Tritium Behavior in Lead Lithium Eutectic (LLE) at Low Tritium Partial Pressure

Masashi Shimada, Ph.D.

Fusion Safety Program,
Idaho National Laboratory,

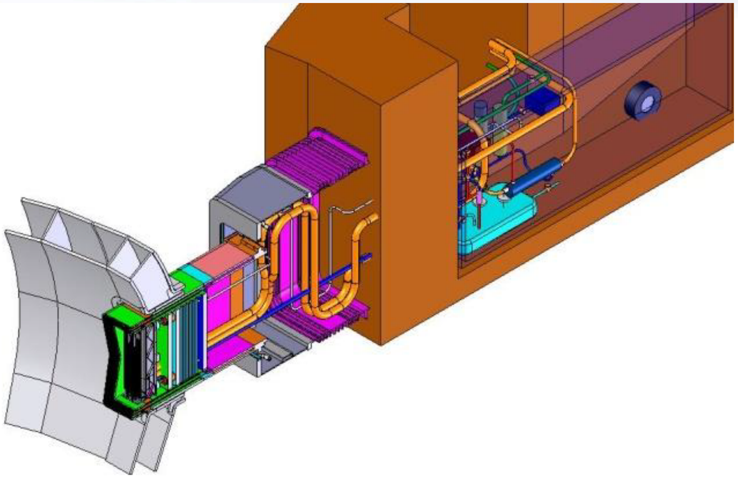
www.inl.gov

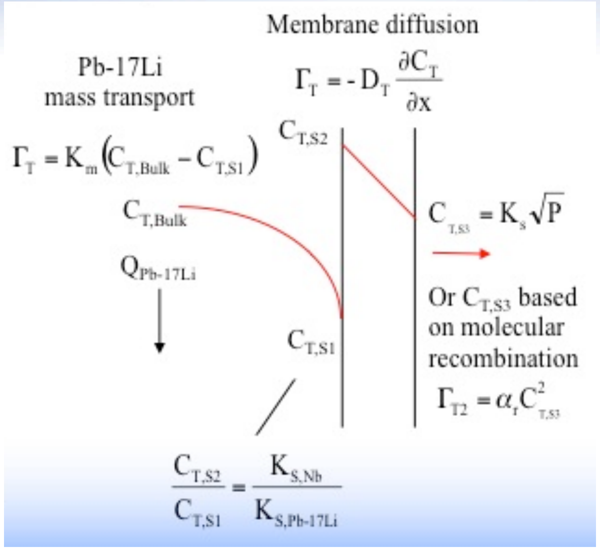
Idaho National Laboratory

Outlines

1. Motivation
2. Experimental apparatus
3. TMAP modeling
4. Experimental results
5. Modeling results
6. Future work

Motivation in fusion safety





- Challenges in blanket development
  - Tritium permeation during normal operation is main operational safety concern.
  - Mass transport properties (e.g. diffusivity, solubility, and permeability) of tritium behavior in blanket/structural/barrier materials at realistic blanket conditions (e.g. low tritium partial pressure << 100 Pa) has very limited database and underlying physics is not fully understood.
  - H Solubility from Lead-Lithium Eutectic (85 at.% Pb and 15 at.% Li), which is the candidate breeder/coolant material for DCLL has 6 orders of magnitudes scattering in literature database.

Motivation for low tritium partial pressure permeation

Reference: "Tritium permeation through 304 stainless steel…" A.S. Zarchy, and R.C. Axtmann, *Journal of Nuclear Materials* **79** (1979) 110

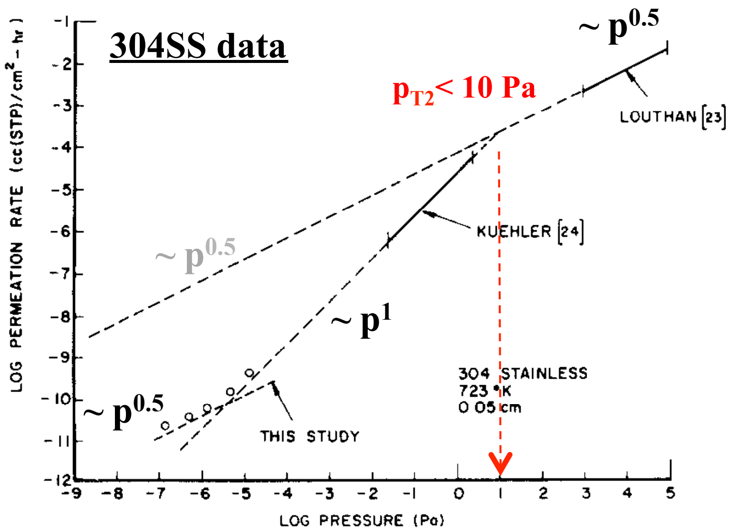


Fig. 4. Permeation rates of hydrogenic gases through 304 stainless steel as measured in three different studies. Results have been normalized for isotopic effects and differences in sample thicknesses.

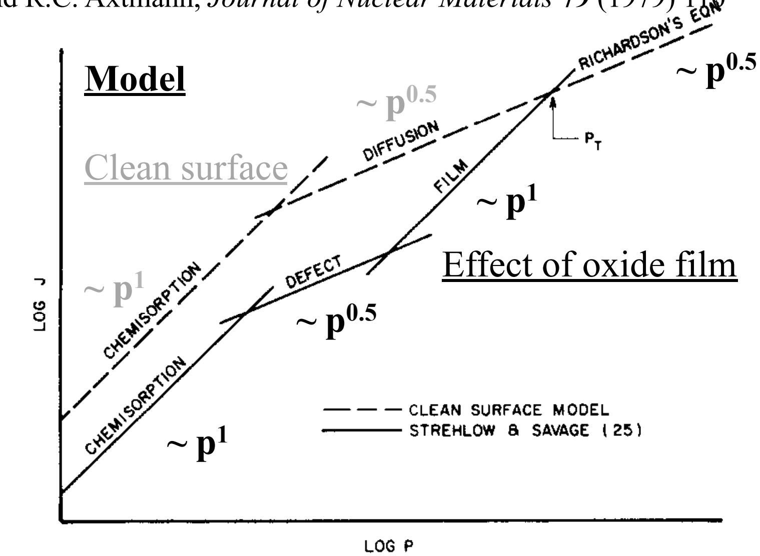


Fig. 6. Overall permeation behavior of hydrogenic gases through metals. P_T is the transition pressure between metal-limited and film-limited permeation.

