Evaluation of In-Situ Tritium Transport Parameters for Type 316 Stainless Steel during Irradiation

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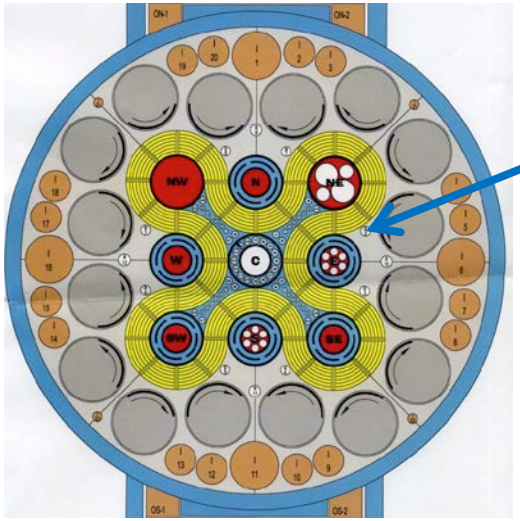
Motivation and Scope

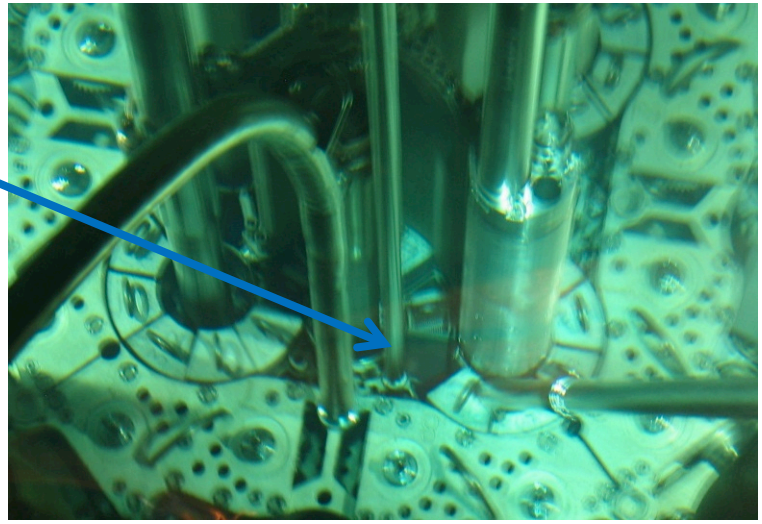
- TMIST-2 Experiment
  - Measured in-reactor steady state tritium permeation through Type 316 stainless steel as a function of tritium partial pressure and temperature
  - Tritium permeation irradiation enhancement of ~3X was observed relative to ex-reactor data and accepted literature values
- Transient Analysis of TMIST-2 Data
  - Sought to elucidate insight into the mechanism for the observed permeation enhancement
  - Three independent, but related, methods were used to analyze the TMIST-2 transient data

