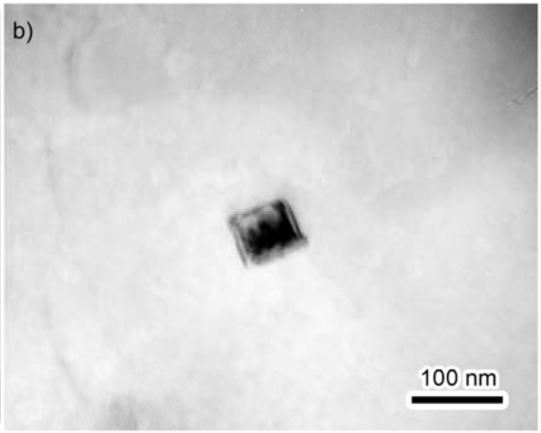
SEM analysis of New “Steel C” Microstructure

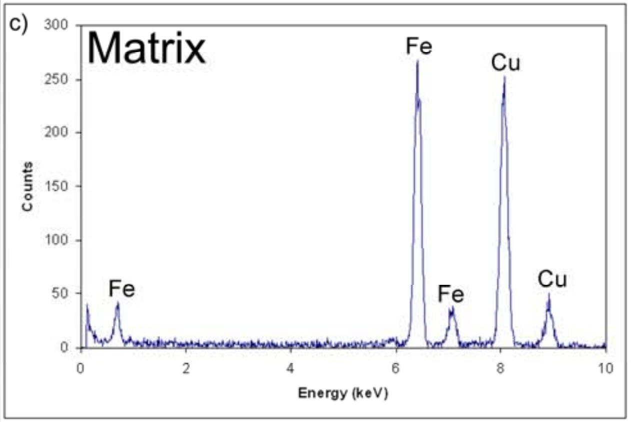


Al rich particle, most likely a sulfide

TEM analysis of New “Steel C” Microstructure





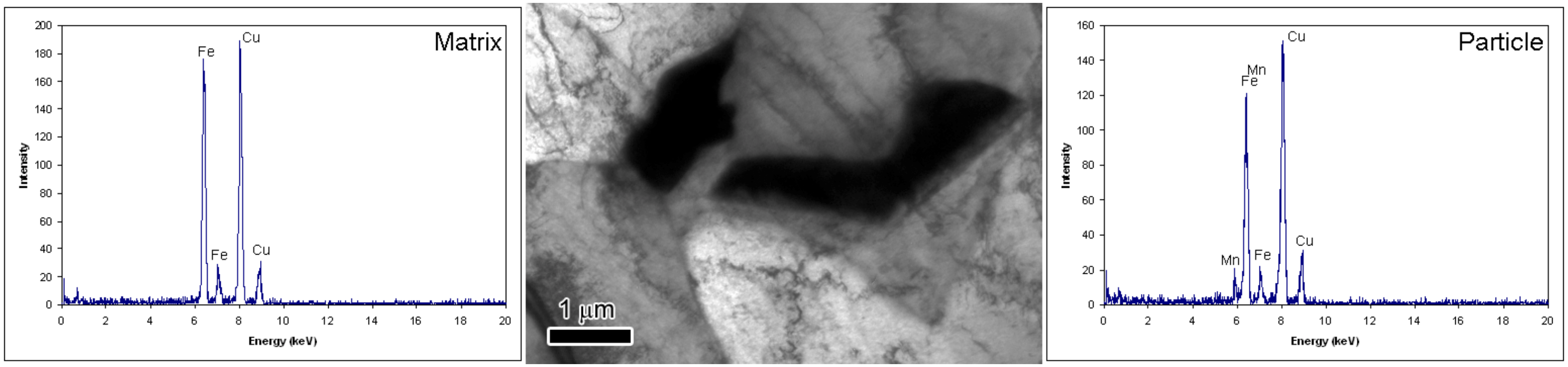


- a) EDS spectrum from particle
- b) Bright field TEM image of typical rectangular particle
- c) EDS spectrum from matrix

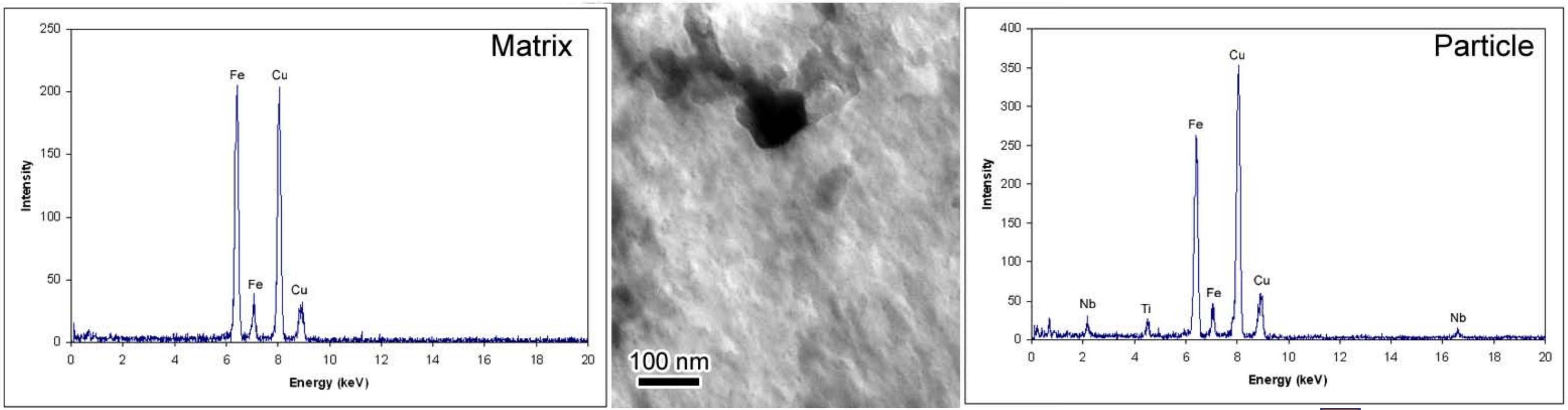
- EDS analysis of fine precipitate inside ferrite grain suggests that precipitate is composed of Ti and Nb

(window detector: C, N, O not detected)

TEM analysis of Air Liquide Steel Microstructure

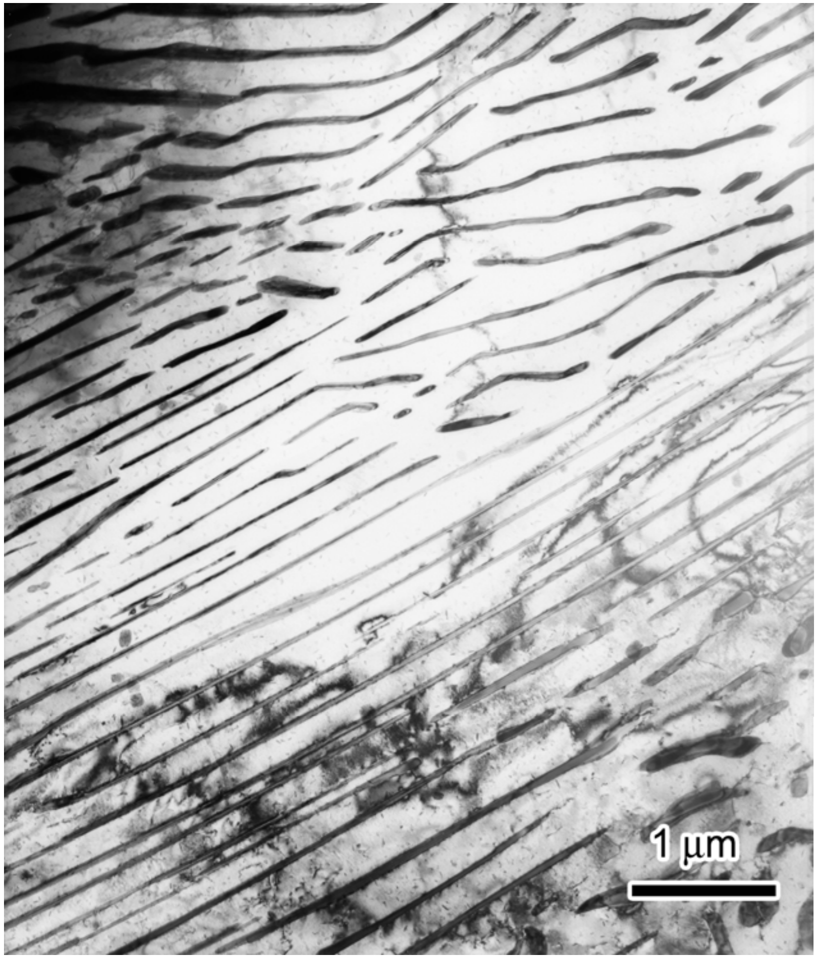


Large intergranular particles (cementite)



Small intragranular particles (carbides with Nb and Ti)

TEM analysis of Air Products Steel Microstructure



Pearlite colonies.