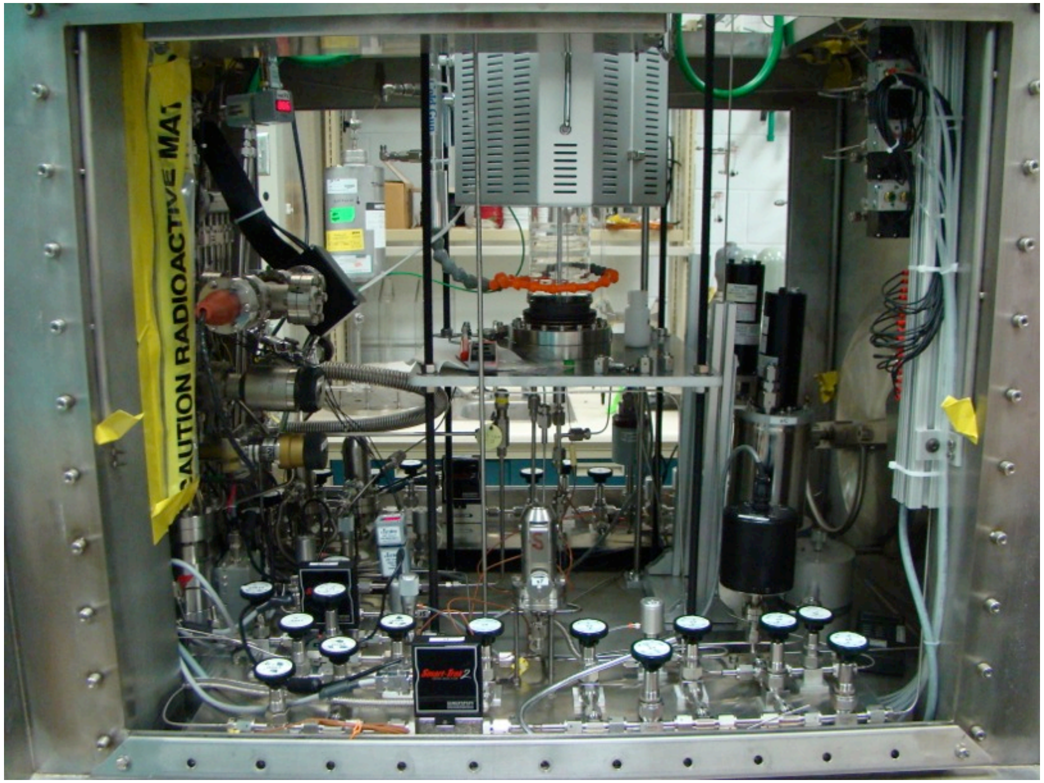
- Importance of tritium permeation at low tritium partial pressure:
  - Tritium permeation rate is lower at low tritium partial pressure (p_{T2}< 10 Pa for 304 SS)
  - Tritium permeation to the environment can be significantly reduced
  - Data from low tritium partial pressure is limited.
  - How about multi-components (H/D/T) permeation

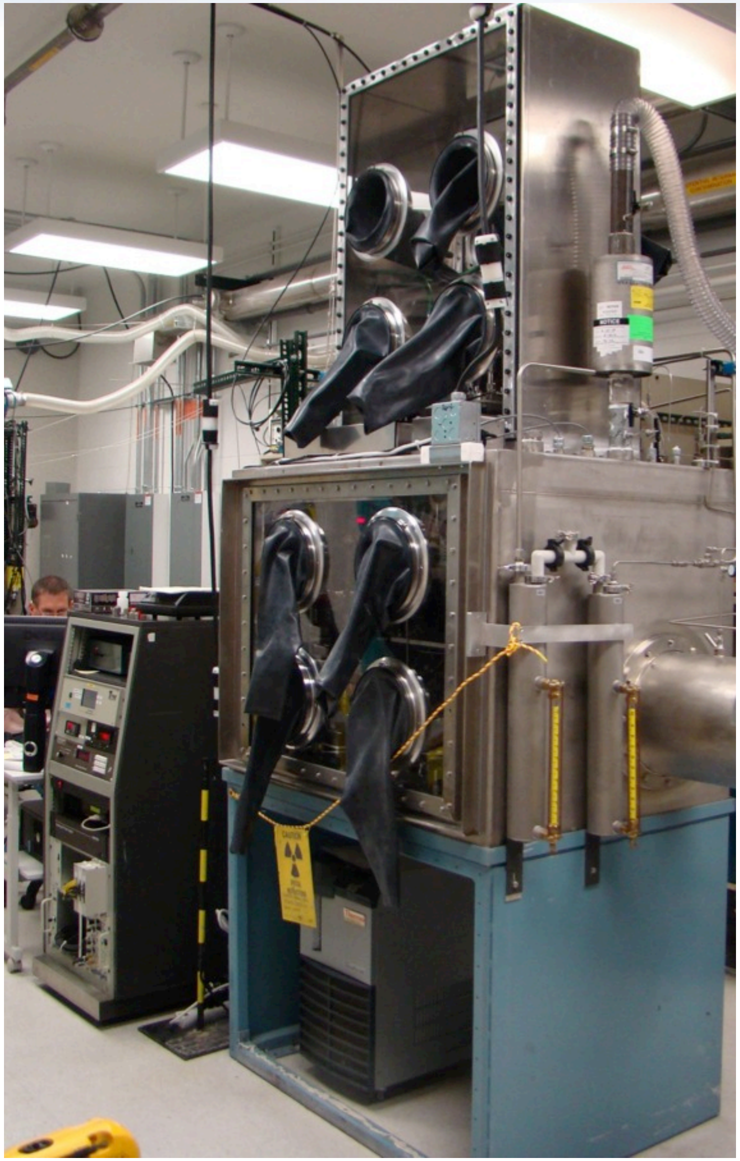
Outlines

1. Motivation
2. Experimental apparatus
3. TMAP modeling
4. Experimental results
5. Modeling results
6. Future work

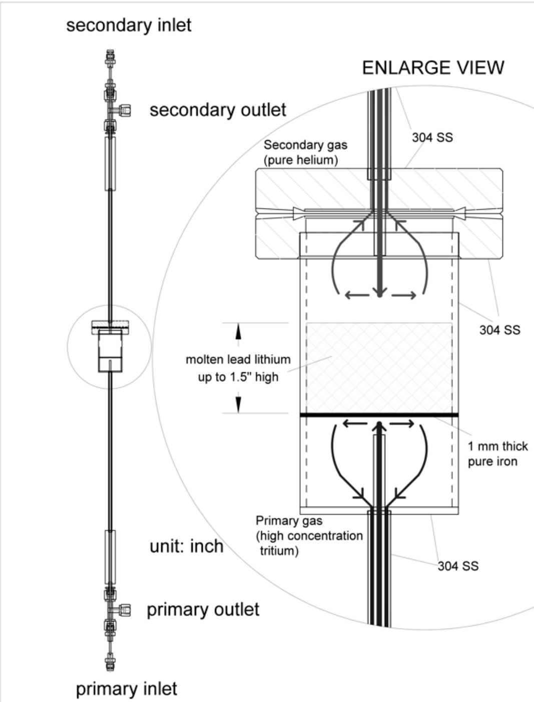
Tritium Gas Absorption Permeation (TGAP) experiment