TMIST-2 In-Reactor Tritium Permeation Experiment



ATR Core Cross Section

TMIST-2 Experiment in ATR at Position B-2

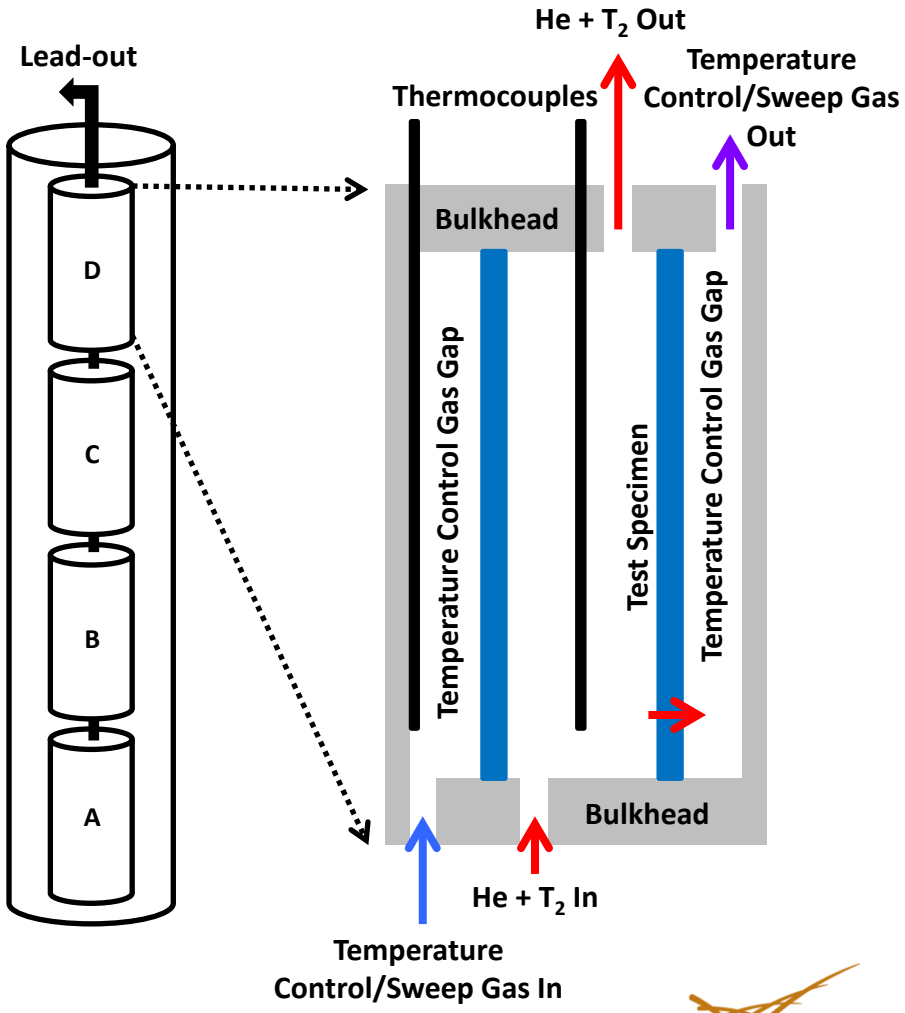


TMIST-2 Lead-Out in ATR Core

- Irradiated for five cycles (257.5 EFPD at 18 MW_t) to a dose of 1.63 dpa-304SS
- Test conditions included tritium partial pressures of 0.1, 5, and 50 Pa; temperatures of 292 and 330° C
- Experiment designed by PNNL, capsules fabricated at PNNL, test train fabricated at INL

TMIST-2 In-Reactor Tritium Permeation Experiment

- Four independent active temperature-controlled capsules separated by bulkheads welded to outer pressure tube
- One test specimen per capsule with active permeating length of 10 cm
- Two thermocouples per capsule
  - Two junctions on centerline
  - Two junctions outside temperature control gas gap
- Gas lines and thermocouples brazed at bulkheads to separate capsule gas volumes
- Test specimens welded to bulkheads with areas outside active length held at much lower temperature and coated with Al to minimize extraneous permeation



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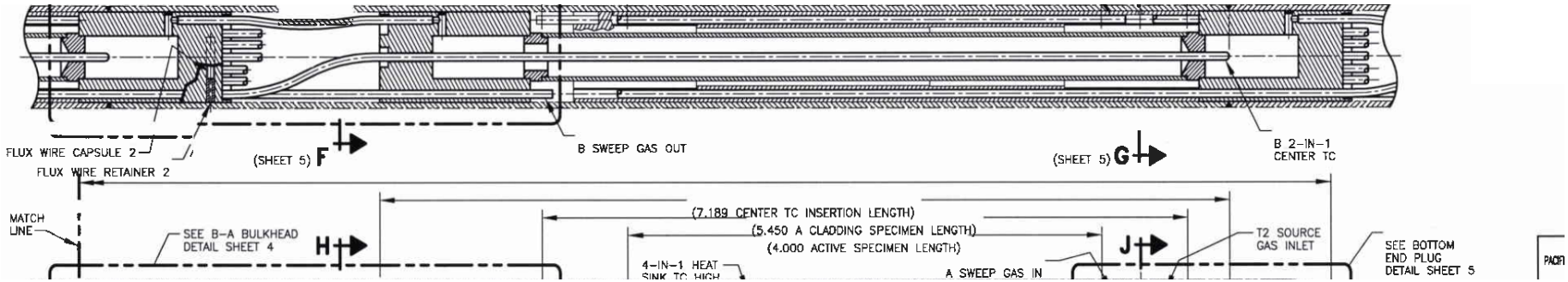
TMIST-2 In-Reactor Tritium Permeation Experiment

- Copper sweep gas outlet lines used to minimize tritium loss between capsule and ion chambers/bubblers
- Tritium partial pressure and specimen temperature changed independently in stepwise fashion during irradiation
- All combinations of temperature and pressure tested at least twice at difference fluence



