Overview of Tritium and molten salt FLiBe research at Safety and Tritium Applied Research (STAR) facility

Masashi Shimada
Fusion Safety Program, INL

www.inl.gov

Idaho National Laboratory

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U.S. DEPARTMENT OF ENERGY | Office of Science

Outline

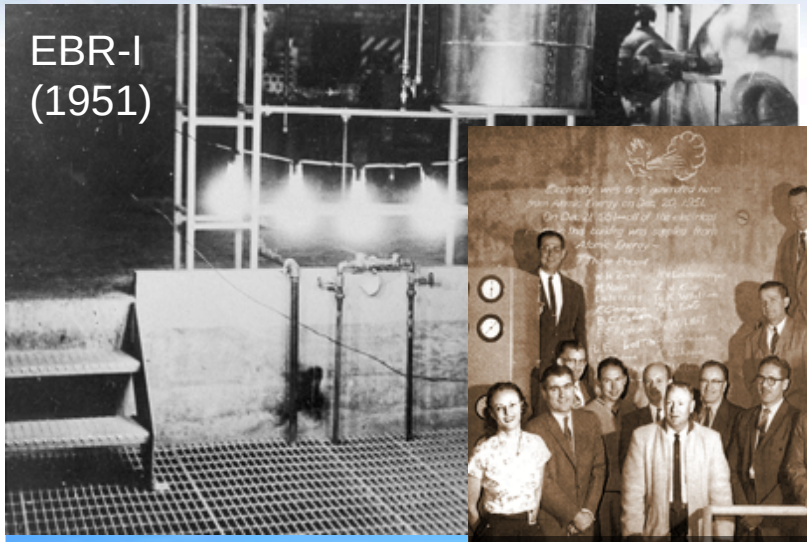
1. Motivation and overview of STAR research

2. Molten salt research at STAR during JUPITER-II project (April 2001 – March 2007)

3. Present capabilities

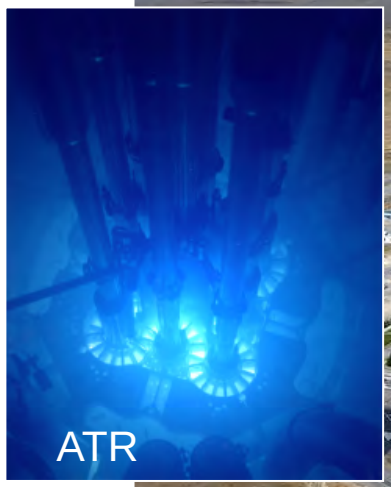
Idaho National Laboratory (INL) and fusion safety

- INL overview:
  - Geographically, the largest lab in 10 multi-program US national laboratories
  - 52 reactors were designed and built in Idaho
  - The world's first usable electricity from nuclear energy generated in EBR-I in 1951
  - The nation's lead laboratory for nuclear energy research and development



EBR-I scientists

- Advanced Test Reactor (ATR)
  - Light water moderated/cooled with Be neutron reflector
  - Max: 250 MW_{th} and Four Leaf Clover" design
  - Materials and fuels testing, isotope production (e.g. ^{60}Co)
- Safety and Tritium Applied Research (STAR) Facility
  - Fusion safety and tritium research


ATR

STAR facility at Advanced Test Reactor Complex

Comparison of site size:

- INL
  - 890 sq. mile
- Rhode Island
  - 1214 sq. mile (0.73)
- Boston
  - 89.6 sq. mile (9.9)

NOTE: the number in the () is the ratio of the INL site to each site.

STAR facility is

- Supported by DOE SC Office of Fusion Energy Sciences

- Restricted to a facility total tritium inventory of less than 1.6 gram (15,390 Ci), to remain below DOE Hazard Category 3 threshold
  - ➔ less than Hazard Category 3 Nuclear Facility
  - Current tritium inventory is ~ 0.35 gram (~3500 Ci)

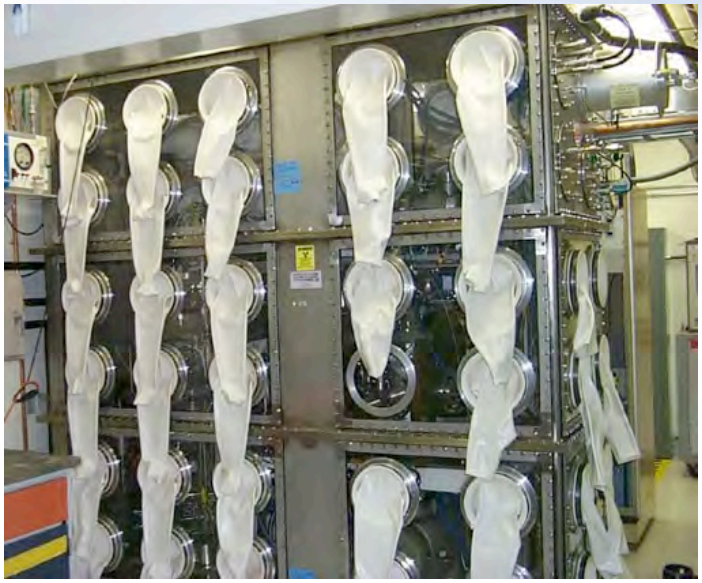
- Specializes in:
  - Tritium (fusion fuel)
  - Activated materials (neutron-irradiated tungsten)
  - Advanced coolant (FLiBe, PbLi, He)
  - Toxic material (Be)

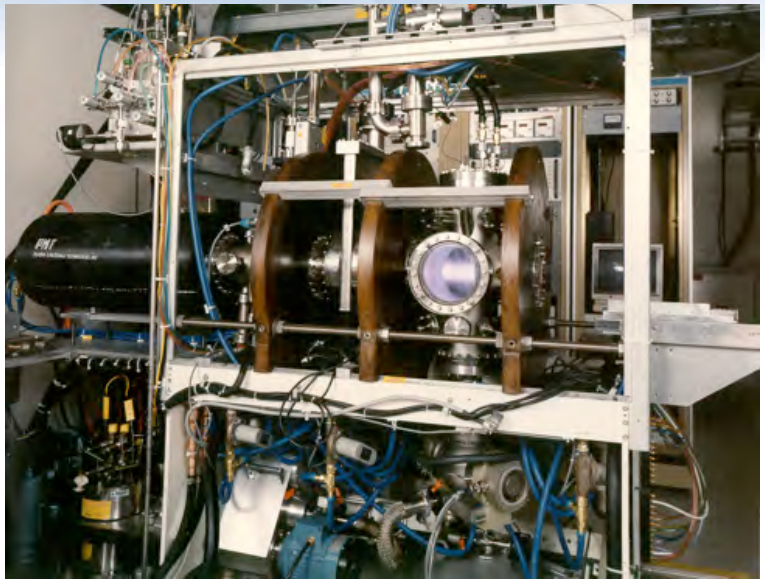
- Containment strategy for hazardous materials:
  - Ventilated enclosures and Laboratory hood for handling tritium
  - Gloveboxes for handling beryllium, Flibe etc..

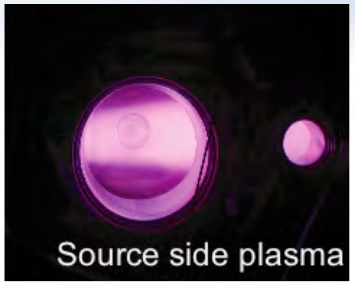
Motivation of STAR research

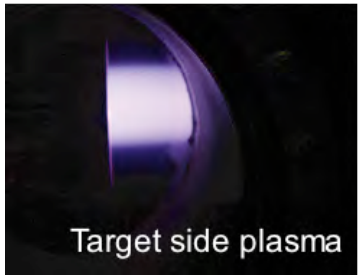
- Investigate tritium related and non-tritium related experimental fusion safety research
  1. **Tritium related fusion safety**
     - In-vessel tritium source term
       - Tritium retention in fusion materials
     - Ex-vessel tritium release term
       - Tritium permeation in fusion materials
  2. **Non-tritium related fusion safety**
     - Dust explosivity
       - Beryllium dust
     - Steam reactivity
  3. **Tritium related fission safety**
     - Tritium removal in molten salt ?