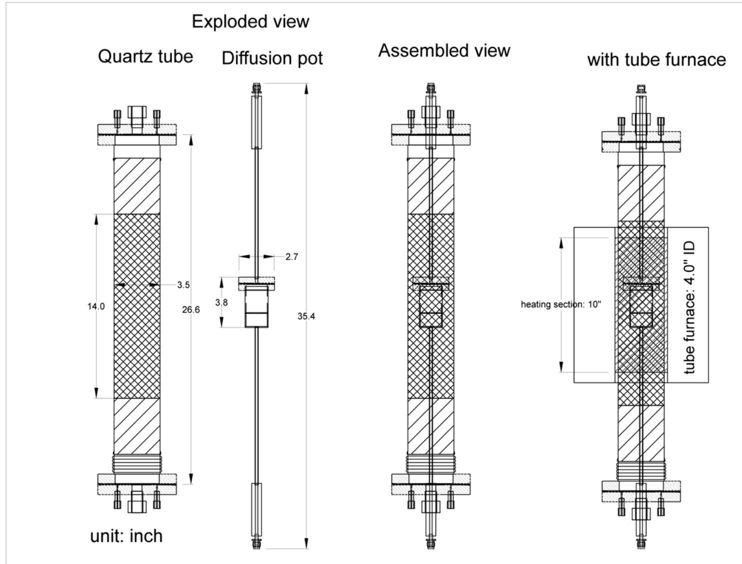
- The experimental apparatus is inside Contamination Area (CA) for tritium
  - Tube furnace in Ventilated Enclosure (VE)
  - Exhaust clean-up system in Fume Hood





Tritium Gas Absorption Permeation experiment

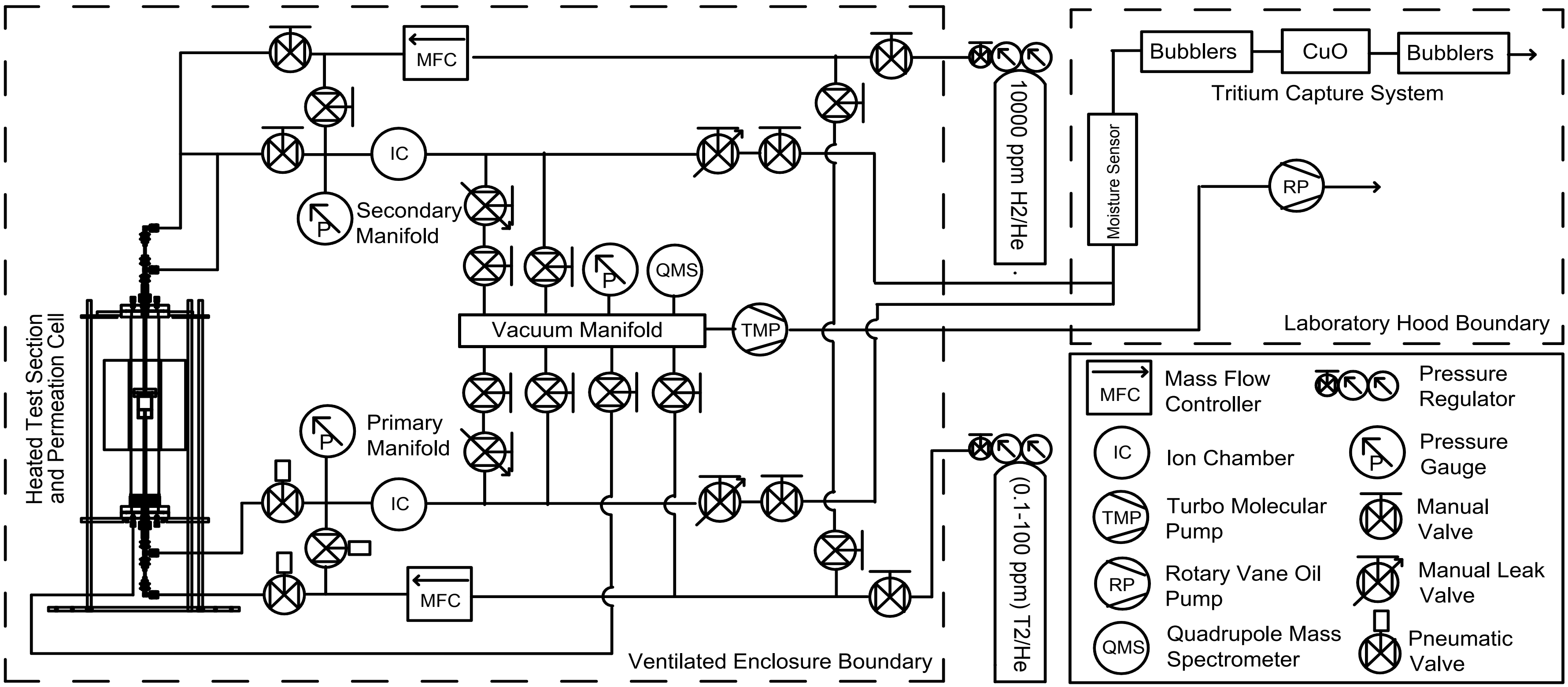




Unique capabilities

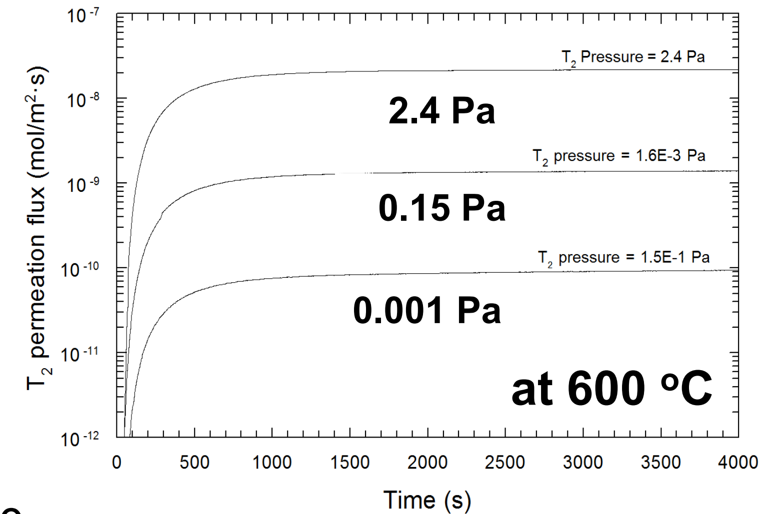
- Designed to measure transport properties (e.g. diffusivity, solubility, and permeability) of tritium at realistic blanket conditions (e.g. low tritium partial pressure < 1000 Pa) for disc geometry sample
- Capable of testing liquid breeder material (e.g. PbLi and FLiBe) and disc shaped metal
- Uniform temperature (+/- 10 C) within the test section utilizing 12” tube furnace