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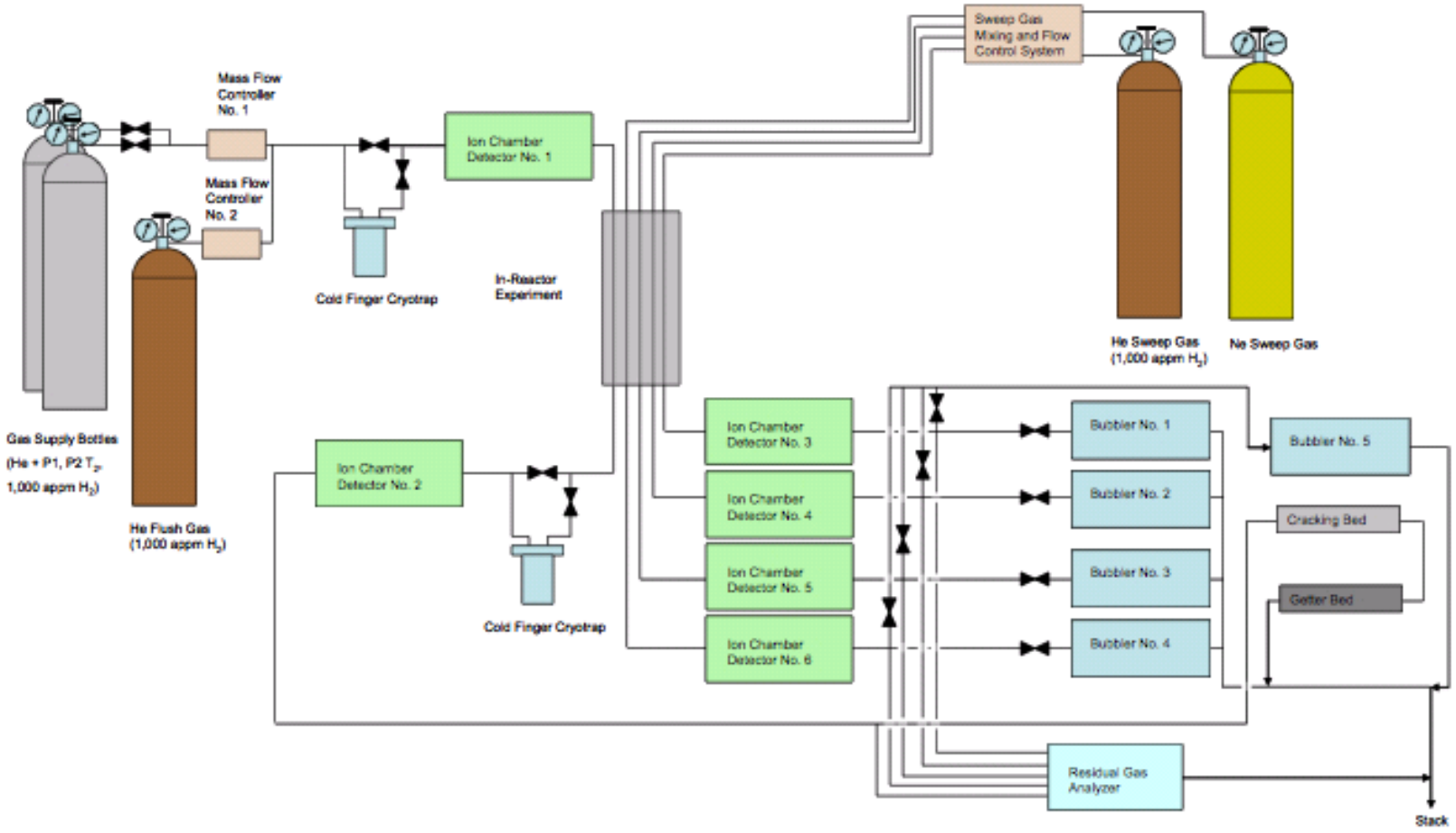
TMIST-2 In-Reactor Tritium Permeation Experiment