Tritium retention studies for fusion PFC development in Tritium Plasma Experiment







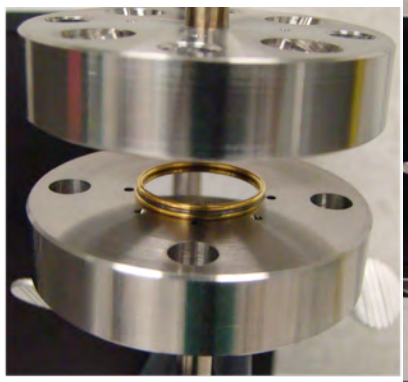


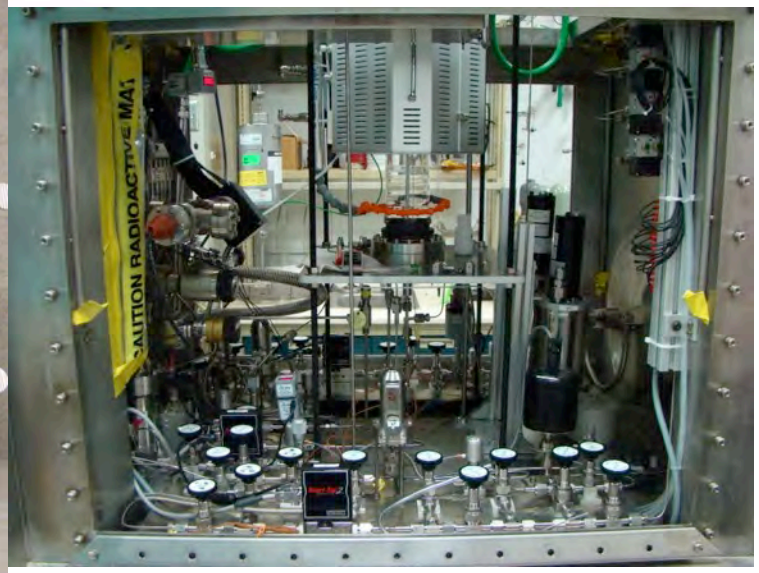
Unique capabilities

- *Designed to measure tritium behavior (e.g. retention and permeation) in activated materials with tritium plasma (e.g. tritium/deuterium ratio ~ (0.01-0.1) at divertor relevant ion flux condition*
- *Capable of testing neutron-irradiated (< 1 mSv/hr @ 30cm) specimens (W, RAFM steels, etc.)*
- *TPE is contained within double enclosure (PermaCon Box and Glovebox)*
- *Collaborations includes: US-JA TITAN collaboration (2007-2012), US-JA PHENIX collaboration (2013-2018), IAEA CRP on irradiated tungsten (2013-2018)*

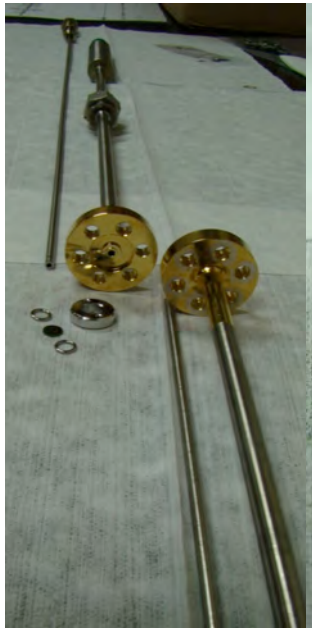
Tritium absorption and permeation studies for fusion blanket development

Test section for NFRI





Test section for PHENIX



Unique capabilities

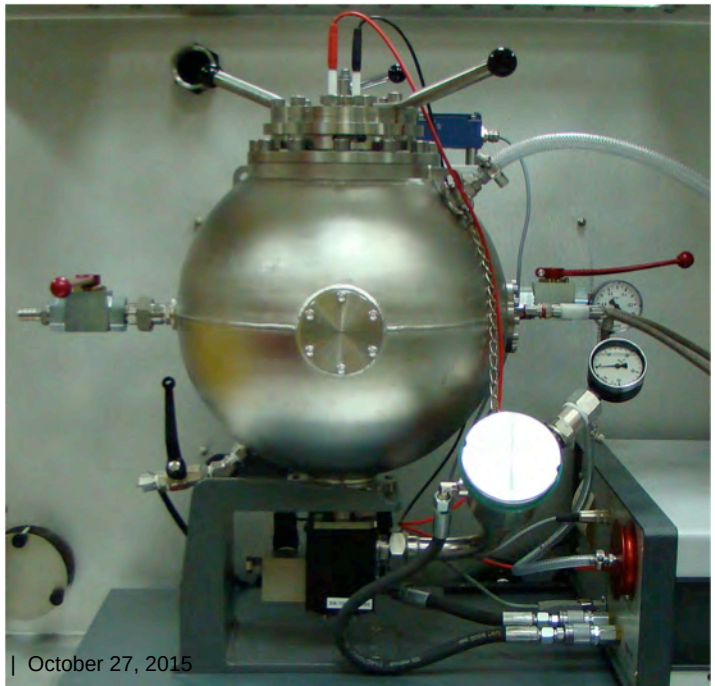
- Designed to measure tritium transport properties (e.g. diffusivity, solubility, and permeability) in activated materials at realistic fusion sweep gas conditions (e.g. low tritium <10 Pa & hydrogen partial pressures < 1000 Pa, moderate < 700 C)
- Capable of testing liquid or ceramic breeder materials (e.g. PbLi, Li_2TiO_3, etc.) and disc shaped metal specimens (W, RAFM steels, etc.)

Hydrogen/deuterium absorption studies for fusion blanket development

- This static gas absorption system studies deuterium gas absorption in materials. It is located inside Laboratory fume hood for hydrogen safety.
- **Unique capabilities**
  - Sub atmospheric absorption (< 0.1 MPa) up to 950 C
  - Utilize three calibrated capacitance manometers (0.002, 1.3, 133 kPa)
  - Capable of testing liquid or ceramic breeder materials (e.g. PbLi, Li_2ZrO_3, KO functional materials etc.) and PFC/structural material (W, RAFM steels, etc.)
  - Capable of testing neutron-irradiated materials

Explosive dust evaluation for fusion vacuum vessel safety in ExCEED

- Developed for evaluating explosivity of mobilized beryllium dust for ITER VV safety (particularly in the presence of hydrogen generated by beryllium-steam reactions)
- **Unique capabilities**
  - One-of-a-kind dust explosion chamber for toxic and hazardous dusts
  - A 20 Kühner liter sphere, a standard device for dust explosion testing
  - A typical measurement test series varies the dust concentration to identify the maximum pressure and maximum rate of pressure rise and the concentration at which these occur
- Future work may include combined hydrogen/dust explosions for fusion safety