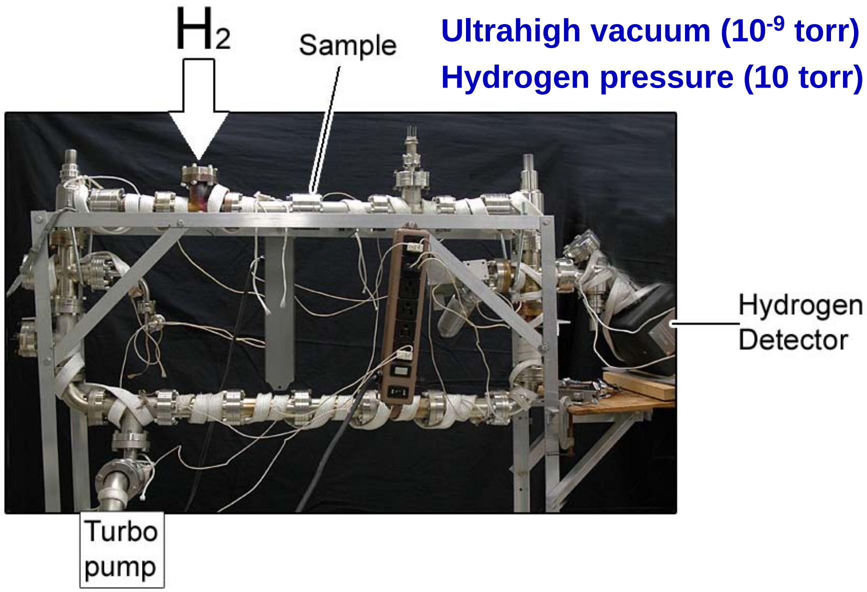
Left: cementite plate arrangement

Right: cross-section of platelets

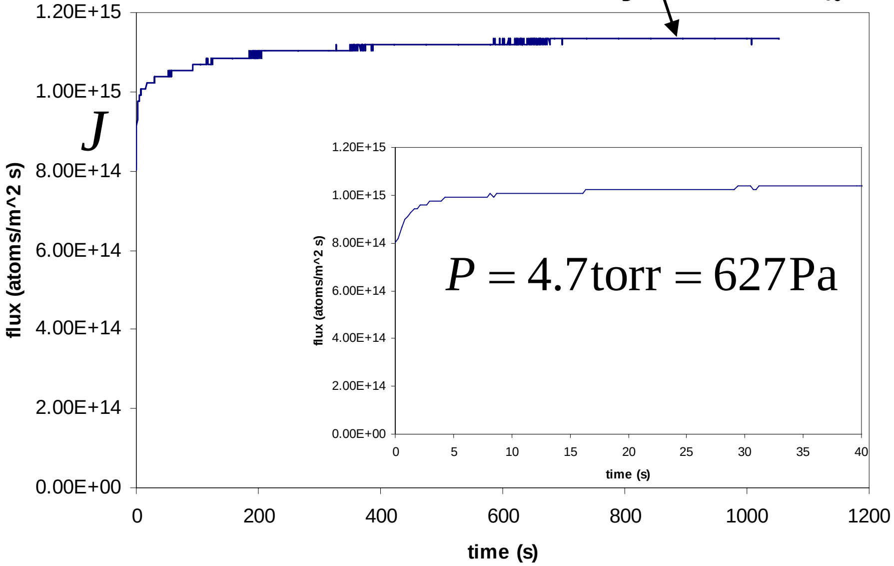
Hydrogen Permeation Measurements

Ultrahigh vacuum (10^{-9} torr)

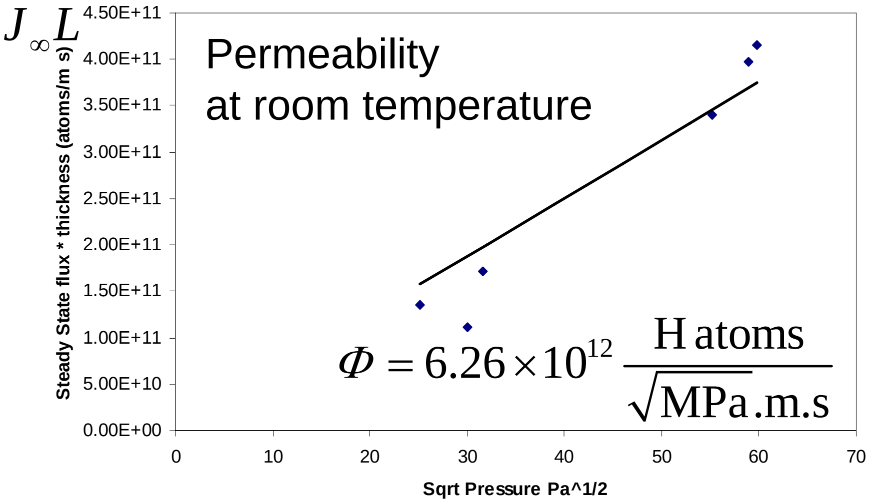
Hydrogen pressure (10 torr)



Steady state: J_∞

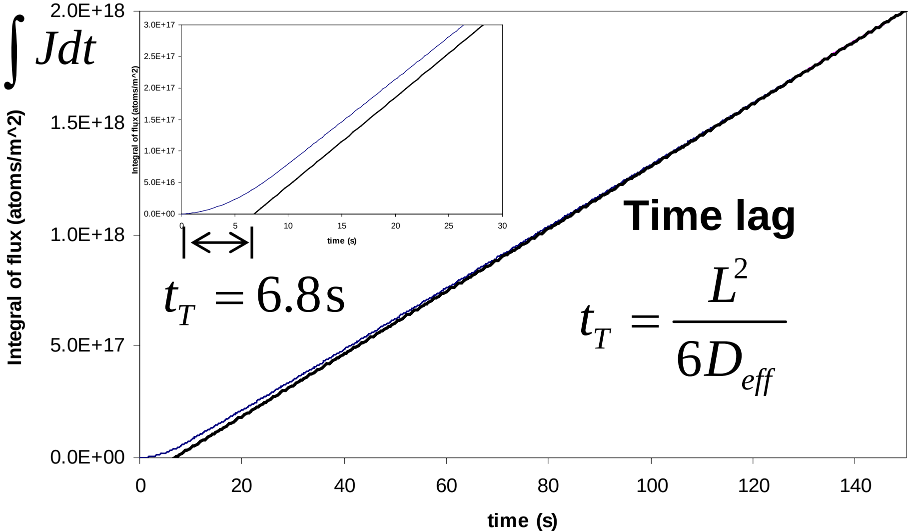


$$P = 4.7\,\text{torr} = 627\,\text{Pa}$$



Permeability at room temperature

$$\Phi = 6.26 \times 10^{12} \frac{\text{H atoms}}{\sqrt{\text{MPa}}\,\text{.m.s}}$$



$$t_T = 6.8\,\text{s}$$

Time lag

$$t_T = \frac{L^2}{6D_{eff}}$$

- Oregon Steel Mills sample: thickness $L = 120\,\text{microns}$
- room temperature

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Material: X70/80 acicular ferrite microstructure

$$C = K\sqrt{f} \qquad f = P\exp\left(\frac{Pd}{RT}\right) \qquad d = 15.84\ \text{cm}^3/\text{mol}$$

$$K = 6.54696\times10^{18}\ \frac{\text{H atoms}}{\text{m}^3\sqrt{\text{Pa}}}$$

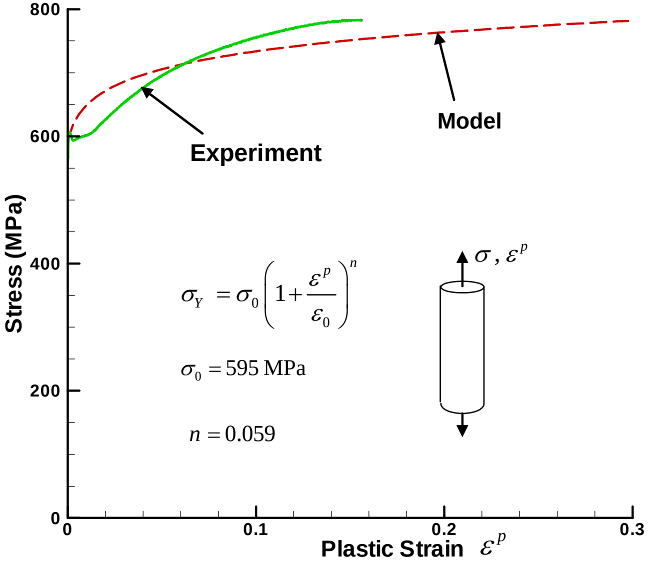
$$C_0 = 2.084\times10^{21}\ \text{H atom}/\text{m}^3 \qquad P = 1\ \text{atm}$$