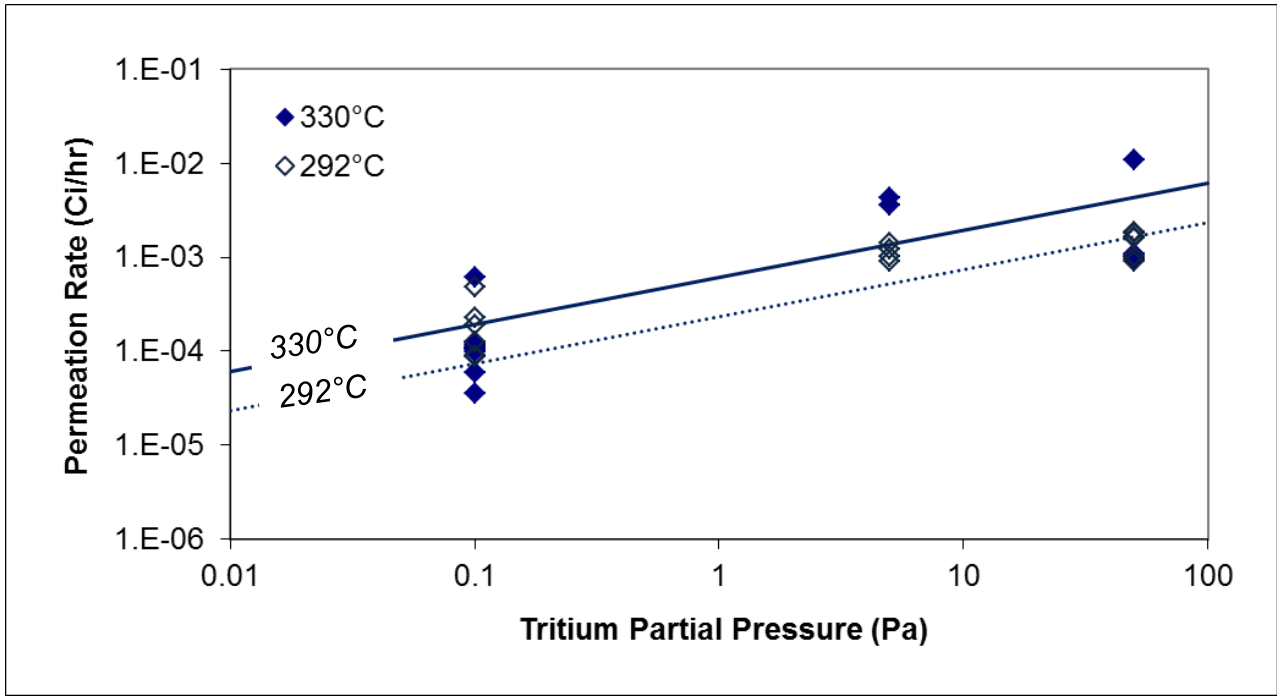


- Ion chambers used to establish steady-state (typically 6-8 days)
- Steady-state permeation rate quantified by repeated liquid scintillation counts of tritium captured in bubblers

In- vs. Ex-Reactor Permeation Behavior

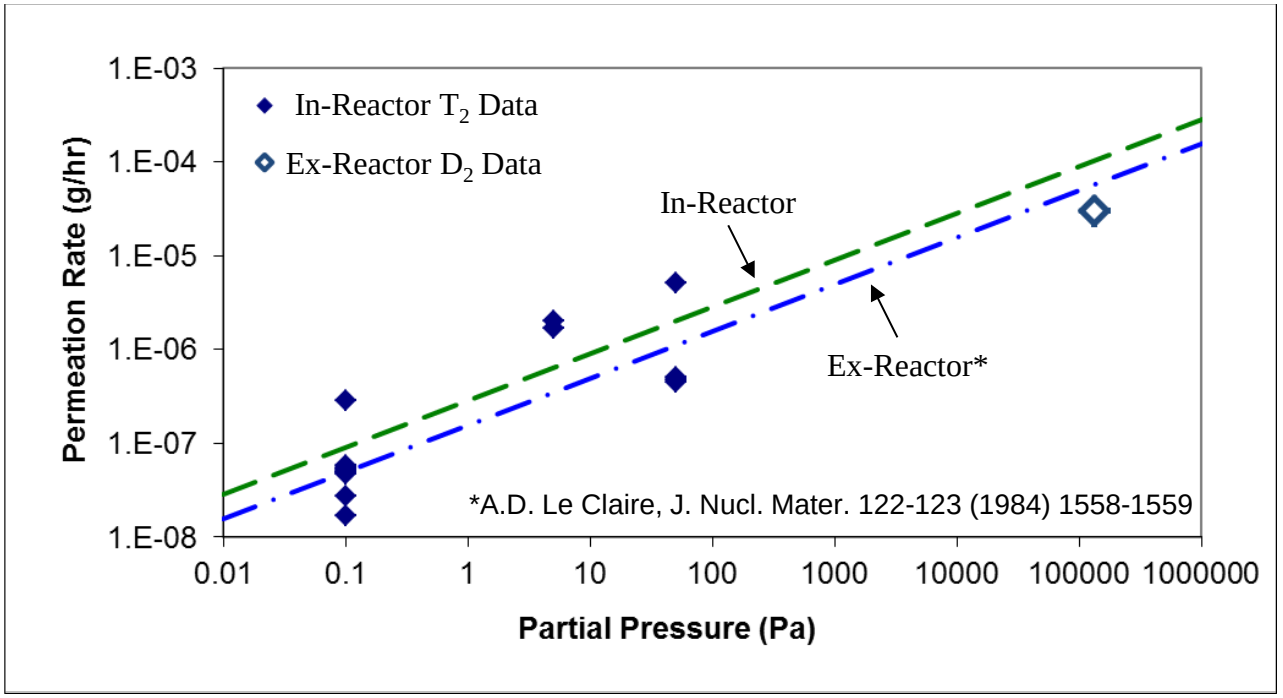
- Ex-reactor permeation measurements
  - > 100 Pa → Diffusion-limited → $P^{0.5}$
  - < 100 Pa → Surface-limited → P^{1}
- Experiment designed to measure in-reactor steady-state permeation rate and pressure dependence to elucidate key performance phenomena
  - Direct dissociative chemisorption
    - Associated with diffusion-limited permeation
    - Disrupted ex-reactor by:
      - Surface impurities at low pressure
      - Oxide films
  - Radiation-enhanced dissociation
    - Radiolysis of T_2 in gas phase
    - Physical or chemical changes in surface in-reactor