Outline

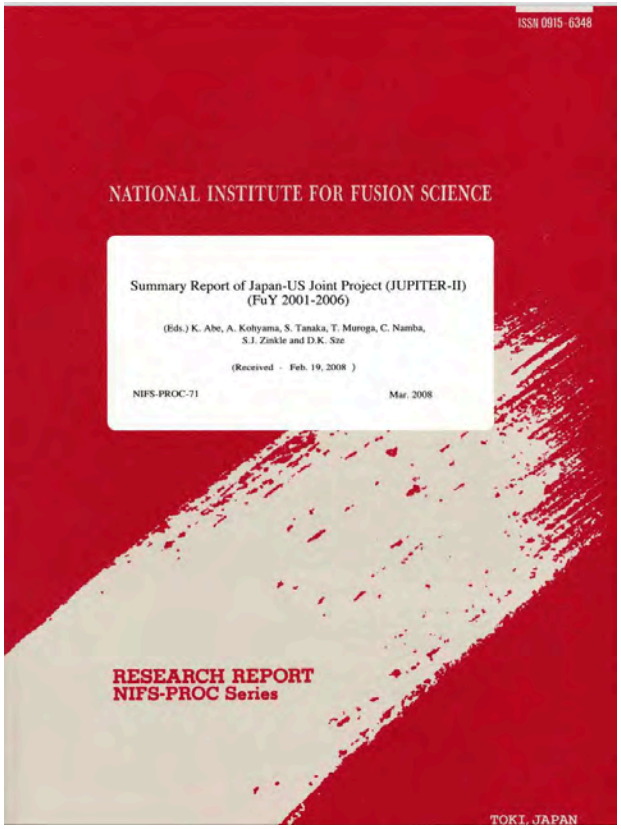
1. Motivation and overview of STAR research

2. Molten salt research at STAR during JUPITER-II project (April 2001 – March 2007)

3. Present capabilities

Overview of JUPITER-II program (Apr. 2001 – Mar. 2007)

- JUPITER-II
  - **J**apan-**U**SA **P**rogram of **I**rradiation/**I**ntegration **Te**st for Fusion **R**esearch –**II**
  - Six years (2001-2006) under the collaboration implemented between MEXT (Ministry of Education, Culture, Sports, Science and Technology) and US DOE
- Task 1: Self-cooled liquid blanket
- **Task 1-1: FLiBe system**
- (Task 1-1-A) FLiBe Handling/Tritium Chemistry
  - Experimental work with FLiBe at STAR, INL for self-cooled liquid blanket of a fusion reactor.
  - Maintaining Flibe under a reducing atmosphere is a key issue to transform TF to T_2 with a faster reaction rate compared with the residence time in blanket.
  - The purpose of the task is to clarify whether or not the Redox control of Flibe can be achieved with Be through the following reaction.
    - Be + 2 TF ➔ BeF_2 + T_2
- (Task 1-1-B) FLiBe Thermofluid Flow Simulation
  - Simulation work at U. of Kyoto and UCLA



Reference:

K. Abe, A. Kohyama, S. Tanaka, T. Muroga, C. Namba, S.J. Zinkle, and D.K. Sze "Summary Report of Japan-US Joint Project (JUPITER-II)" ***NIFS- PROC-71 (2008)***

JUPITER-II (2001-2006)
Task 1-1-A: FLiBe Chemistry Control, Corrosion, and Tritium Behavior

- **Molten salt handling and purification**
  - Developed Material Handling Protocol with Safety Emphasis
  - Proved Effectiveness of Hydro-fluorination Purification
  - Demonstrated Capabilities for Impurity Quantification
  - **Experimental procedure:**
    - BeF_2 and LiF powders were dried and weighted to the mole ratio of 2:1
    - Melted with helium gas purge first, and then with gas mixture of He, H_2, HF at 520°C to reduce inherent oxides
    - The salt was transfered to another vessel through filtered through 60 µm metal mesh frit

Table 1 Impurities in ingredients and final salt

	O (ppm)	C (ppm)	N (ppm)	Fe (ppm)	Ni (ppm)	Cr (ppm)
BeF2	5700	<20	58	295	20	18
LiF	60	<2	78	100	30	4
Flibe	560	10	32	260	15	16

- **Mobilization studies**
  - Developed and Validated Transpiration System for Vapor Pressure Measurement of Molten Salts
  - Measured FLiBe Vapor Pressure at Low-temperature range Relevant to Fusion Blanket Designs
  - **Experimental procedure:**
    - Mobilization test was performed with Ar, air, and moist air in inert gas glove box.
    - (Ar test) conducted at 500, 600, 700, and 800°C with 25 sccm Ar flow
    - (Air test) conducted at 500, 600, 700, and 800°C with 25 and 50 sccm air flow
    - (Moist air test) conducted at 600, 700, and 800°C with 25 and 50 sccm moist air flow
    - Both Ni and glassy carbon crucibles were used

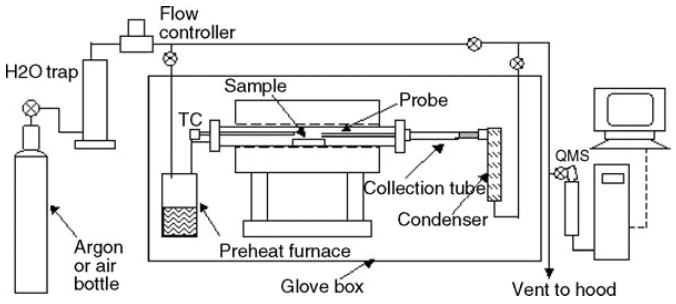


Fig. 1. Transpiration test setup.

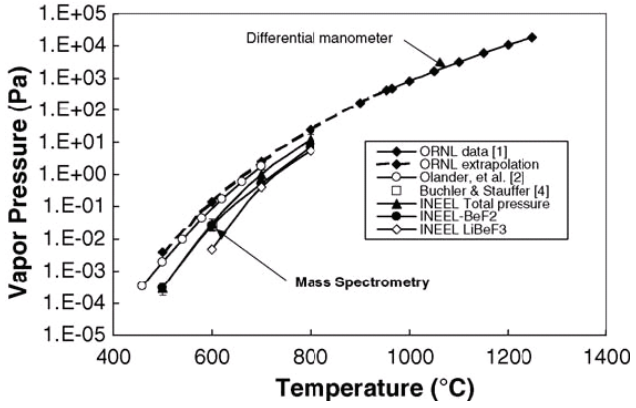


Fig. 2. Total pressure over Flibe.

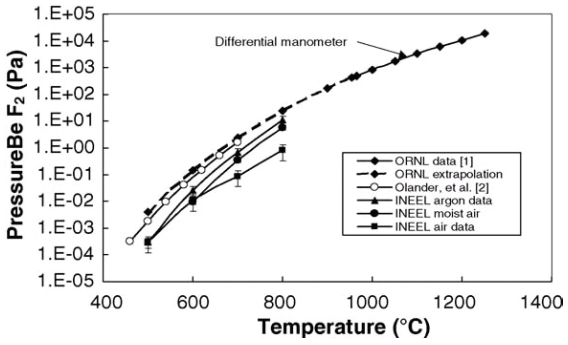


Fig. 3. BeF_2 pressure in various environments.