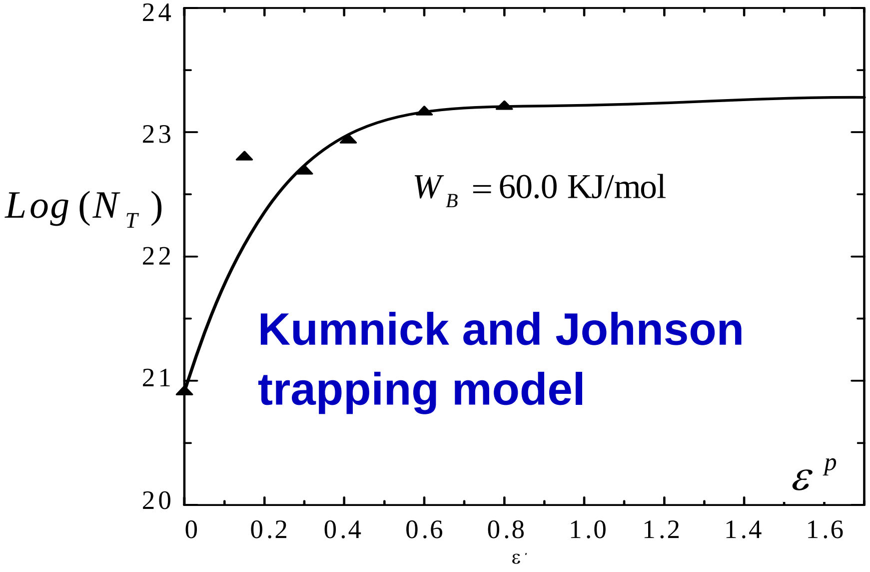
$$C_0 = 2.65932\times10^{22}\ \text{H atom}/\text{m}^3 \qquad P = 15\ \text{MPa}$$

Lattice diffusion coefficient

$$D = 1.271\times10^{-8}\ \text{m}^2/\text{s}$$

Stress-strain





Kumnick and Johnson trapping model

Dislocation trapping modeling

$$N_T = \frac{\sqrt{2}\rho}{a} \qquad W_B = 20.2\ \text{KJ/mol}$$

$$\rho = \begin{cases} \rho_0 + \dfrac{\gamma}{0.15}\varepsilon^p & \varepsilon^p \le 0.15 \\ \text{const.} & \varepsilon^p > 0.15 \end{cases}$$

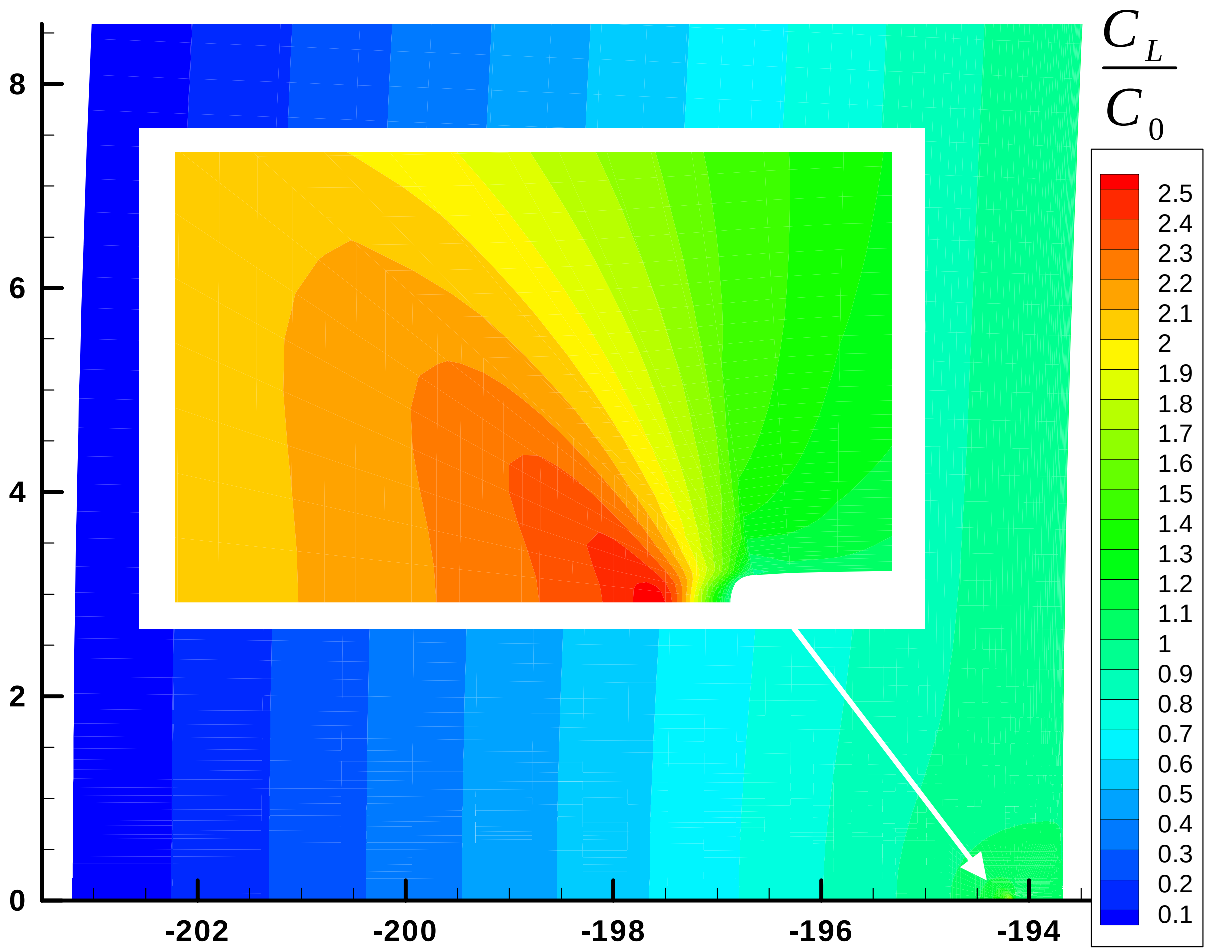
$$\rho_0 = 10^{10}\,m^{-2},\ \gamma = 10^{16}\,m^{-2}$$

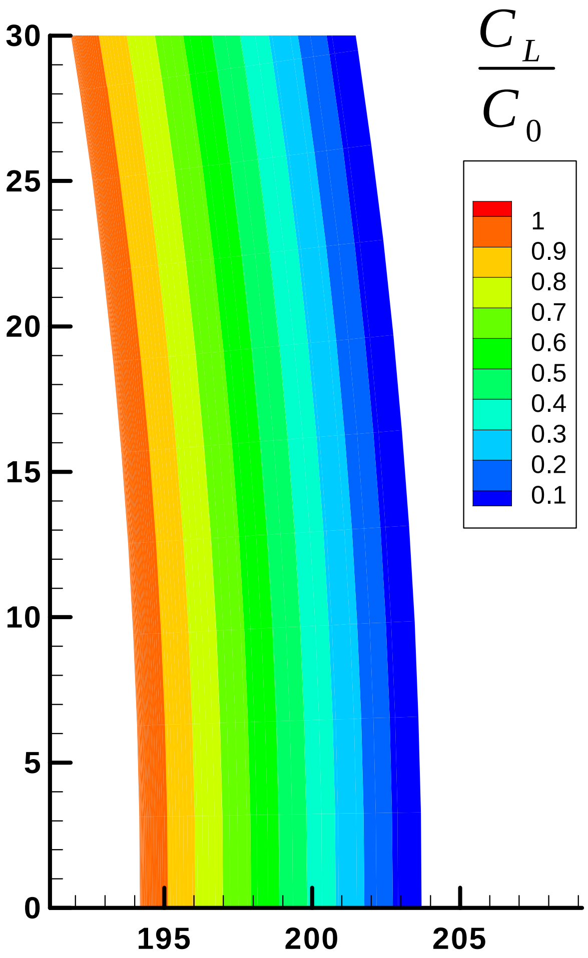
Lattice Hydrogen Concentration at Steady State