Results of In-Situ Permeation Measurements



- Weak temperature dependence over specified range
- $P^{0.5}$ dependence observed
  - Suggests diffusion-limited permeation over specified pressure range

Results of In-Situ Permeation Measurements



- $P^{0.5}$ dependence permits extrapolation of ex-reactor correlation
  - No transition from diffusion- to surface-limited behavior < 100 Pa
  - Indicates in-reactor enhancement in permeability of ~3X

Permeability, Diffusivity, and Solubility

- Permeating flux (J) is dependent on diffusivity (D) and solubility (S)

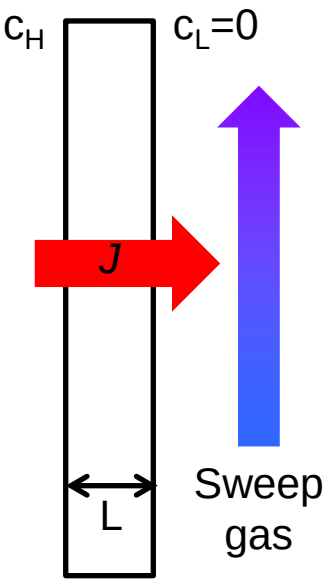
$$J = -D\frac{dc}{dx} = -D\frac{c_L - c_H}{L} = D\frac{c_H}{L}$$

$$K = \frac{[H_{(m)}]}{[H_{2(v)}]^{1/2}} = \frac{c_H}{P^{1/2}} \Rightarrow c_H = KP^{1/2} = SP^{1/2}$$

$$\therefore J = \frac{DSP^{1/2}}{L}$$

Permeability, Φ

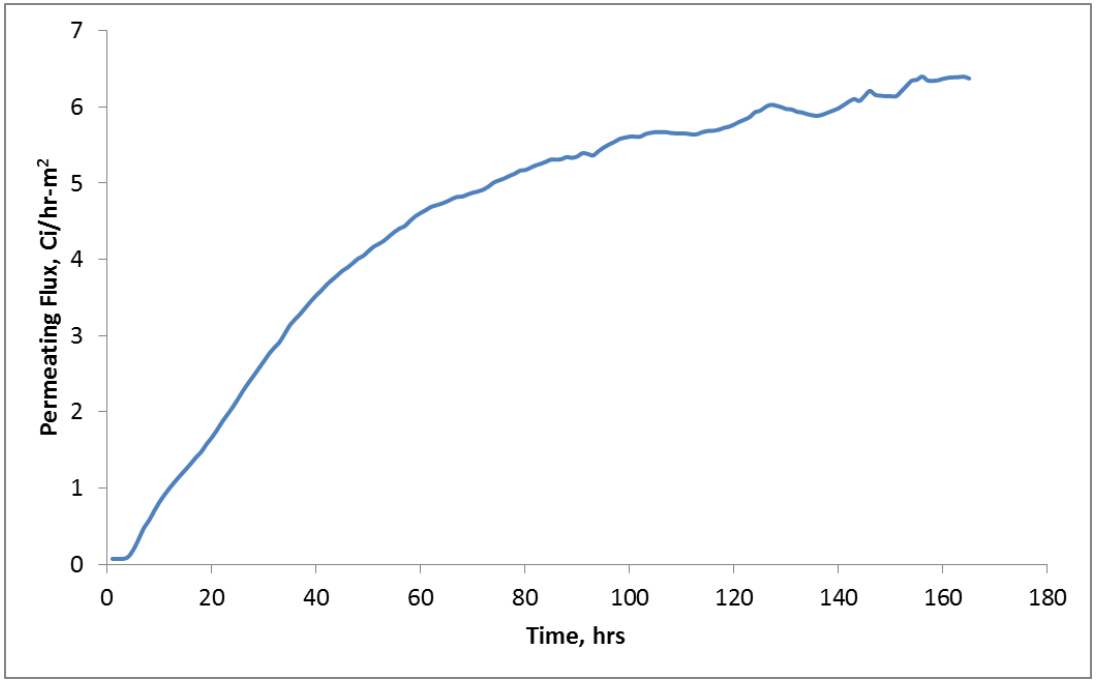
$$\Phi = DS$$



- Is the ~3X radiation enhancement in permeability (Φ) due to enhanced D, S, or both?