JUPITER-II (2001-2006)
Task 1-1-A: FLiBe Chemistry Control, Corrosion, and Tritium Behavior

- **Redox control**
  - Demonstrated active control of the fluorine potential in FLiBe/Nickel systems using metallic Be
  - Proved the inhibition of FLiBe corrosion of Reduced Activation Ferritic Steel in static conditions
  - **Experimental procedure:**
    - The purpose of the task is to clarify whether or not the Redox control of Flibe can be achieved with Be through the Be + 2 TF ➔ $BeF_2 + T_2$ reaction
    - HF was bubbled with He and H_2 through FLiBe with various concentration of dissolved Be (cylindrical Be rod, 0.76 cm OD and 3 cm long) .
    - Ni crucible and Ni tubes were used and all the wet surface was Ni coated

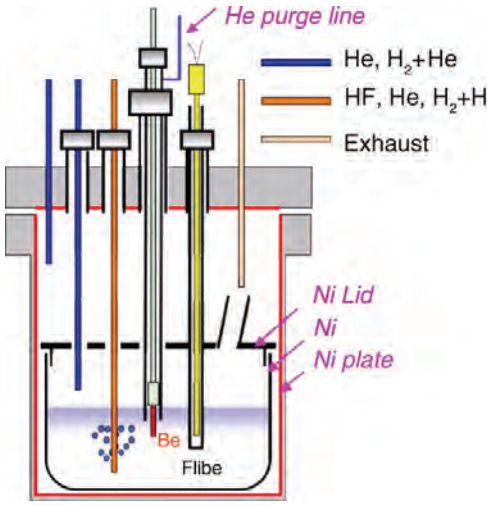
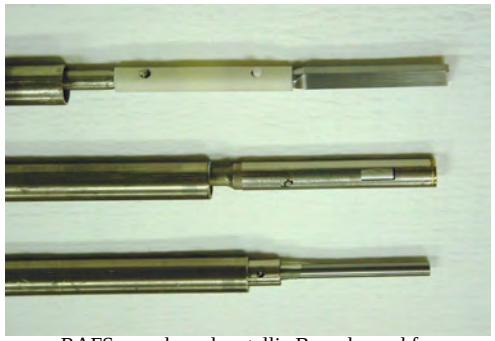


Fig. 5. Reactor for measuring redox



RAFS sample and metallic Be rods used for Redox and corrosion tests before and after immersion in FLiBe bath

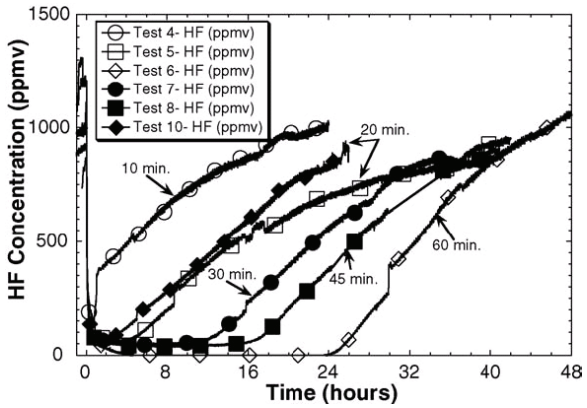


Fig. 6. HF concentration measured by QMS on the outlet of the REDOX experiment for several Be immersion times.

JUPITER-II (2001-2006)
Task 1-1-A: FLiBe Chemistry Control, Corrosion, and Tritium Behavior

- **D_2 and T_2 permeation**
  - Measured transport properties (diffusivity and solubility) of D_2 and T_2 in FLiBe between 550 and 700 C
  - Investigated the effect of FLiBe Redox condition on T_2 transport
  - **Experimental procedure:**
    - (D_2 test) was conducted in a cylindrically symmetric dual probe permeation pot
      - Ni crucible and Ni tubes are used
      - at 600 and 650°C at 9.0x10^4 Pa in NI Probe 1
    - (T_2 test) was conducted in a permeation pot with 2mm thick Ni membrane
    - at 550, 600, 700 and 800°C with 1 and 20 sccm (0.1 ppm-10 vo..% T_2/Ar)
    - Measured with QMS and GC

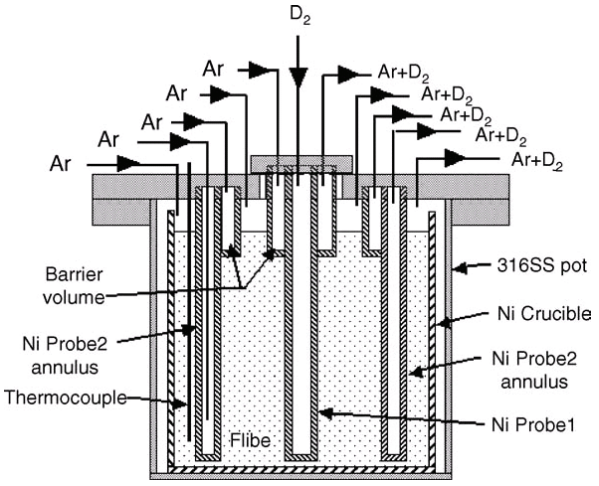


Fig. 10. Schematic illustration of cylindrically symmetric, permeation probe assembly.

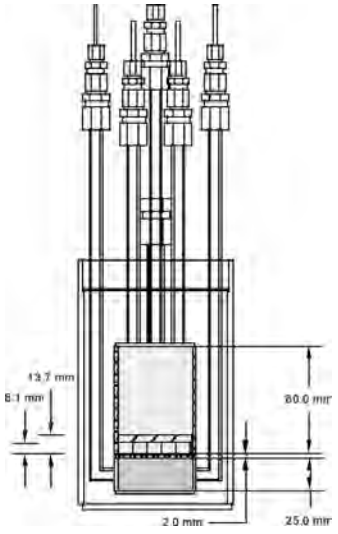


Fig. 1. Permeation cell scheme with relevant dimensions.

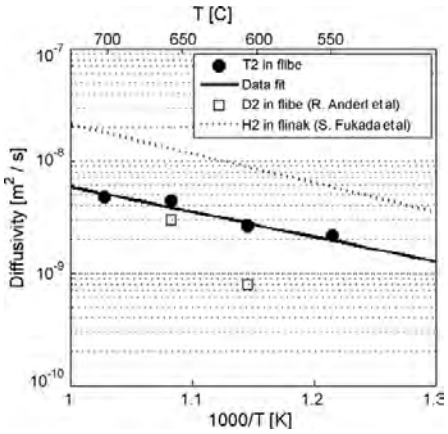


Fig. 3. Measured T diffusivity in flibe.

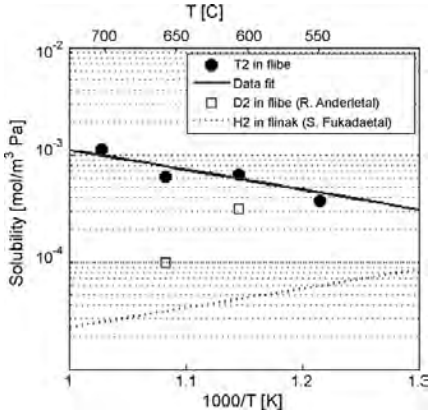


Fig. 4. Measured T solubility in flibe.