Kumnick and Johnson trapping model

Time to steady-state: 2.0 hrs

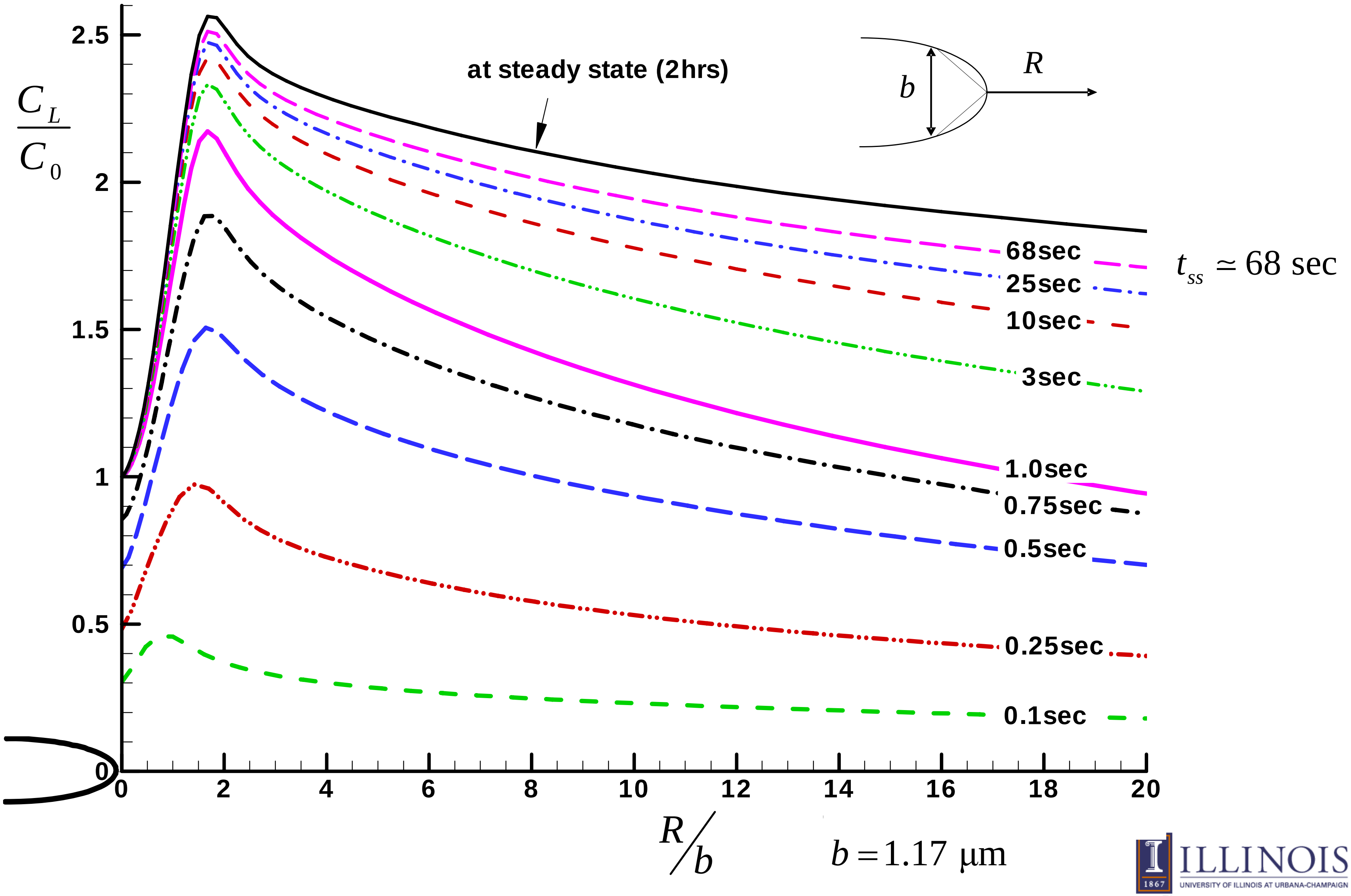
$$t_{ss} = 68\,\text{sec}$$





$C_0 = 2.65932 \times 10^{22}$ H atom / m^3 $\quad P = 15$ MPa

Evolution of Hydrogen Concentration at NILS

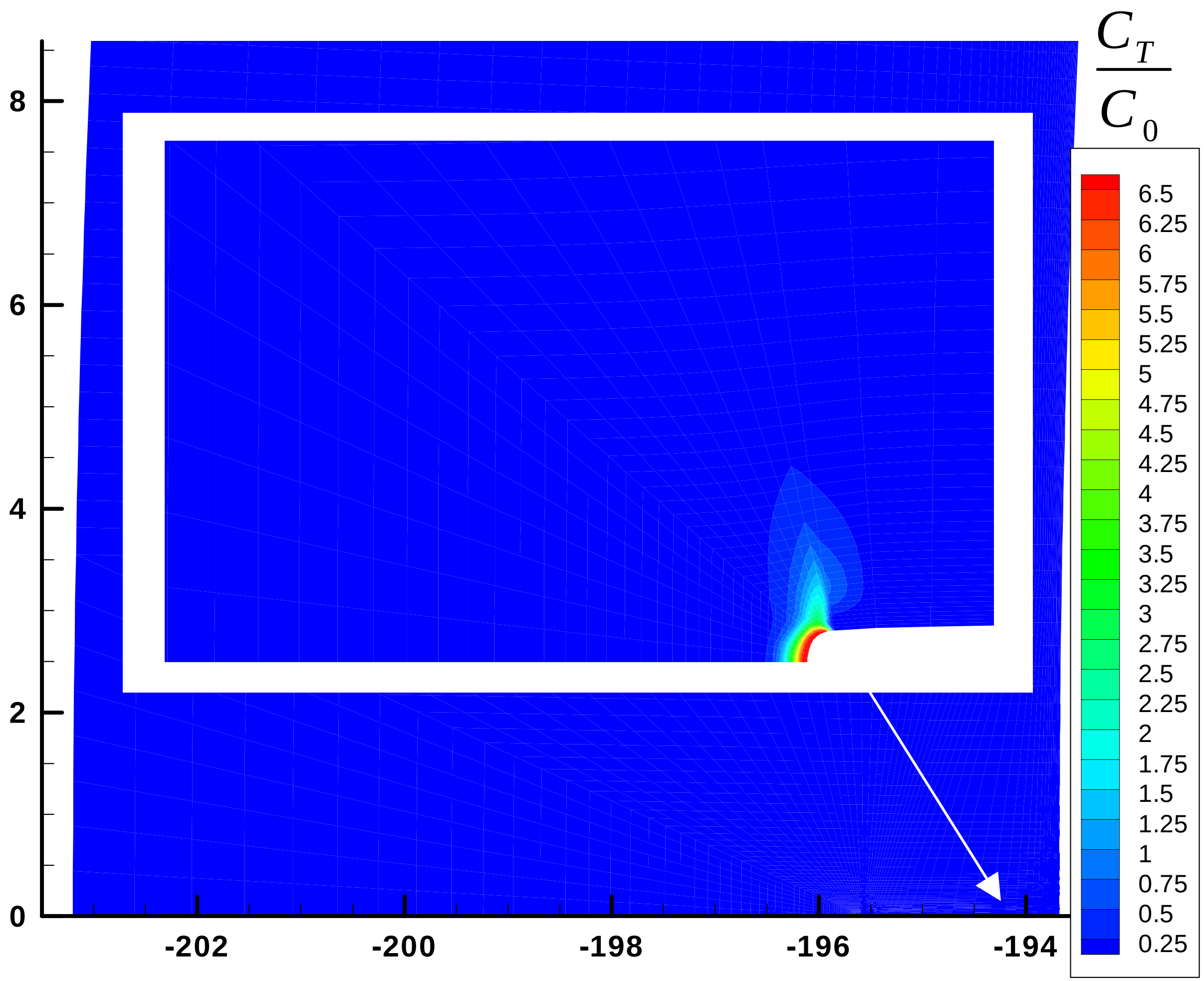