Analytical Approach

- Attempt to extract diffusivity data by analyzing ion chamber transient data
  - Test steps exhibiting increase in flux only
  - Still evaluating methodologies for analyzing steps with decrease in flux
  - Assume slab geometry due to favorable aspect ratio per Kishimoto (1985)



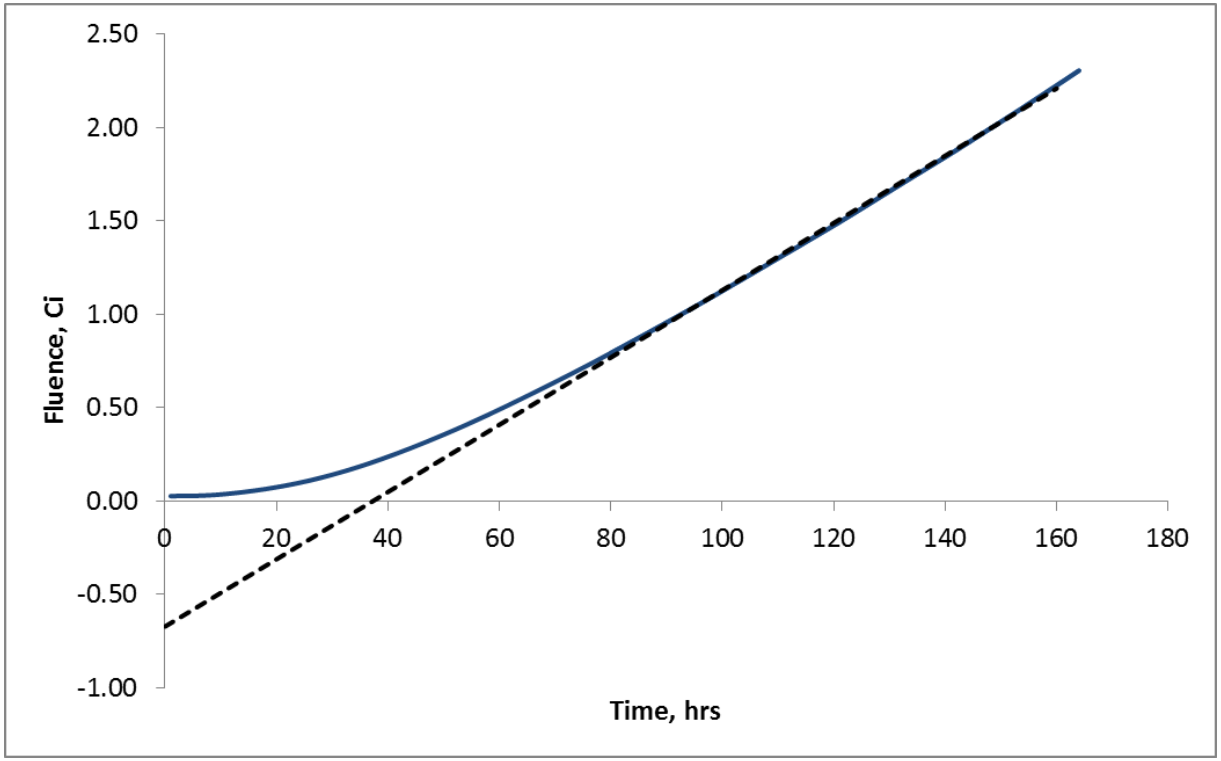
Considerations and Assumptions

- In-reactor specimens located >15 m from ion chambers
  - Transport time
    - Quantified by time lag measured in ion chambers positioned at supply-side inlet and outlet (≤3 hr)
    - Transport time considered negligible relative to time needed to reach steady-state (≥144 hr)
  - Isotopic exchange
    - Possibly significant due to large surface area of tubing walls between test specimens and ion chambers
    - Tubing swamped with protium between test steps to minimize tritium cross-talk between permeation measurements
- Surface processes
    - Measured $P^{0.5}$ dependence indicates that surface decomposition and recombination are fast relative to bulk diffusion

Analysis Methods

- Time-lag analyses (Frisch 1957)
  - Integrate flux transient to obtain plot of quantity vs. time
  - Fit linear (t➔∞) portion of curve
  - Locate t-axis intercept and calculate *D*
  - Use measured permeability and estimated *D* to calculate *S*