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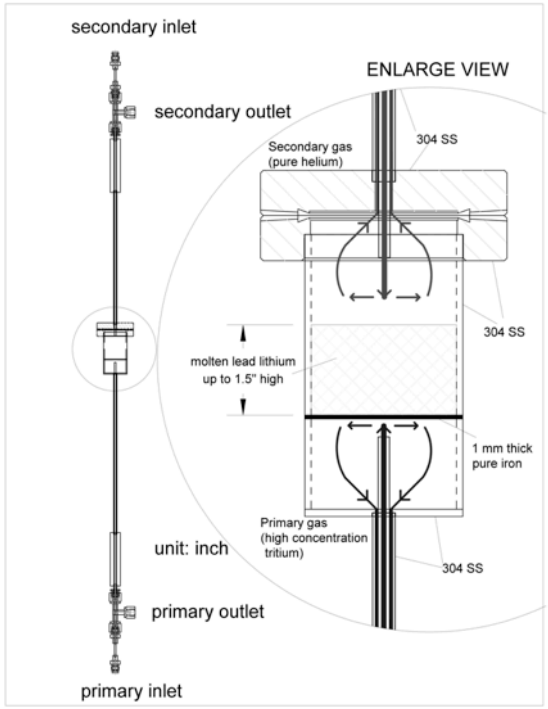
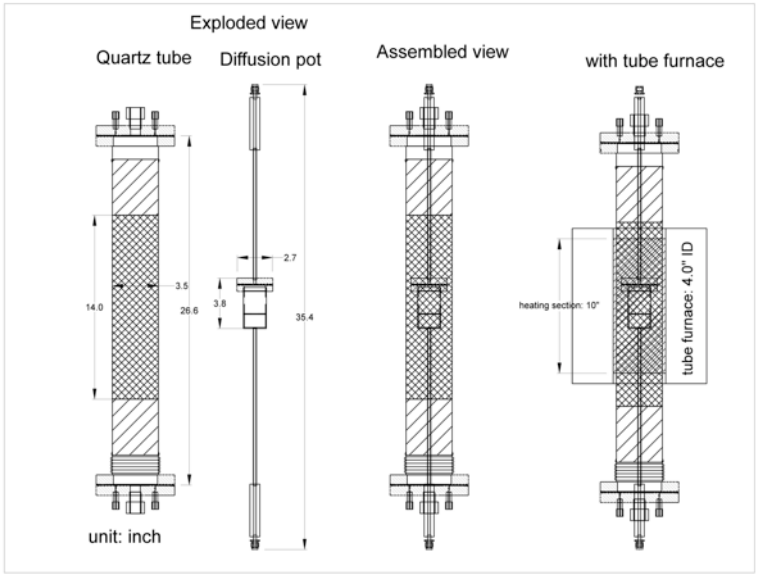
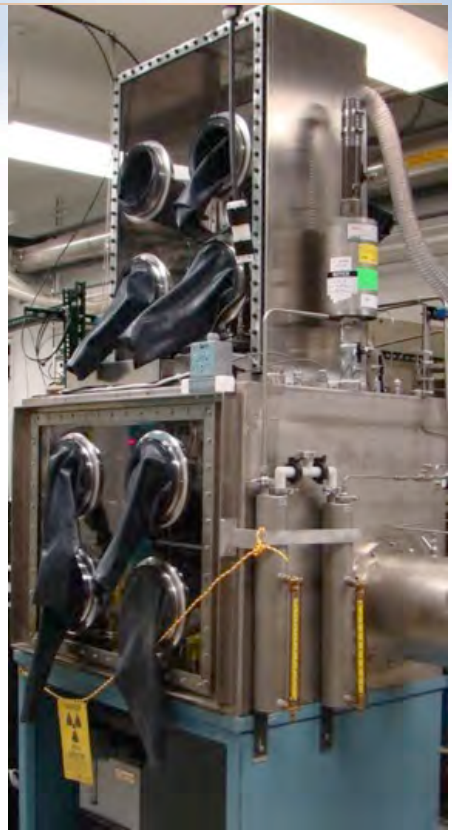
TGAP design is based on the FLiBe permeation pot