Simplified P&ID of Tritium Gas Absorption Permeation experiment

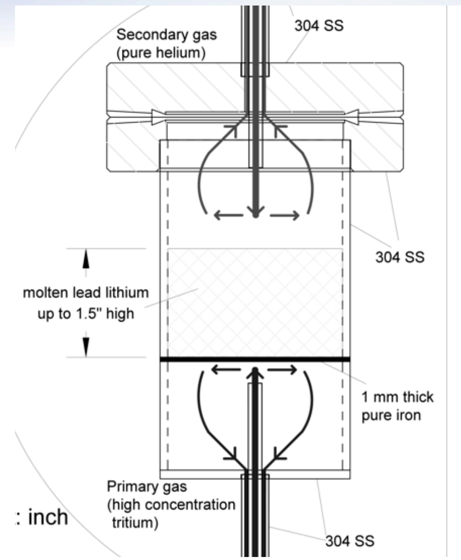


Idaho National Laboratory

Experimental procedure



H_2/He



T_2/He

- t < 0:
  - Primary and Secondary was purged with **1 % H_2/He** **200 sccm, p_t=10^5 Pa**
  - Bake out at 600 C with for 2 hours to remove oxide
  - Test section was kept at uniform temperature (+/- 10 C) for 1 hour at t<0.
  - Traps in the α-Fe were saturated by hydrogen.
- At t=0,
  - Tritium (0.001, 0.15, 2.4 Pa **T_2/He**) were introduced in the primary.
- t >0 :
  - Fast breakthrough time was obtained (within a minutes) and tritium equilibrates within 30 minutes

Outlines

1. Motivation
2. Experimental apparatus
3. TMAP modeling
4. Experimental results
5. Modeling results
6. Future work

Tritium Migration Analysis Program (TMAP)

- The TMAP calculates the time-dependent response of a system of solid structures or walls (may be a composite layer), and a related gas filled enclosures or rooms by including
  - Movement of gaseous species through structures surfaces, governed by dissociation/recombination, or by solution law such as Sieverts' or Henry's Laws
  - Movement in the structure by Fick's-law of bulk diffusion with the possibility of specie trapping in material defects
  - Thermal response of structures to applied heat or boundary temperatures
  - Chemical reactions within the enclosures
- User specified convective flow between enclosures
- Equations governing these phenomenon are non-linear and a Newton solver is used to converge the equation set each time-step

NOTE: TMAP ver.4 can be obtained from DOE OSTI ENERGY SCIENCE AND TECHNOLOGY SOFTWARE CENTER

TMAP Capabilities (cont.)

- TMAP does not treat plasma surface physics, such as sputter or sputtered material re-deposition. TMAP's basic equations are:

Surface Movement

Non-equilibrium

$$J_m = K_d p_m - K_r C_m^2$$

Equilibrium

$$C_m = K_s \sqrt{p_{m_2}}$$

or

$$C_m = K_s \cdot p_{m_2}$$

Bulk Diffusion

$$\frac{\partial C_m}{\partial t} = \frac{\partial}{\partial x}\left(D \frac{\partial C_m}{\partial x}\right) + S_m - \sum_{i=1}^{K} \frac{\partial C_t^i}{\partial t}$$

Bulk Trapping

$$\frac{\partial C_t}{\partial t} = \alpha_t f_t C_m - \alpha_r C_t$$

Surface

J_m – Net specie surface flux (m^{-2}-s)
K_d – Molecular disassociation coefficient (m^{2}-s^{-1})
K_r – Atom recombination coefficient (m^{2}-s^{-1})
K_s – Sievert's solubility coefficient (m^{2}-s^{-1})

Bulk

C_m – Mobile specie concentration (m^{-3})
D – Specie diffusion coefficient (m^{2}-s^{-1})
K – Number of traps (-)
C_t – Trapped specie concentration (m^{-3})
α_t – Trapping rate coefficient (s^{-1})
f_t – Probability of landing in a trap site (-)
C_m – Mobile concentration (m^{-3})
α_r – Release rate coefficient (s^{-1})

$$\alpha_t = \frac{D}{\lambda^2};\ f_t = \frac{c_t^o - C_t}{N};\ \alpha_r = \nu_o \exp\left(-\frac{E_t}{kT}\right)$$

λ – jump distance or lattice constant (m)
c_t^o – Trap site concentration (m^{-3})
N – Bulk material atom density (m^{-3})
ν_o – Debye frequency (s^{-1})
E_t – Trap energy (eV)

Input parameter for TMAP modeling

- To apply TMAP to experimental data, property data for H diffusivity, solubility, and surface recombination/dissociation coefficients in α-Fe are required.
- Physical data of tritium partial pressure and sample temperatures are also required
- Mass transport property data used for H permeation in α-Fe are:
  - H diffusivity in alpha-iron [m^2/s]
    - y=4.43e-8*exp(-638.7/temp)
  - H solubility in alpha-iron [1/(m^3 Pa^0.5)]
    - y=4.2e23*exp(-2922.6/temp)
  - H/T recombination coefficient in alpha-iron [m^4/s]
    - y=4.6e-24/temp*exp(+4365.9/temp)
  - H/T dissociation coefficient in alpha-iron [m^4/s]