$$t = \frac{L^2}{6D}$$

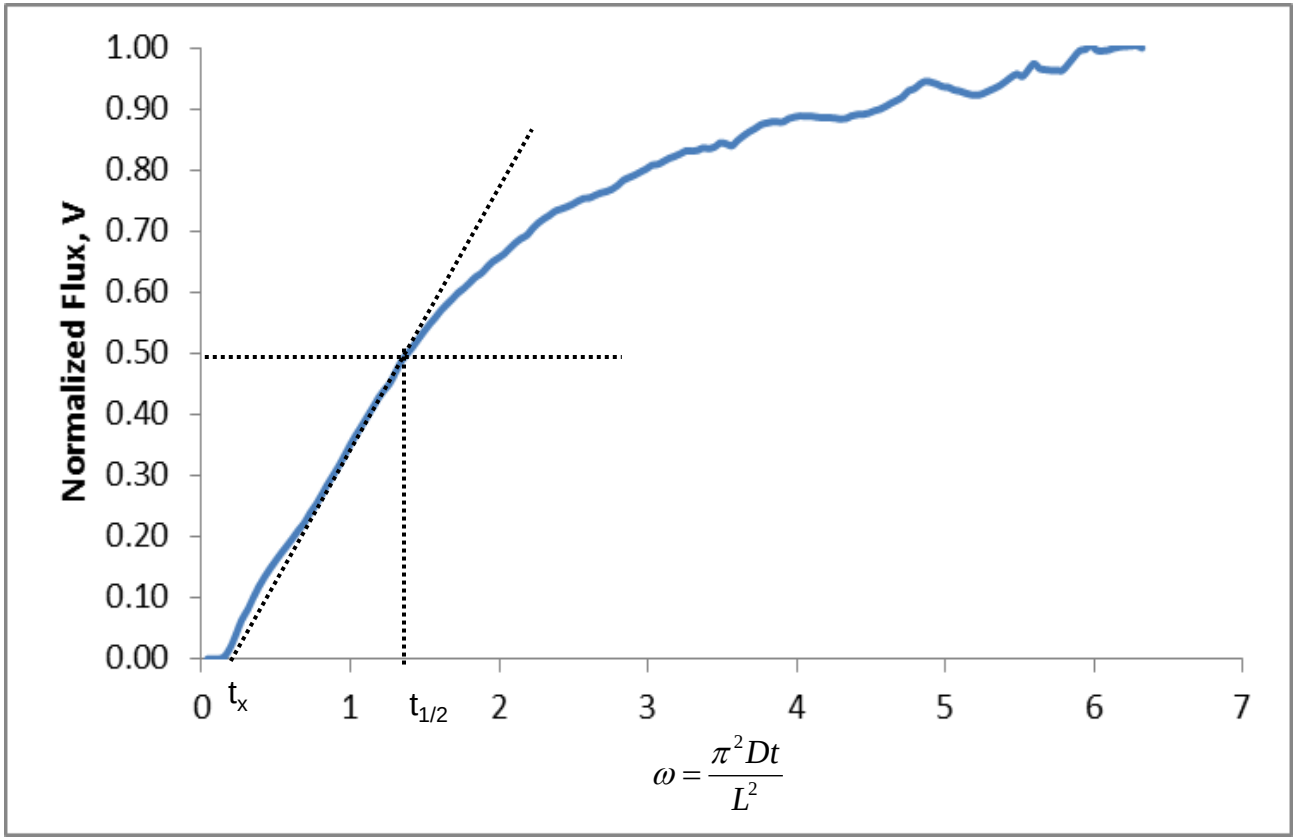


Analysis Methods

- Rise-Time Analysis (Parker 1961)
  - Used for measuring thermal diffusivity via the flash method
  - Plot normalized flux (*V*) vs. ω
  - Solve for *D* using half-height (*V*=0.5) and intercept methods

$$D = \frac{1.38L^2}{\pi^2 t_{1/2}}$$

$$D = \frac{0.48L^2}{\pi^2 t_x}$$

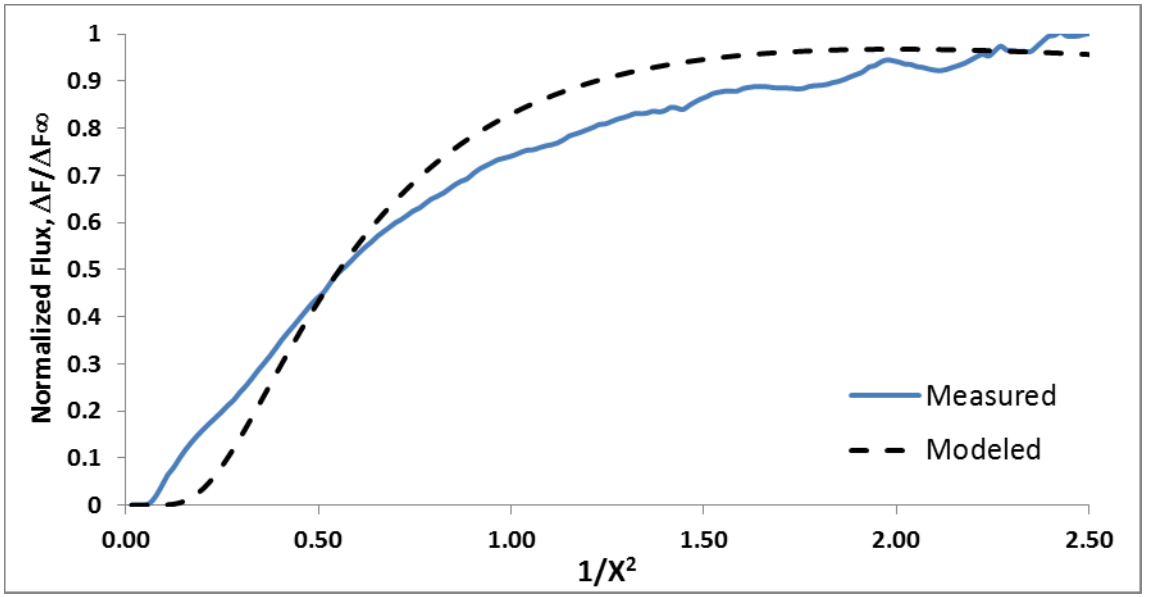


Preliminary Results

- Exponential Analysis (Pasternak et al. 1970)
  - Plot normalized flux versus function of time
  - Obtain best-fit *D* to approximate experimental data