Tritium Gas Absorption Permeation experiment



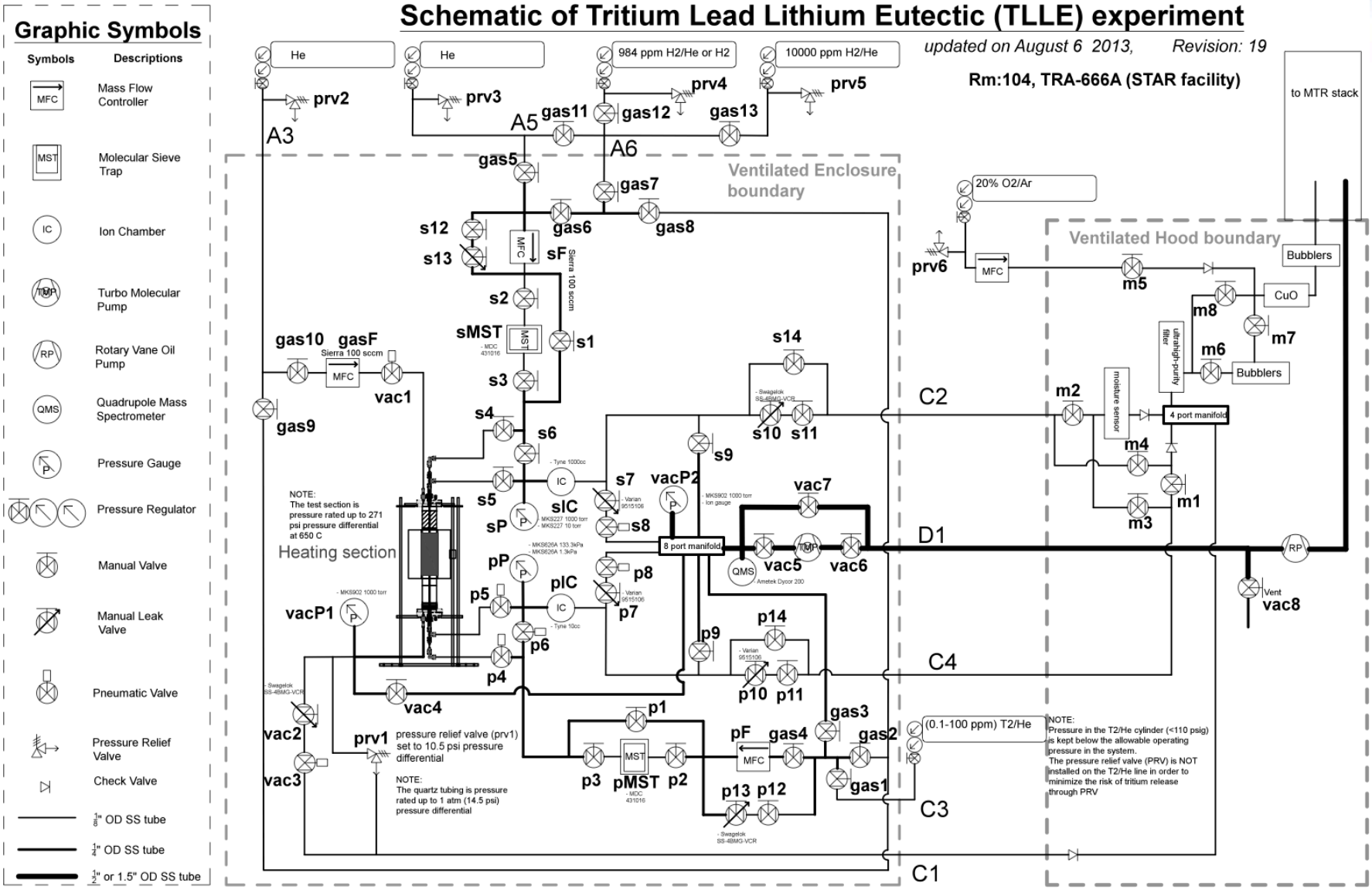




Unique capabilities

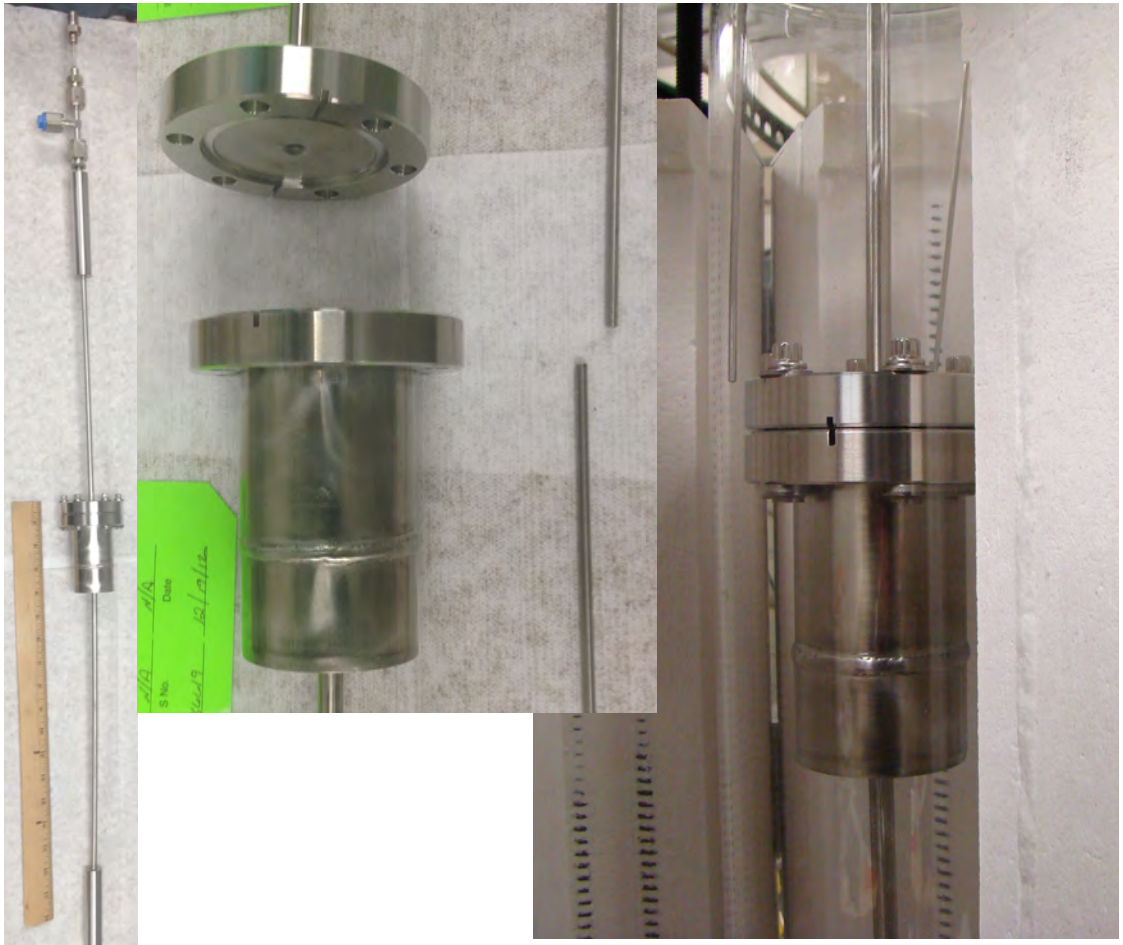
- Designed to measure transport properties (e.g. diffusivity, solubility, and permeability) of tritium at realistic blanket conditions (e.g. low tritium partial pressure < 1000 Pa) for disc geometry sample
- Capable of testing liquid breeder material (e.g. PbLi and FLiBe) and disc shaped metal
- Uniform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

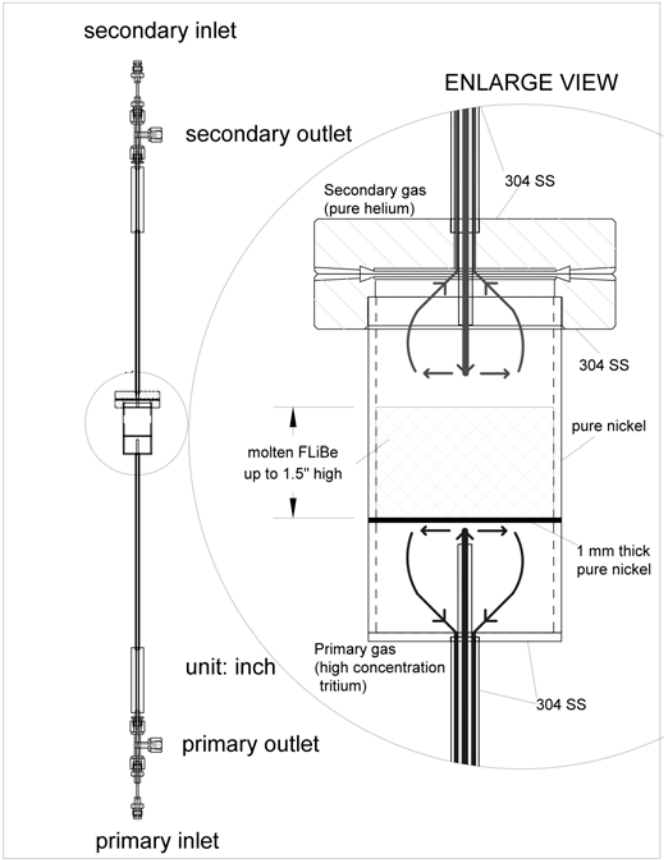
Schematic of tritium gas permeation system



Test section for Lead Lithium Eutectic (LLE)

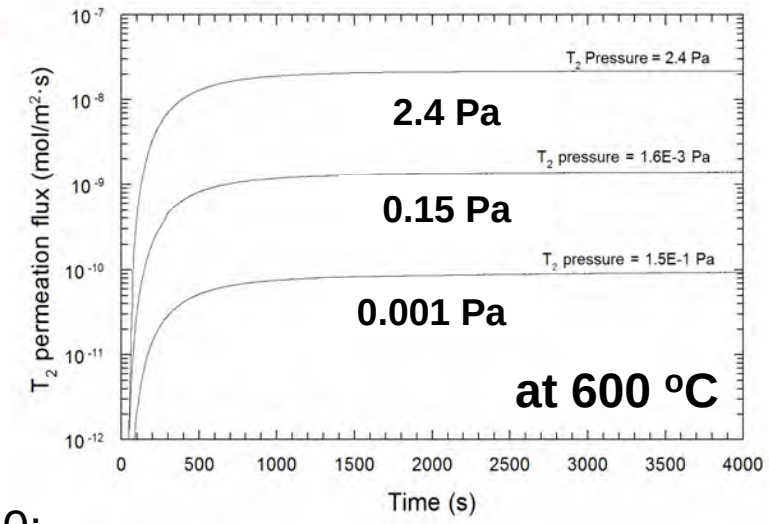
- **Photo of Test section for lead lithium eutectic**



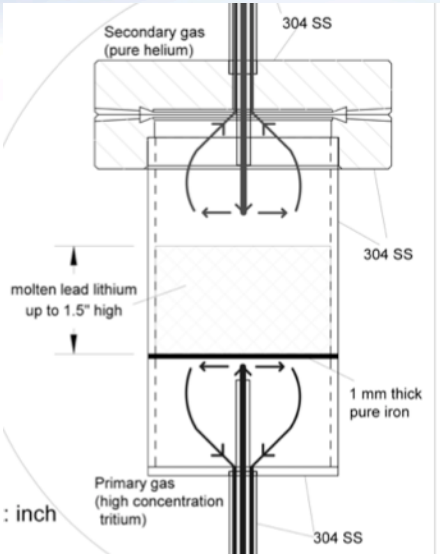


- This test section for FLiBe can be fabricated based on LLE drawing with FLiBe compatible material (e.g. nickel) on all wet surface

Experimental procedure (Fe/LLE)