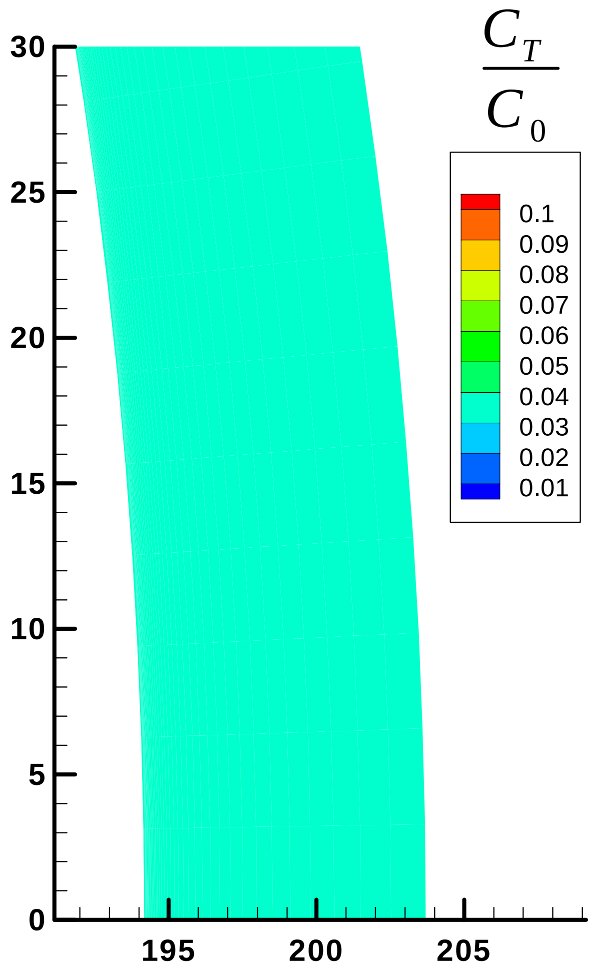
$t_{ss} \simeq 68$ sec

$b = 1.17\ \mu\text{m}$

Trapped Hydrogen Concentration at Steady State

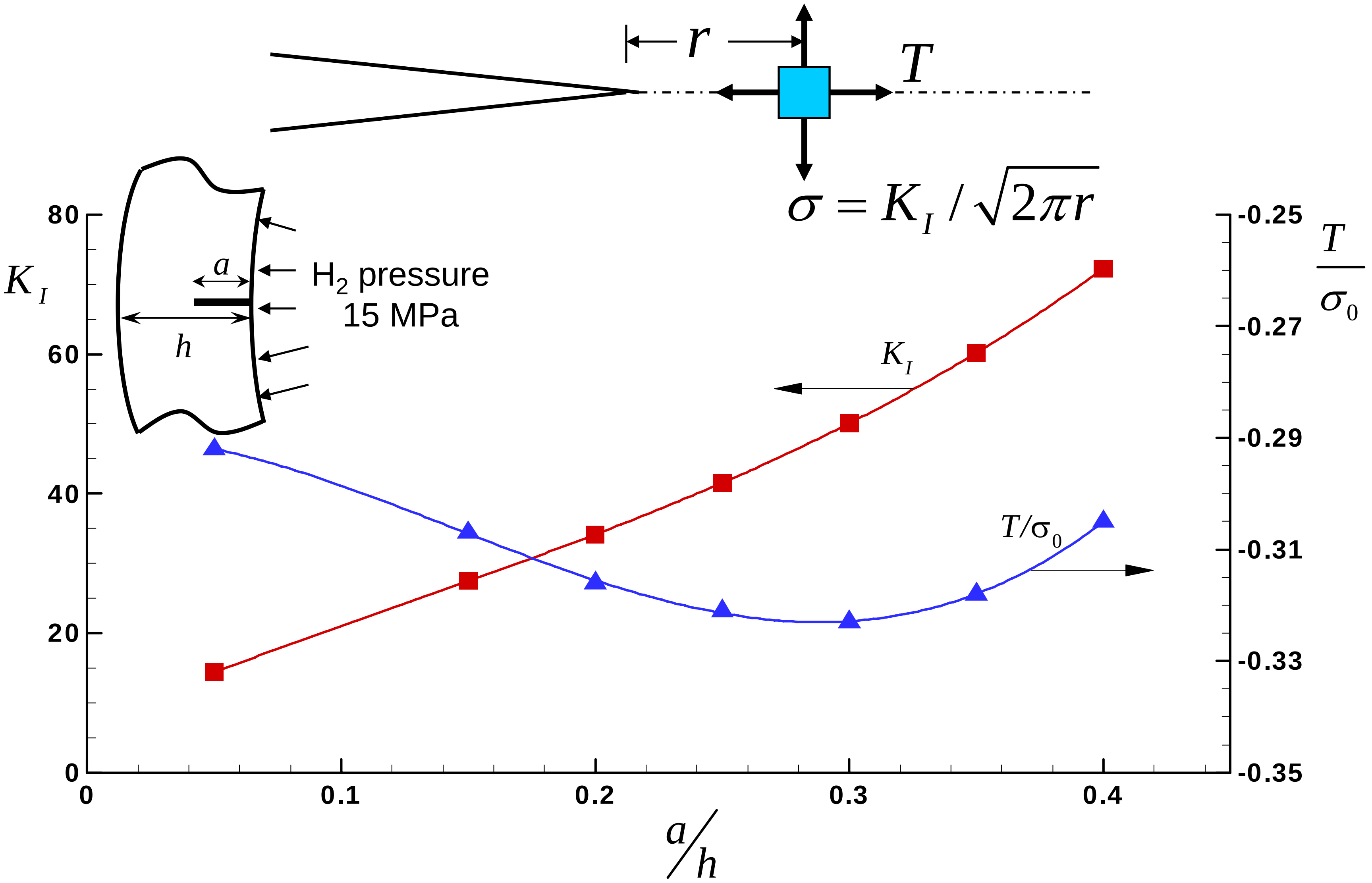
Kumnick and Johnson trapping model





$C_0 = 2.65932 \times 10^{22}\ \text{H atom}/\text{m}^3 \qquad P = 15\ \text{MPa}$