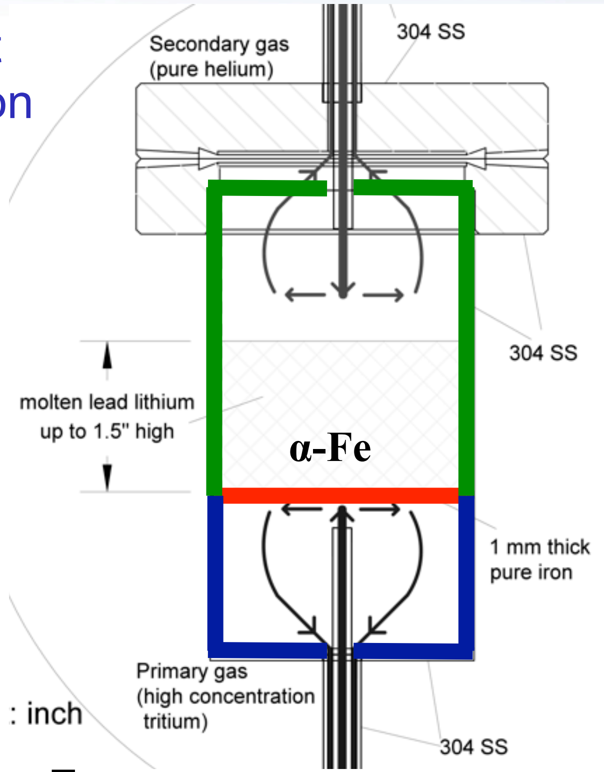
$$K_d = K_r \cdot K_s^2$$

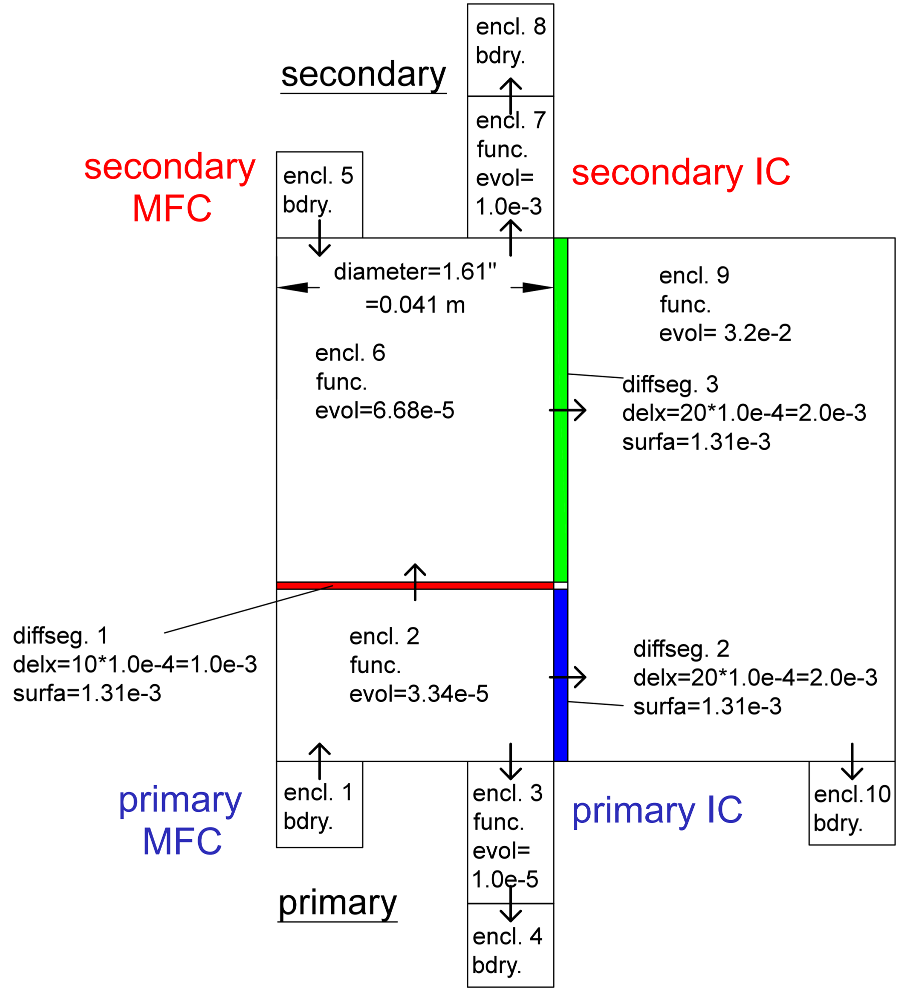
- Adjusted T diffusivity and T solubility to fit exp. data as two fitting parameters

[1] Yamanishi, et. al., Trans. Japan Institute of Metal (1983)
[2]Tahara and Hayashi, Trans. Japan Institute of Metal, v.26 (1985) 869
[3] Eichenauer et. a., Z. Metallkunde, v.49 (1958) 220
[4] Nagasaki et. a., JNM v.191-194 (1992) 258