H_2/He

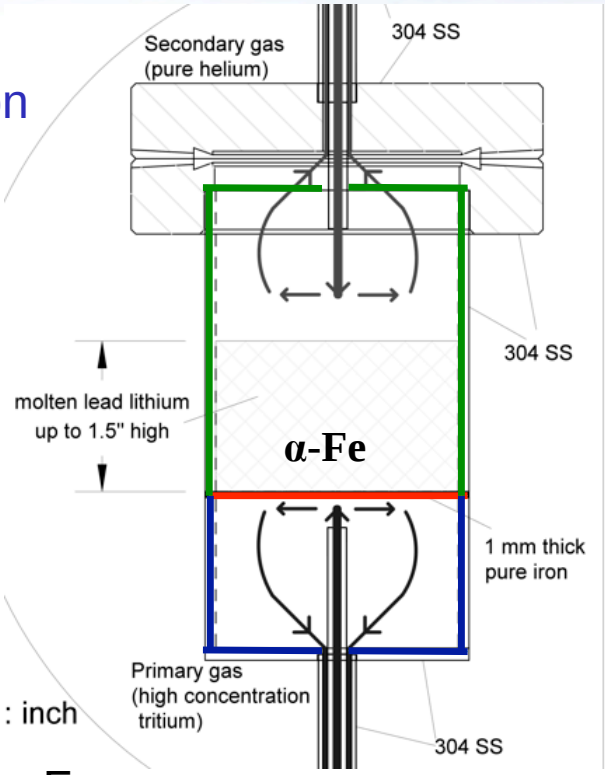


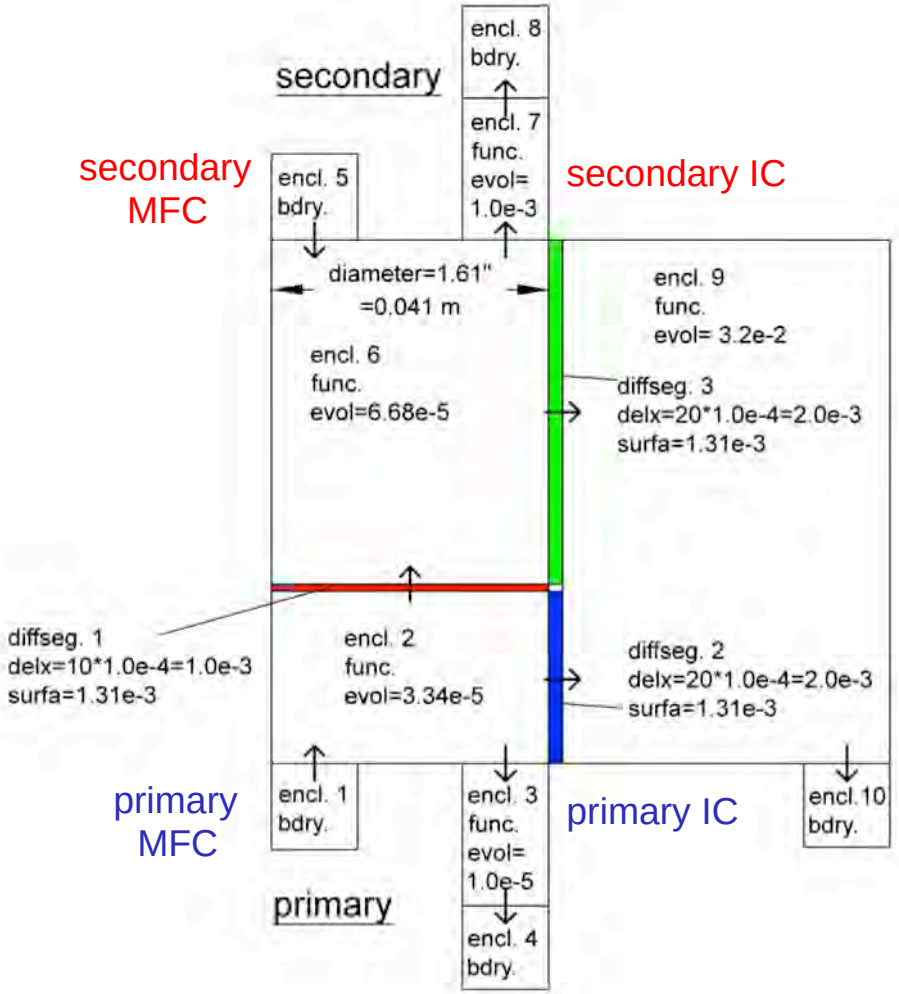
T_2/He

- t < 0:
  - Primary/Secondary was purged with **1 % H_2/He 100/200 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.

TMAP configuration for (1 mm) α-Fe

Experiment configuration

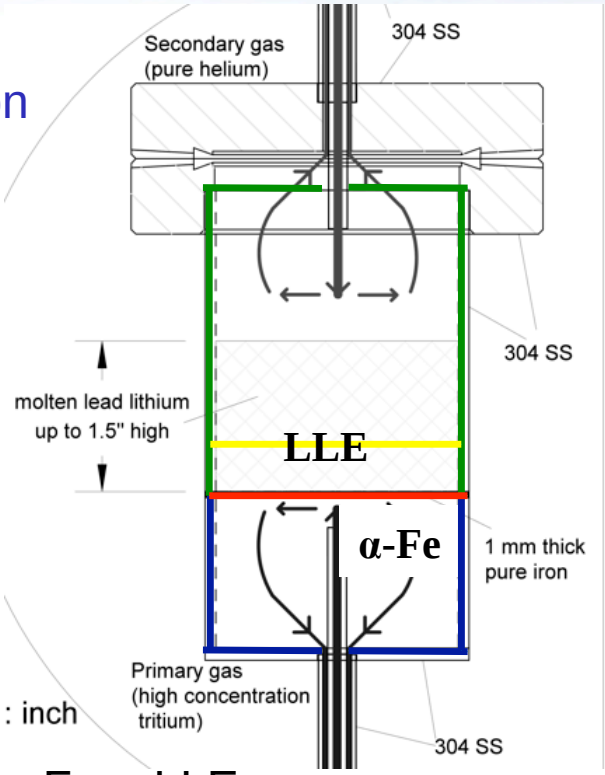


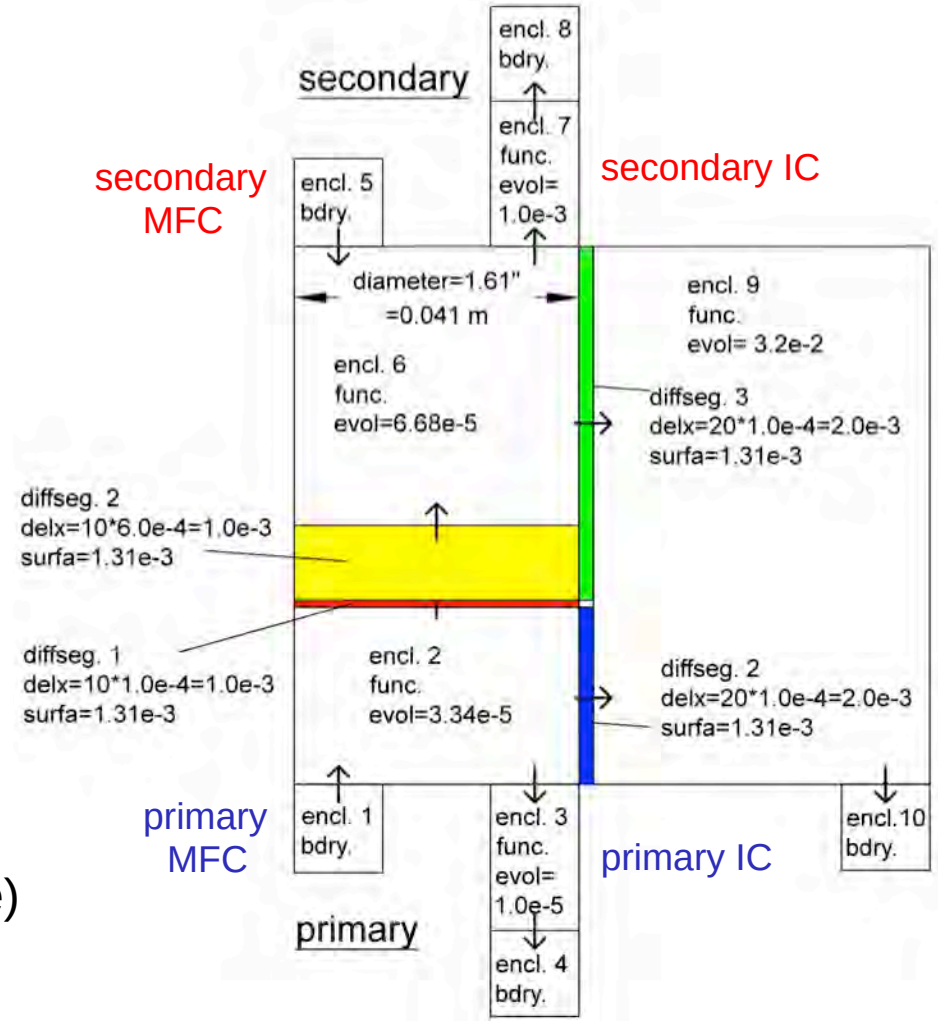


- TMAP for α-Fe
  - 1D geometry
  - 4 enclosure species (H_2, T_2, HT He)
  - 2 diffusion species (H, T)
  - 3 segments
  - 10 enclosures