Fracture Mechanics Parameters
From the Full Pipeline to the Laboratory Specimen

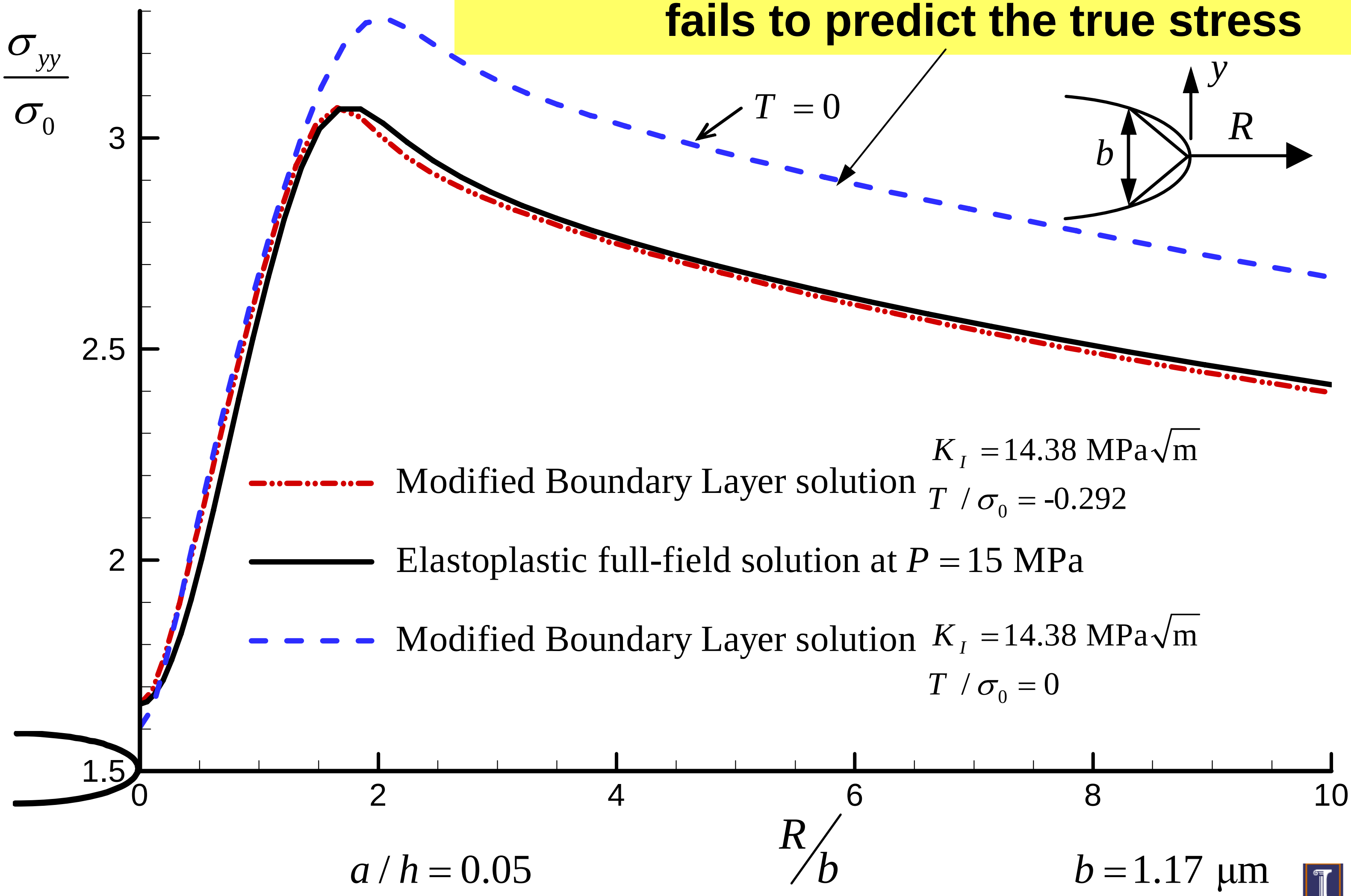


$$\sigma = K_I / \sqrt{2\pi r}$$

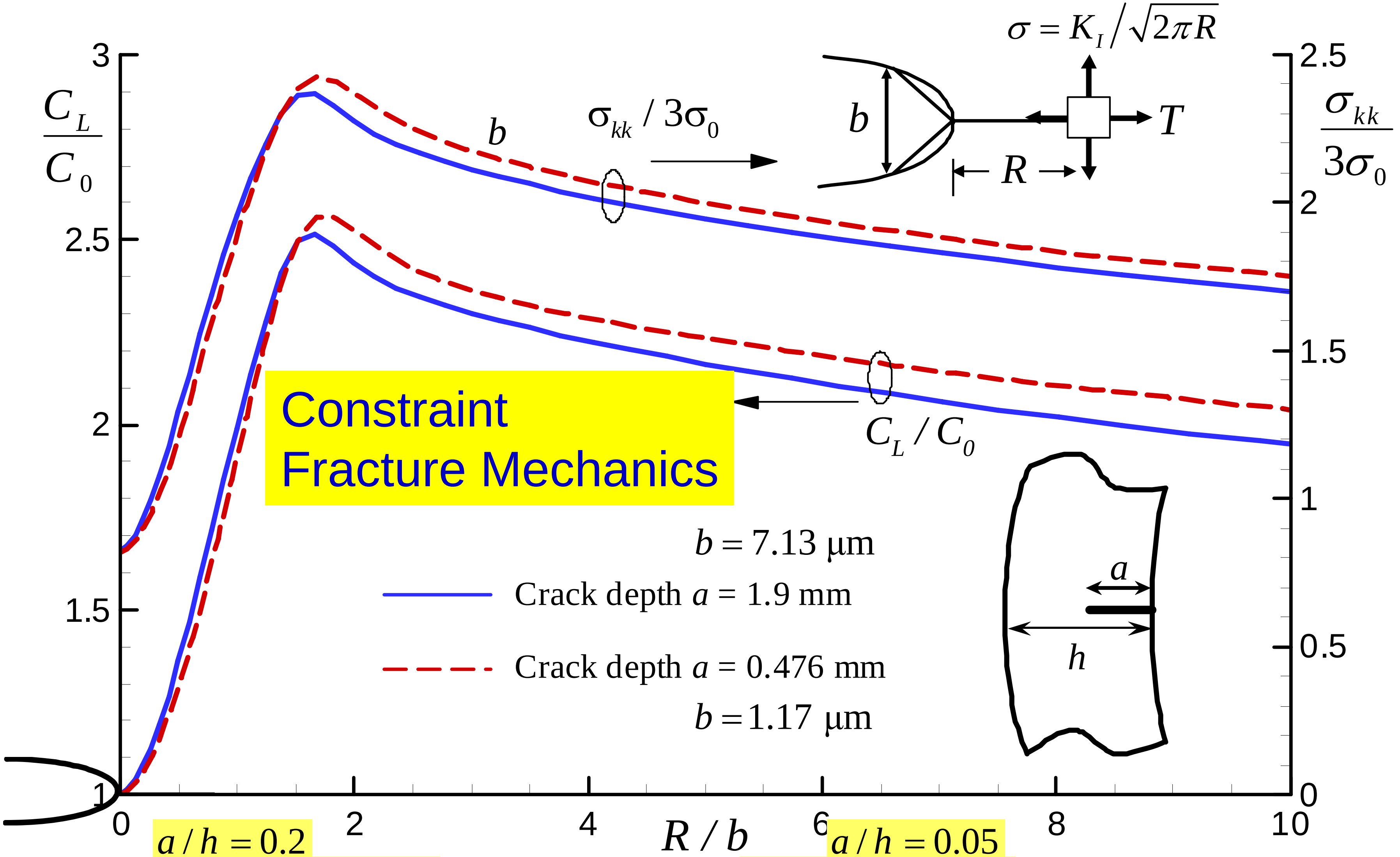
Crack depth/pipe thickness

Full Field (pipeline) vs Boundary Layer Solution (laboratory specimen)

Neglecting the *T* -stress in the MBL formulation fails to predict the true stress



Crack-Tip Fields Scale with K_I and T-stress Independence from Crack Depth



Constraint Fracture Mechanics

$a / h = 0.2$
$K_I = 34.12$ MPa$\sqrt{\text{m}}$
$T / \sigma_0 = -0.316$

$a / h = 0.05$
$K_I = 14.38$MPa$\sqrt{\text{m}}$
$T / \sigma_0 = -0.292$

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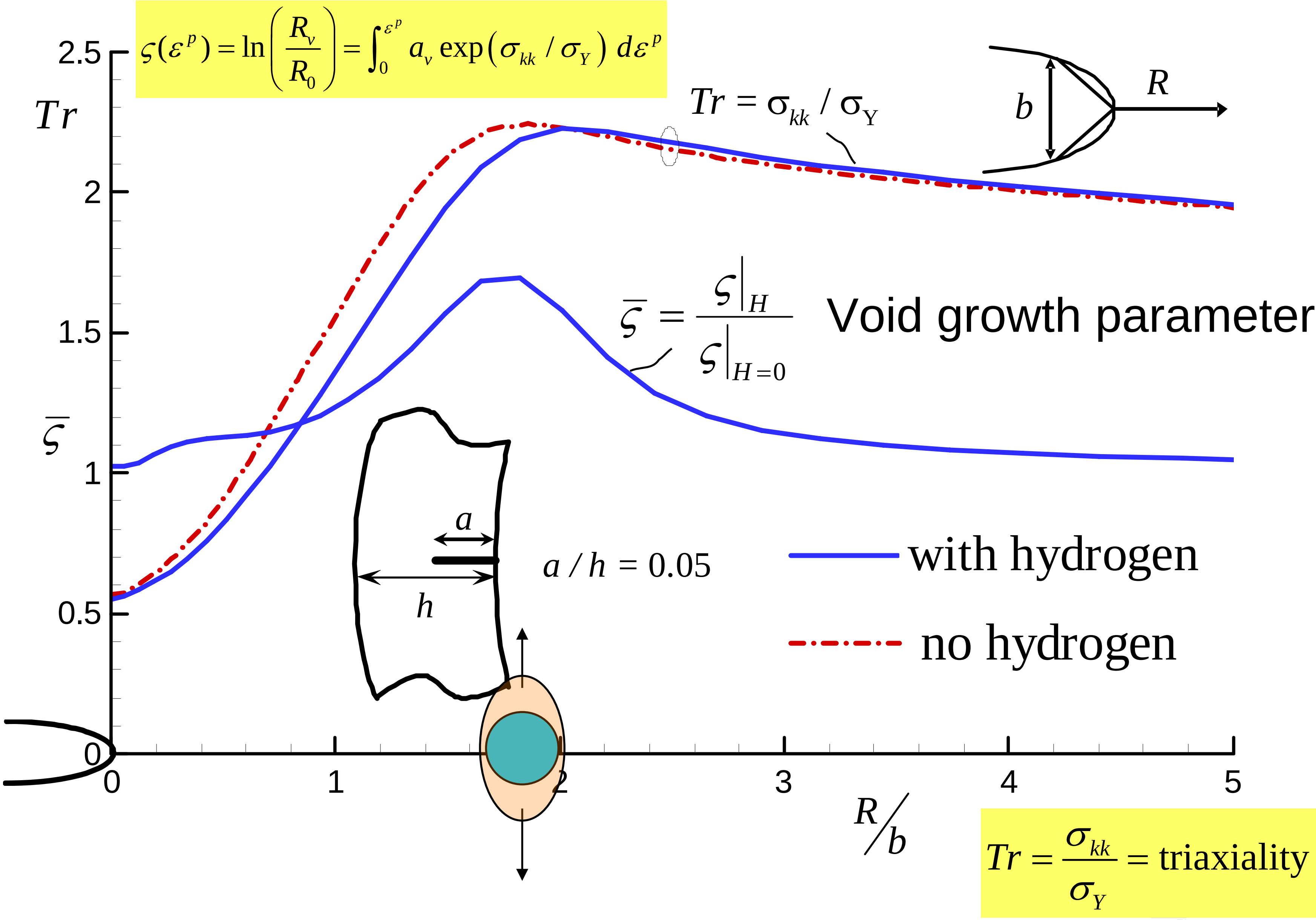
Hydrogen Accelerates Void Growth



$$\varsigma(\varepsilon^p) = \ln\left(\frac{R_v}{R_0}\right) = \int_0^{\varepsilon^p} a_v \exp\left(\sigma_{kk} / \sigma_Y\right) d\varepsilon^p$$