TMAP configuration for (1 mm) α-Fe

Experiment configuration

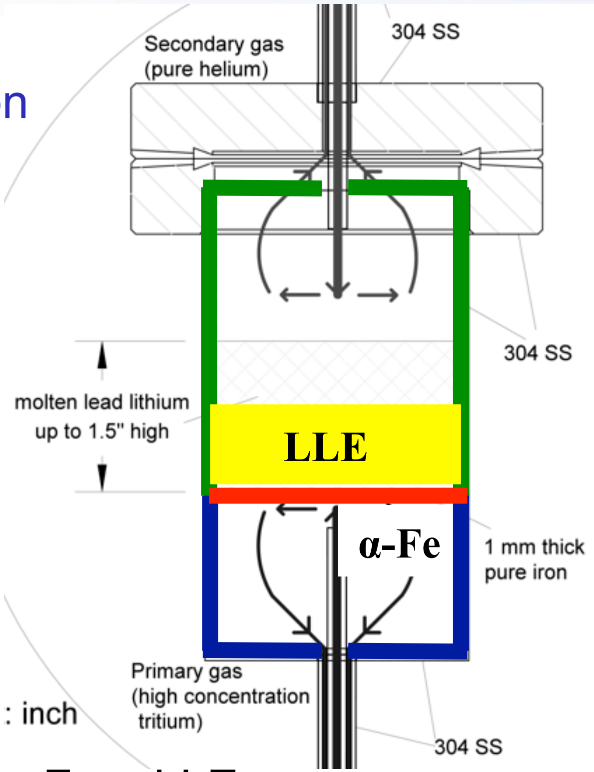


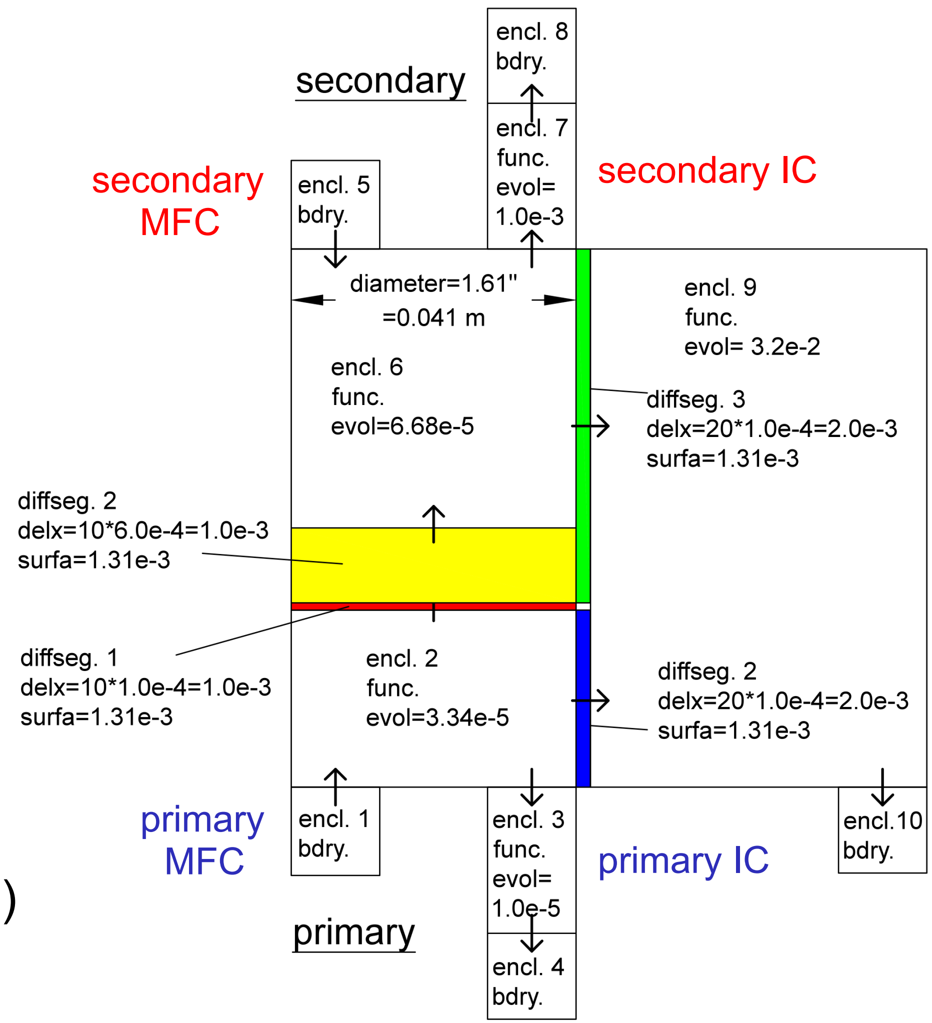


- TMAP for α-Fe
  - 1D geometry
  - 4 enclosure species (H_2, T_2, HT He)
  - 2 diffusion species (H, T)
  - 3 segments
  - 10 enclosures

TMAP configuration for (1 mm) α-Fe + (6mm) LLE

Experiment configuration



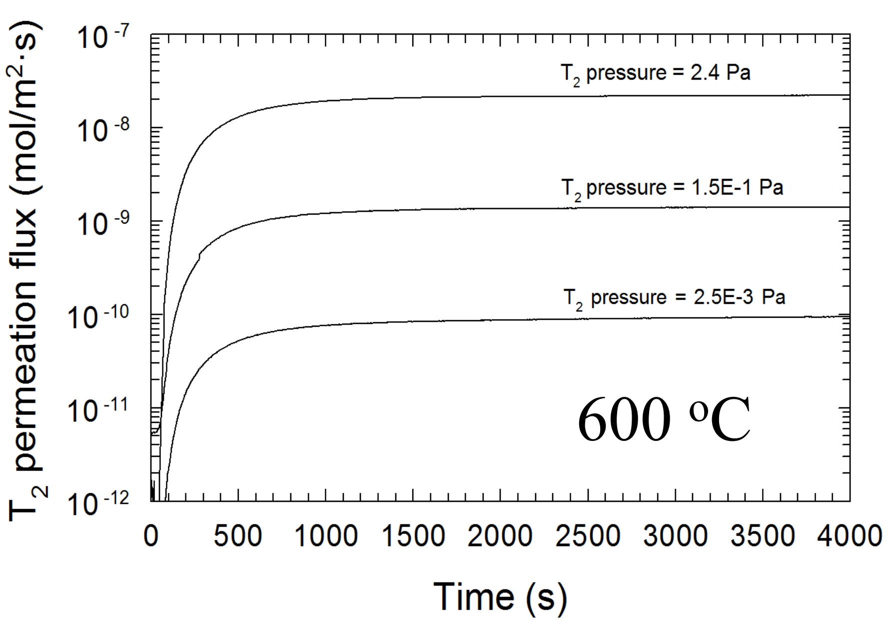


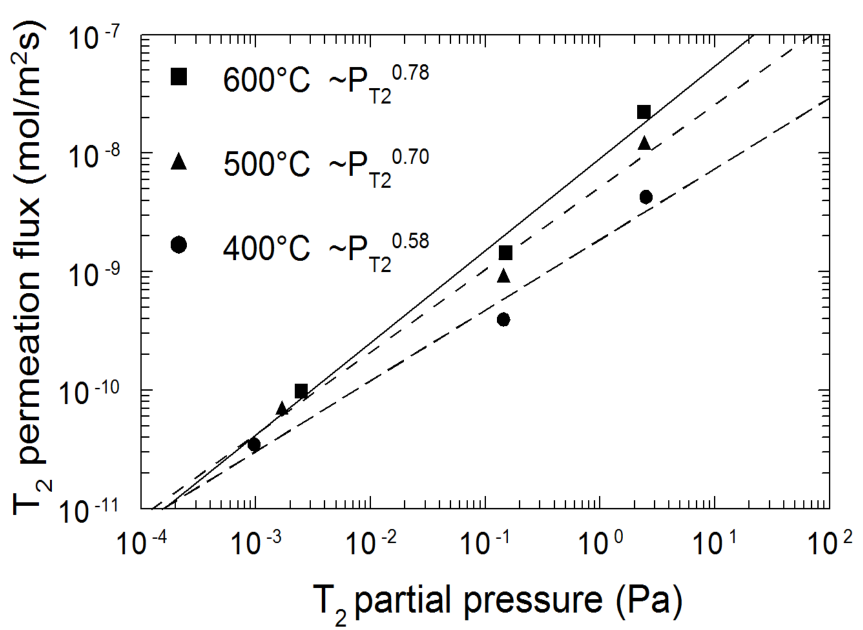
- TMAP for α-Fe + LLE
  - 1D geometry
  - 3 enclosure species (H_2, T_2, HT, He)
  - 2 diffusion species (H, T)
  - 4 segments
  - 10 enclosures