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

Experiment configuration



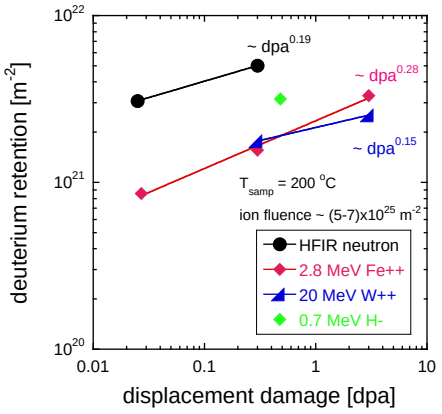


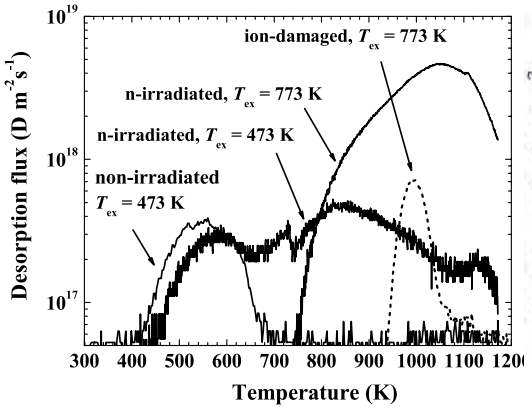
- TMAP for α-Fe + LLE
  - 1D geometry
  - 3 enclosure species (H_2, T_2, HT, He)
  - 2 diffusion species (H, T)
  - 4 segments
  - 10 enclosures

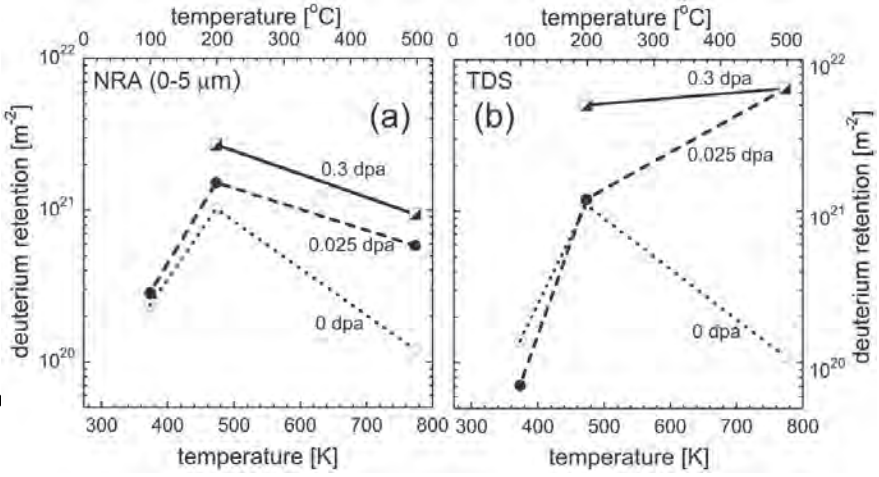
History of HFIR irradiation in tungsten:

Tritium retention in HFIR neutron-irradiated tungsten

- ✓ **US-Japan TITAN program (2007-2013):**
  - Low-temperature (<100 °C) low-dose (0.025 & 0.3 dpa) HFIR neutron-irradiated tungsten
  - Tritium was trapped in bulk (>10 μm), and retention increases at high temp. (500 °C)
  - Measurement of microstructural evolution and characterization of radiation defects and defect annealing before/after plasma exposure is required to reveal trapping mechanism.
- ✓ **US-Japan PHENIX program (2013-2018):**
  - High-temperature (500, 800, & 1200°C) medium-dose (0.3 &1.5 dpa) HFIR neutron-irradiated tungsten RB* irradiation with Gd thermal neutron shield
  - Positron annihilation spectroscopy (PAS) at ORNL and INL will characterize radiation defects and before/after plasma exposure to reveal defect annealing mechanism.
  - Nuclear reaction analysis, thermal desorption spectroscopy, and PAS is used to determine tritium migration depth and trap density for tritium retention assessment.







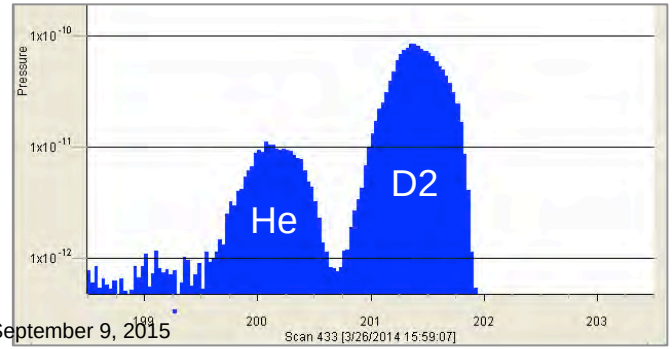
M. Shimada et.al. *Fus. Eng. Des.* **87**(2012) Y. Hatano et.al. *Nucl. Fusion* **53**(2013) M. Shimada et.al., *Nucl. Fusion* **55**(2015)

Thermal desorption spectroscopy (TDS)

- Both TPE and NIMIIX study retention.
- To investigate retention,

$$R = \frac{\Phi_{out}}{\Phi_{in}}$$

- Heat samples up to 1100ºC
  - *Vacuum infrared tube furnace*
  - *Linear ramp rate*
  - *6 calibrated leaks (x3 He and x3 D2)*
- Analyze residual gases
  - *Two quadrupole mass spectrometers*
  - *High resolution QMS can distinguish:*
    - D2: 4.0282 amu 
    - He: 4.0026 amu 



Summary

- STAR at INL is DOE less than Hazard Category 3 Nuclear Facility for both fission and fusion safety R&D.
  - Maximum tritium inventory allowed 1.6 gram (15,390 Ci)
  - Capable of handling tritium, beryllium, FLiBe, and activated material
- FLiBe was extensively studied at the STAR facility during JUPITER-II program (2001-2006)
- STAR still operates 2x inert (Ar) gas gloveboxes for FLiBe use
- TGAP is capable of studying tritium permeation and absorption in FLiBe.
  - Design is based on the tritium permeation pot of FLiBe
- TPE, NIMIIX and SGAS can be used to investigate deuterium (tritium at TPE) retention in irradiated graphite for FHR.
- STAR can help develop tritium control and capture in FLiBe to enhance technical readiness level of FHR technology

Supporting slides

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]

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

- Mass transport property data used for H permeation in PbLi from Okitsu[5]
- 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. al., JNM v.191-194 (1992) 258
[5] Okitsu et.al., FED v.87 (2012) 1324