$$Tr = \sigma_{kk} / \sigma_Y$$

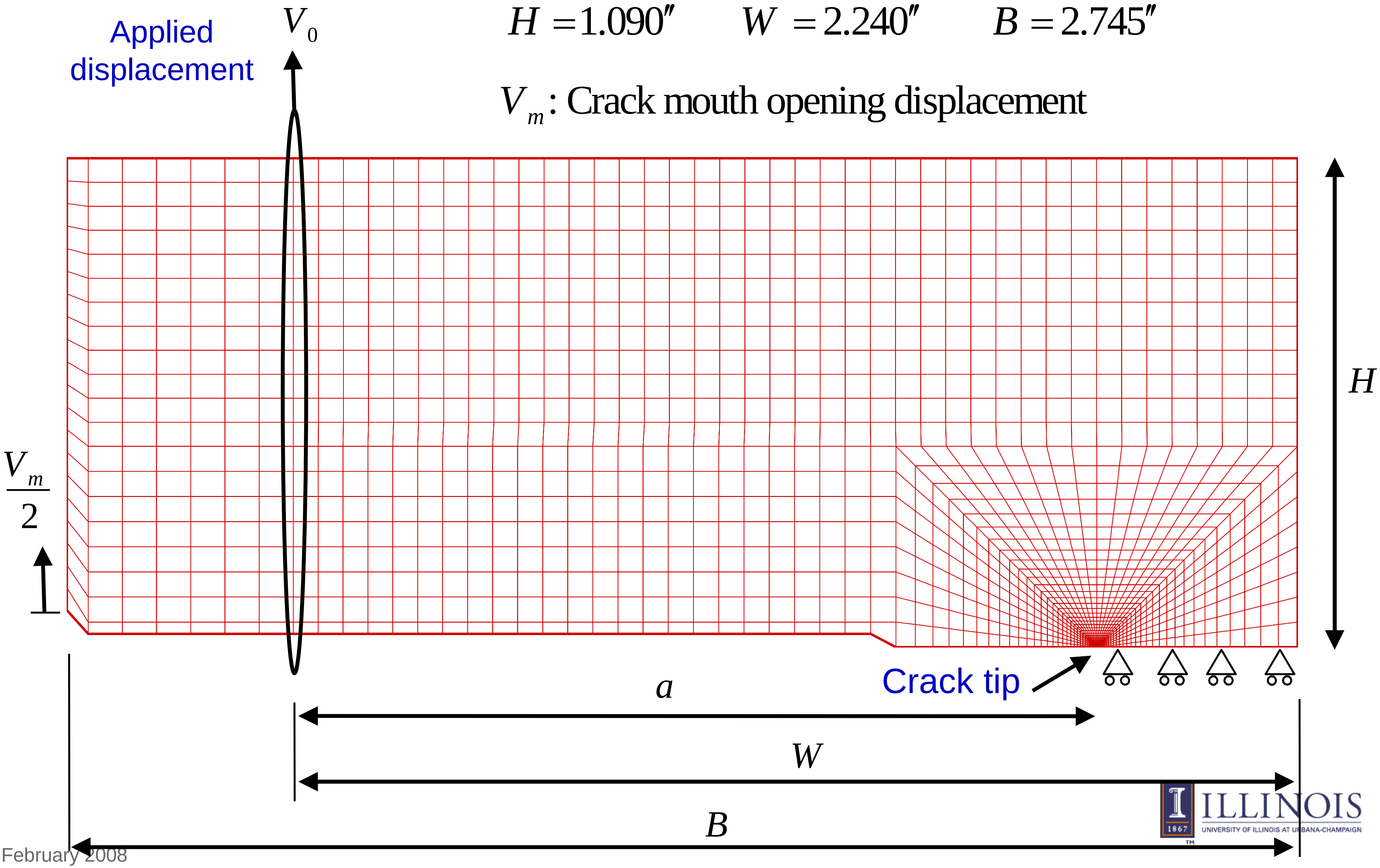
$$\bar{\varsigma} = \frac{\varsigma|_H}{\varsigma|_{H=0}}$$

$$Tr = \frac{\sigma_{kk}}{\sigma_Y} = \text{triaxiality}$$

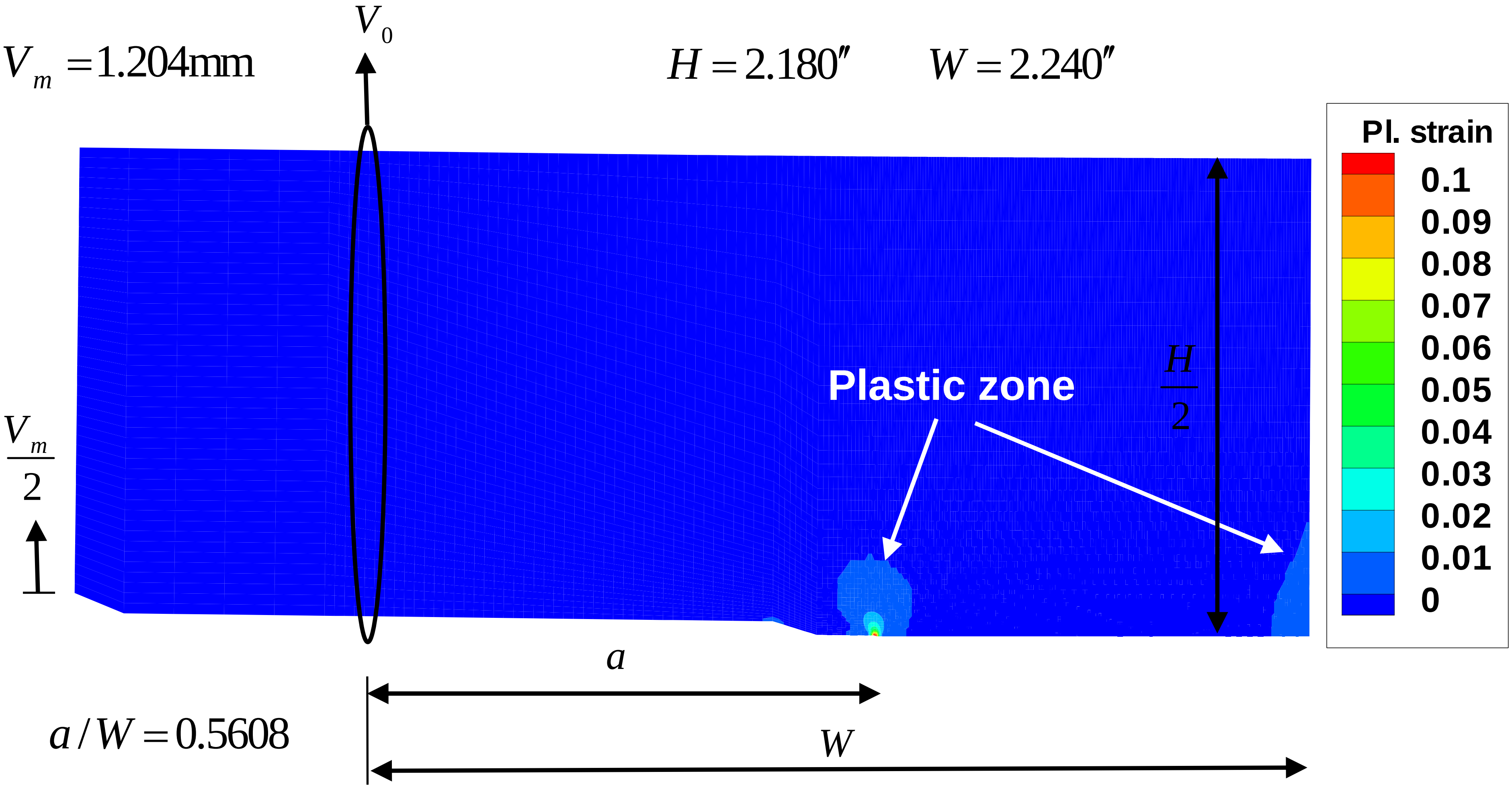
WOL Specimen for Subcritical Crack Growth Finite Element Mesh

$H = 1.090''$ $W = 2.240''$ $B = 2.745''$

V_m: Crack mouth opening displacement



WOL Specimen (X-100) loaded to K_I=158 MPa√m



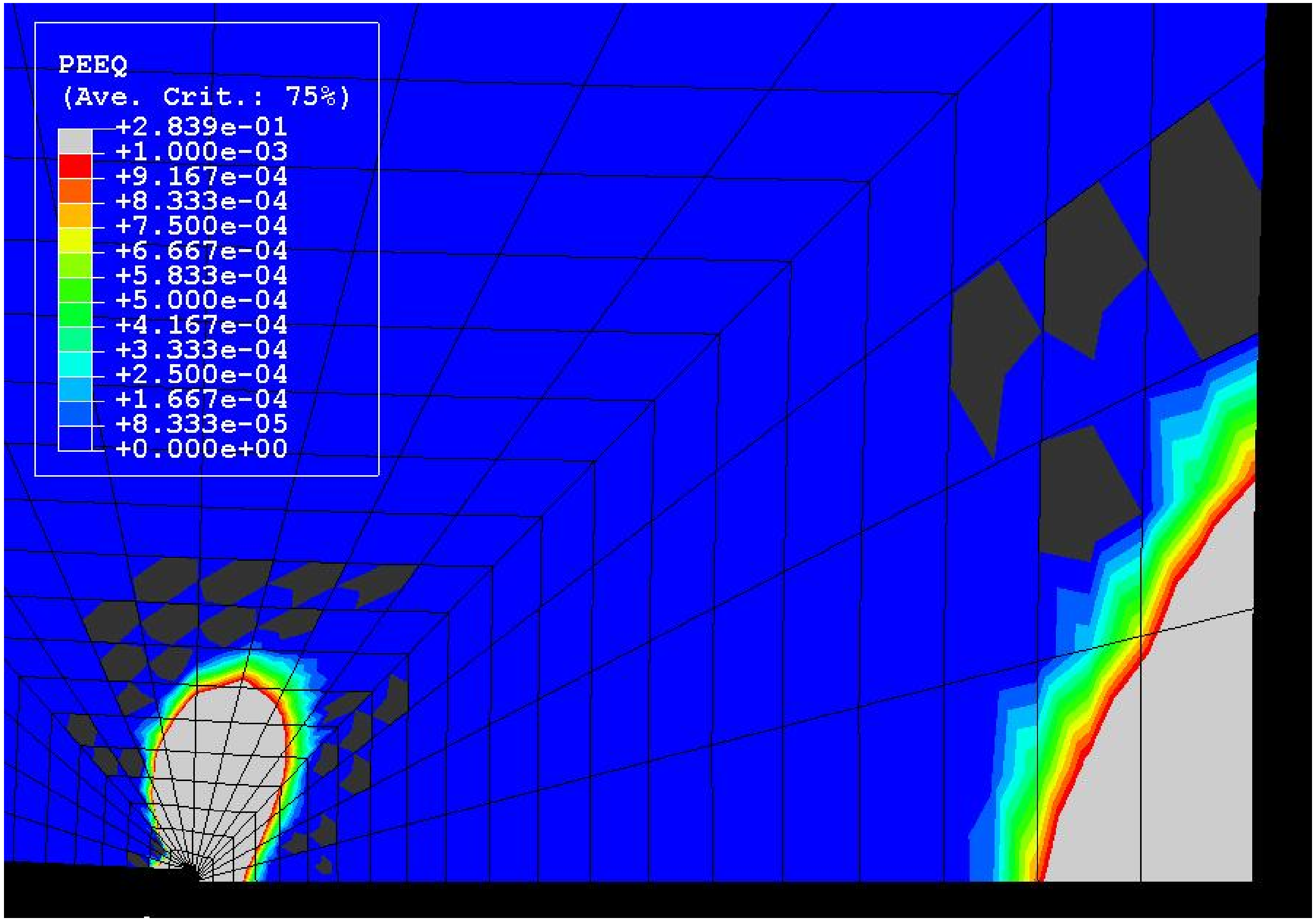
Plasticity is confined to the crack tip under K-dominance