Outlines

1. Motivation
2. Experimental apparatus
3. TMAP modeling
4. Experimental results
5. Modeling results
6. Future work

Tritium permeation through (1mm) α-Fe

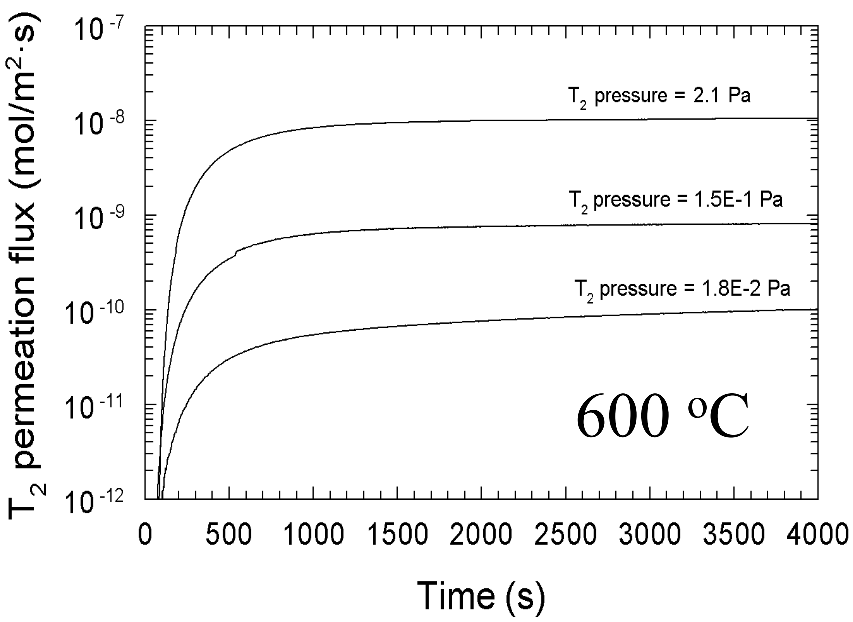


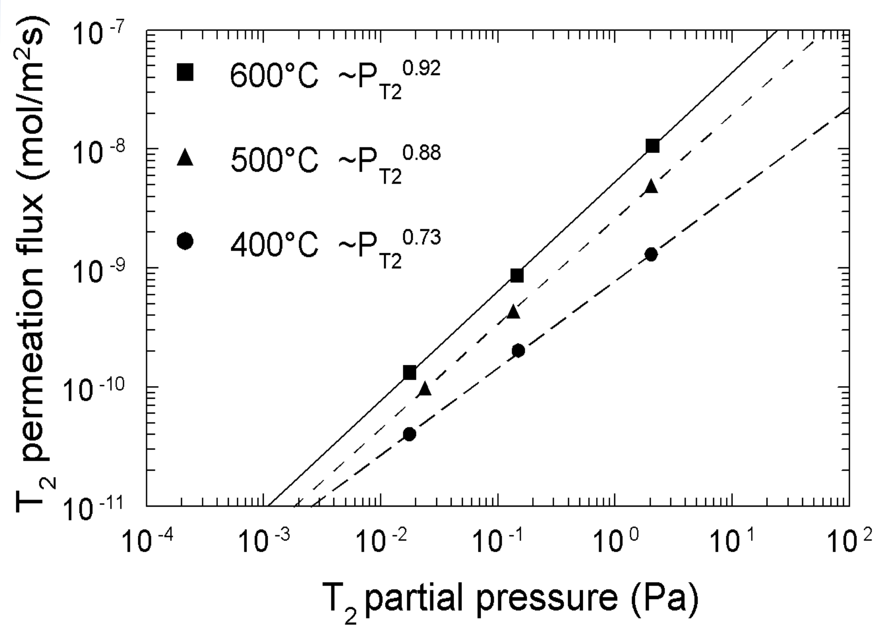


- Tritium partial pressure dependence were $P_{T2}^{0.58} \sim P_{T2}^{0.78}$
- Tritium behavior is in the transition range ($P^{0.5} < P^{x} < P^{1}$) from diffusion limited to surface limited.

- Issues:
  - H_2 and HT concentration in primary are unknown.
  - Should be HT in the secondary

Tritium permeation through (1 mm) α-Fe + (6mm) LLE



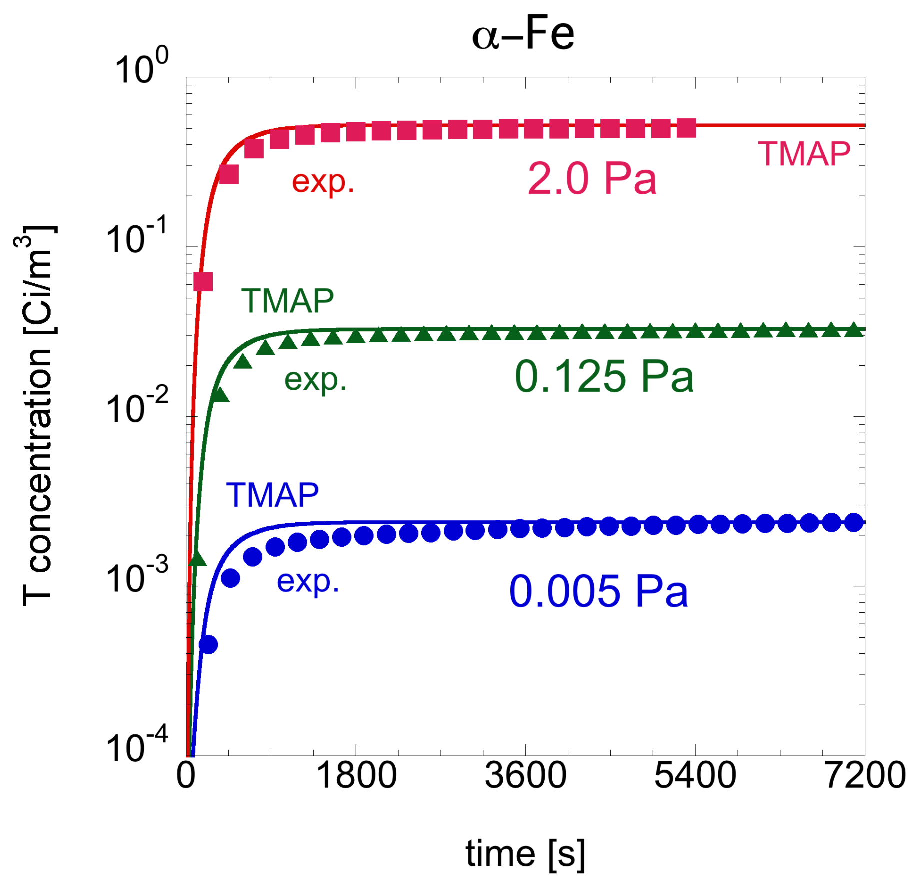


- Tritium partial pressure dependence were $P_{T2}^{0.73} \sim P_{T2}^{0.92}$
- Tritium behavior is in the transition range ($P^{0.5} < P^{x} < P^{1}$) from diffusion limited to surface limited.
- Straight line (R^2~1) fit at all three case
- Issues:
  - Primary tritium partial pressure (especially at lowest case) was higher than that of α-Fe test, making it difficult to compare those two results