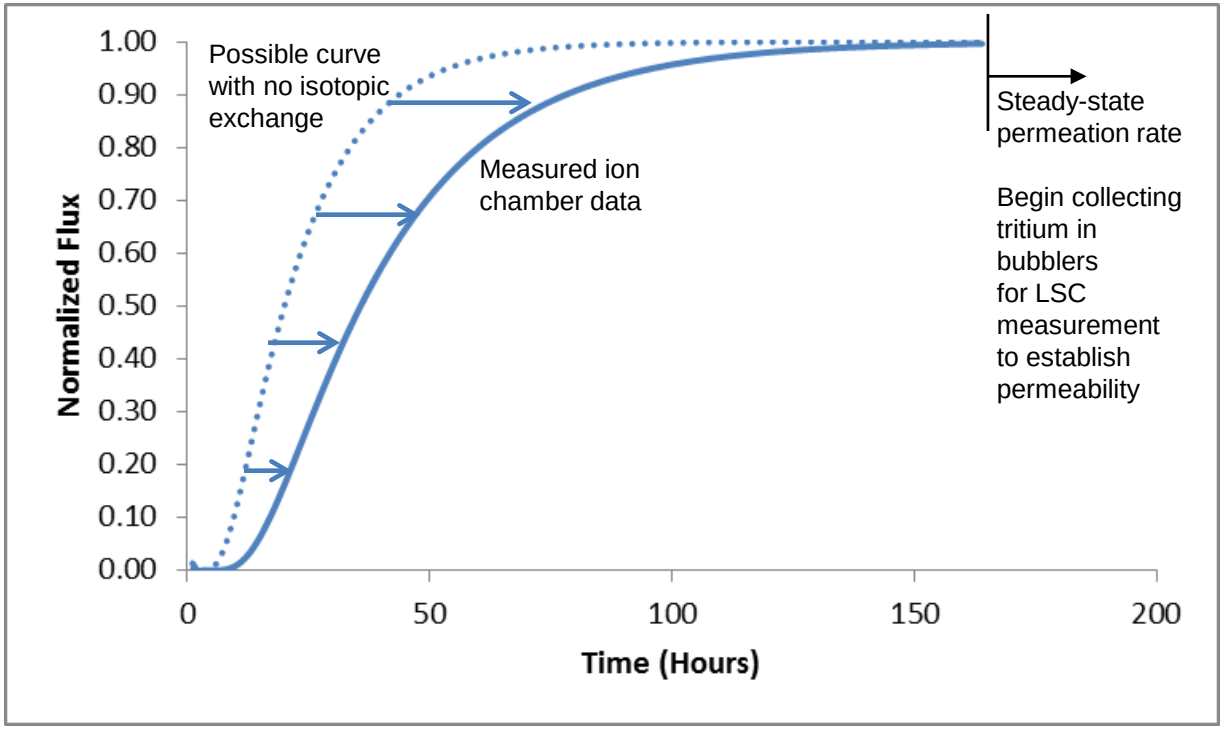
$$X^2 = \frac{L^2}{4Dt}$$

$$\frac{\Delta F}{\Delta F_\infty} = \left(\frac{4}{\sqrt{\pi}}\right) X \exp\left(-X^2\right)$$



Preliminary Interpretation of Results

- Initial estimates of *D* based on rise-time, time-lag, and permeating membrane model:
  - Reasonable agreement between methods (~3.5x10^{-9} cm^2/s)
  - Lower (~100X) than accepted ex-reactor values (~1x10^{-7} cm^2/s)
    - No physical reason to expect in-reactor reduction in *D*
    - Would require solubility enhancement >100X to account for enhanced, in-reactor permeability
    - Non-physical result suggests possible distortion in response curve due to isotopic exchange



Summary and Conclusions

- In-reactor permeability observed to be enhanced by ~3X relative to ex-reactor permeability
- $P^{0.5}$ pressure dependence observed at low pressure (e.g. <100Pa) in-reactor
  - Consistent with diffusion-limited permeation
- Preliminary analyses of transient ion chamber data reveals non-physical results
  - Possible time-lag from isotopic exchange on tubing walls between test specimen and ion chamber
- Next steps
  - Some estimate of ion exchange time lag may be possible from comparison of ion chamber data on inlet and outlet sides (recent work by Longhurst)
  - Any correction for isotopic exchange time lag will be a large fraction of measured time constant, resulting in significant uncertainty