Crack Arrest in WOL Specimen : K_I - dominance

$$V_m = 1.204\text{mm} \qquad a/W = 0.9408$$

$$\xrightarrow{\text{ASTM}} \quad K_I = 57.5\ \text{MPa}\sqrt{\text{m}}$$

$$\xrightarrow{\text{FEM}} \quad K_I = 63.8\ \text{MPa}\sqrt{\text{m}}$$



FEM (Plastic)

$$J = 16008\ \text{N/m}$$

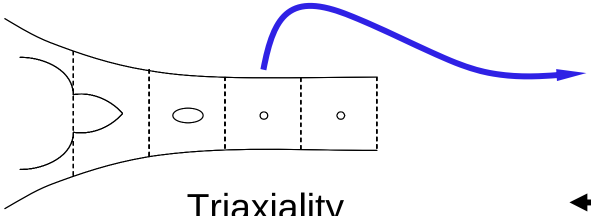
$$K_I = \sqrt{\frac{JE}{1-\nu^2}}$$

$$K_I = 62.2\ \text{MPa}\sqrt{\text{m}}$$

K_I dominance when crack stops

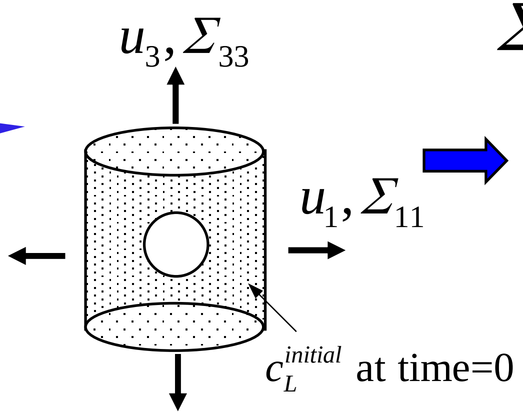
Long Term Objective: Multiscale Fracture Approach

(a) Crack tip fracture process zone

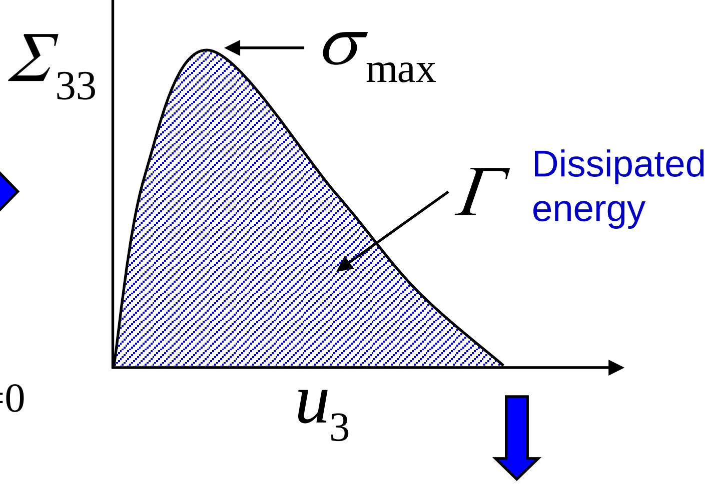


Triaxiality
Hydrogen concentration

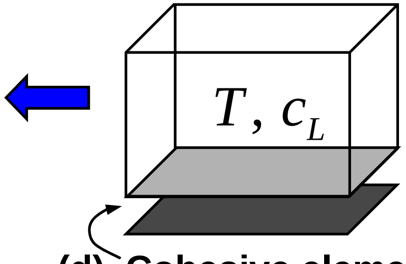
(b) Axisymmetric unit cell model



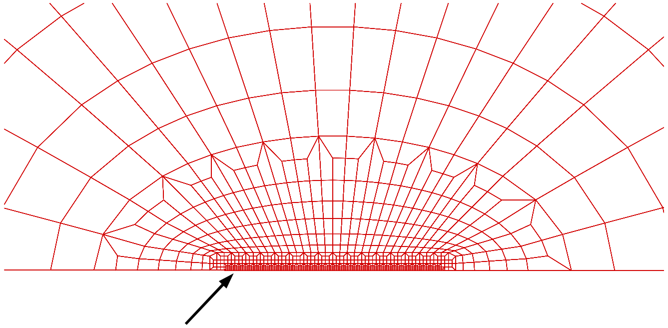
(c) Traction - separation law



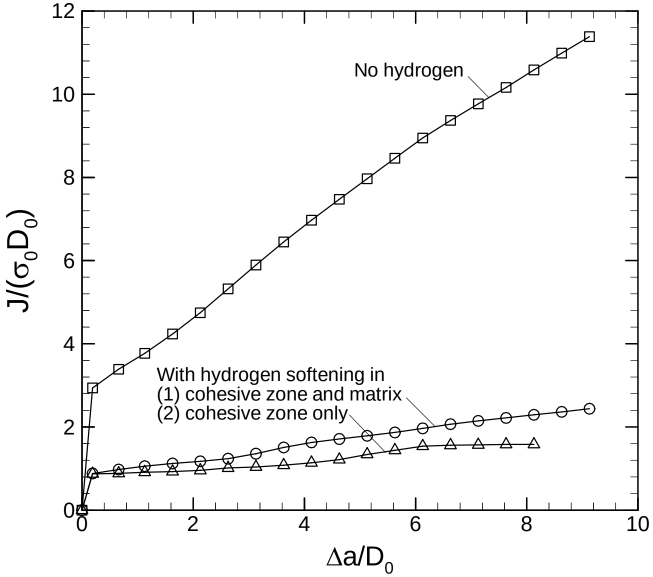
Adjacent finite element



(d) Cohesive element



(e) Cohesive elements characterized by a traction-separation law based on the unit cell model



Conclusions and Future Work

- **Attempted to characterize the hydrogen concentration and stress fields in a pipeline in terms of K_I and T-stress (J-T fracture locus - constraint fracture mechanics)**
  - **Model depends on assumptions (e.g. trapping according to Kumnick and Johnson model, reversible traps, etc) that need to be explored through microstructural characterization and permeation measurements**
  - **Self similarity and no explicit dependence on crack depth**
  - **Transferability of results from laboratory specimens**
  - **If void growth is the mechanism of failure, hydrogen enhances void growth through softening-induced straining**

- **Developed cohesive element technology to simulate decohesion- or ductile-driven processes for crack propagation**
  - **Simulated J-R curve**

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Conclusions and Future Work

- **Coupling fracture mechanisms and microstructural analysis with hydrogen transport, thermodynamics of decohesion, and plastic flow localization to understand**
  - **Interaction of time scales (loading rate, diffusion rate, adsorption rate**
  - **Crack initiation**
  - **Crack propagation**
  - **Devise fracture criteria with predicting capabilities**
    - **Possibly a J_{IC}-T locus**

- **Fracture mechanics/mechanism-based approach to design**
  - **As opposed to the SMYS approach**

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Where We Go From Here

- We have years of experience and extensive knowledge of all aspects of hydrogen embrittlement.
- We have a tremendous collection of analysis tools.
- We can tame the problem

Support by the
U.S. Department of Energy is
Gratefully Acknowledged