Outlines

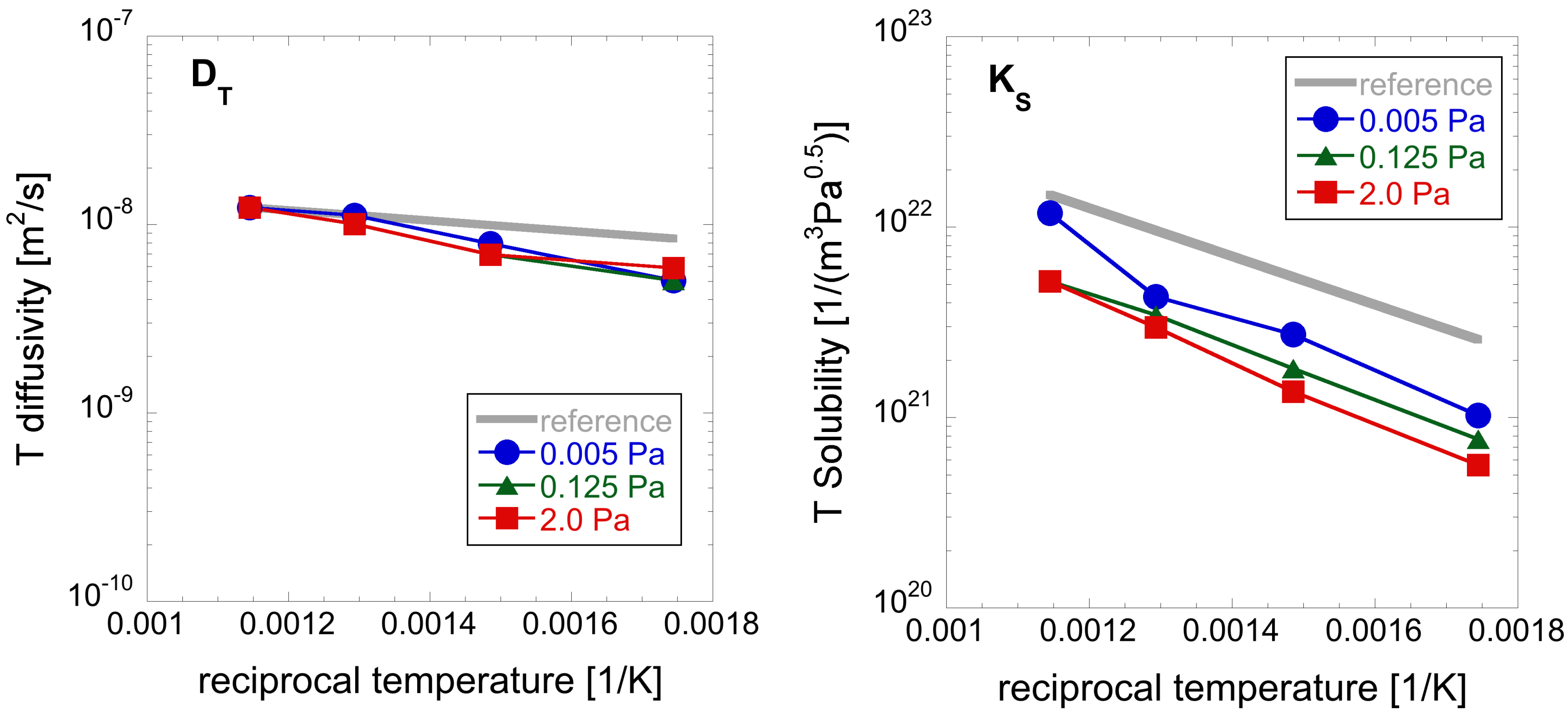
1. Motivation
2. Experimental apparatus
3. TMAP modeling
4. Experimental results
5. Modeling results
6. Future work

Tritium permeation through (1 mm) α-Fe



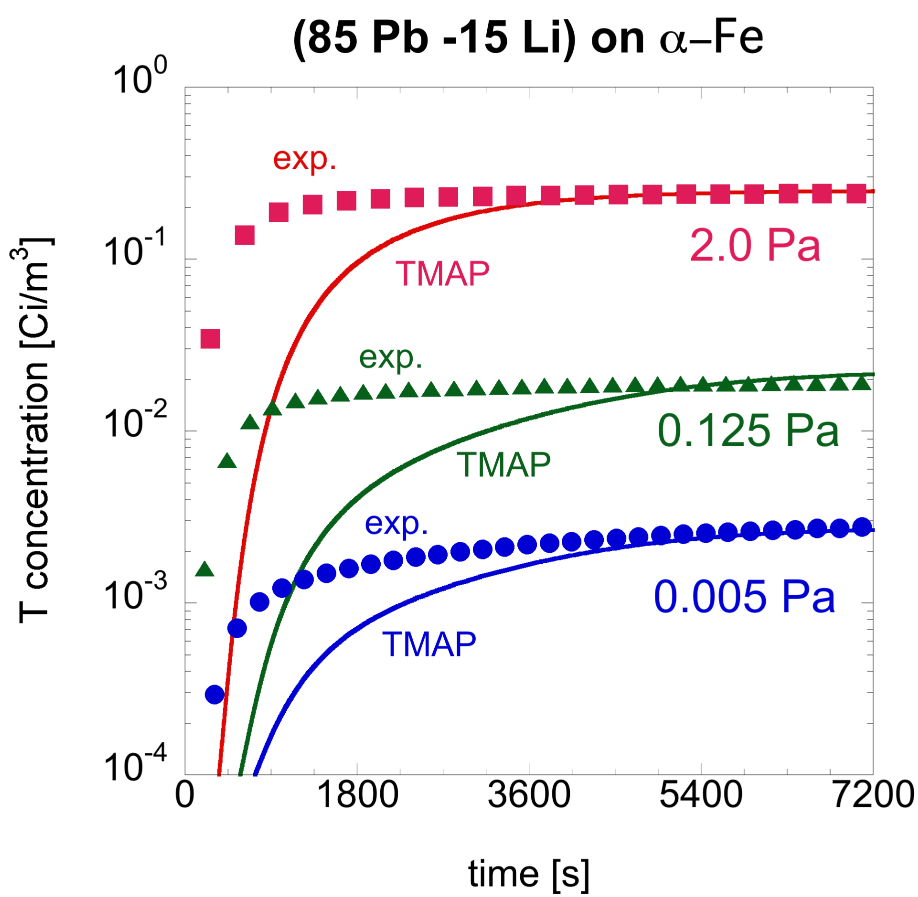
- TMAP can reproduce the experimental results well with two fitting parameters

Tritium permeation through (1 mm) α-Fe



- Tritium diffusivity in α-Fe:
  - Similar to the extrapolation from literature H diffusivity x sqrt(3)
- Tritium solubility in α-Fe:
  - 30-70 % lower than literature data
  - Shows T partial pressure dependence

Tritium permeation through (1 mm) α-Fe + (6 mm) LLE



- Tritium diffusivity in LLE:
  - A factor of 2-3 higher value were needed to fit exp. data
- Tritium solubility in LLE:
  - Similar to literature data

Outlines

1. Motivation
2. Experimental apparatus
3. TMAP modeling
4. Experimental results
5. Modeling results
6. Future work

Summary and future work

- Continue on modeling with TMAP
  - Improve the modeling of dissociation/recombination for hetero-nuclear species (e.g. HT)
  - Modify the metal-LLE interface boundary condition
  - Understand the mechanism for permeation mechanism in LLE
- Follow-up experiment:
  - Test with well characterized metal (e.g. Fe and Ni)
  - Test with PFCs (e.g. W)
  - Test with different thickness/weight LLE