

A Quantitative Case for Direct Internal Recycle of Deuterium & Tritium Through Permeation

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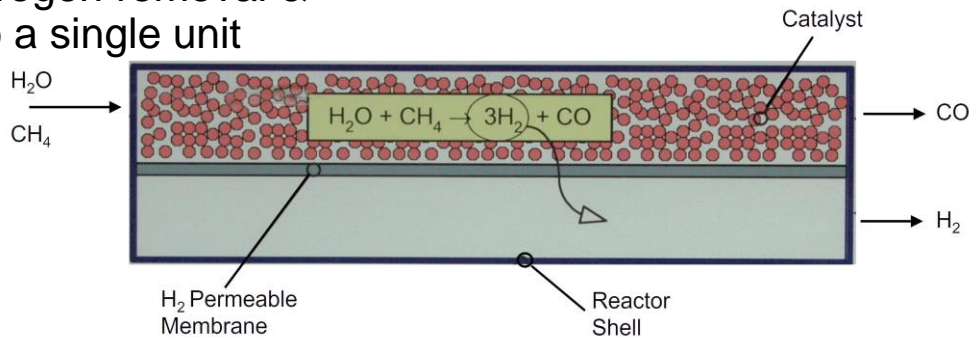
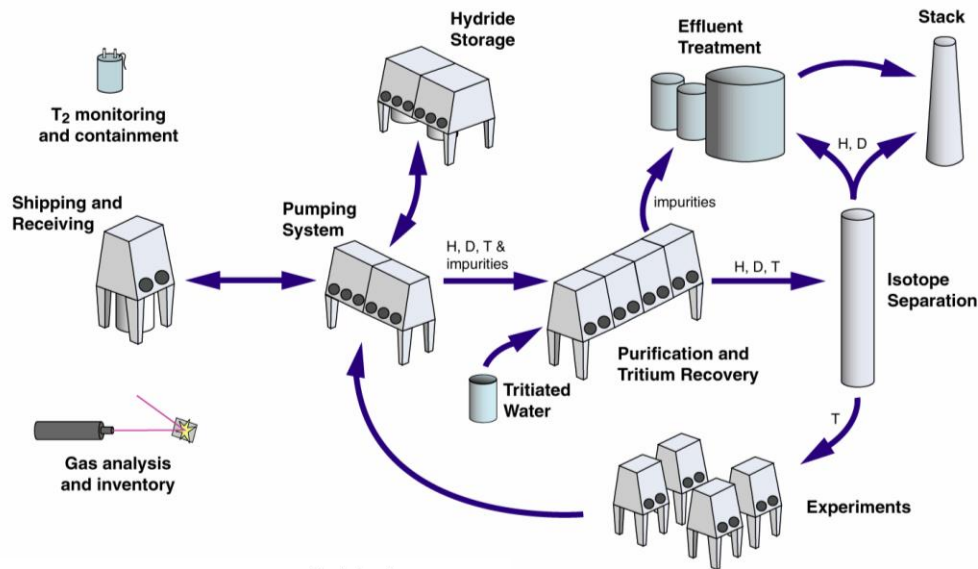
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LANL fusion fuel cycle background

Tritium Systems Test Assembly (1977-2008)

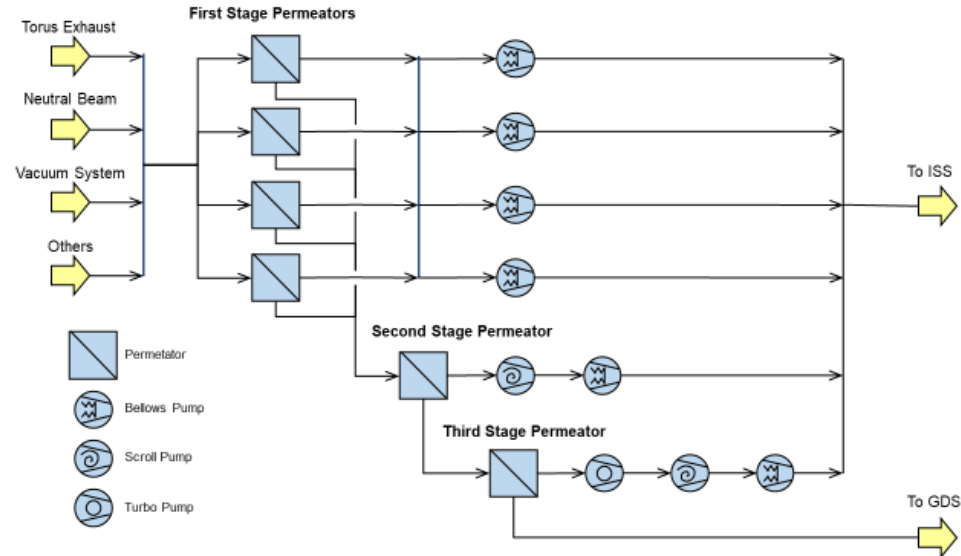
- “Everything but the reactor”
 - Storage, processing, pumping
 - Investigated all tritium streams
 - Processing sized at 1/10th ITER scale
- Palladium Membrane Reactor
 - Developed at LANL
 - Combines hydrogen removal & separation into a single unit



LANL fusion fuel cycle background

Tokamak Exhaust Processing System for ITER: Preconceptual Design

- Dynamic ASPEN models for TEP system design & sizing
- R&D for:
 - Catalyst selection
 - Permeators, Cat beds, Palladium Membrane Reactor
- Tritium-compatible vacuum pump models that could be combined into a pump train
- Lab-validated unit operations models specific to the ITER fuel cycle



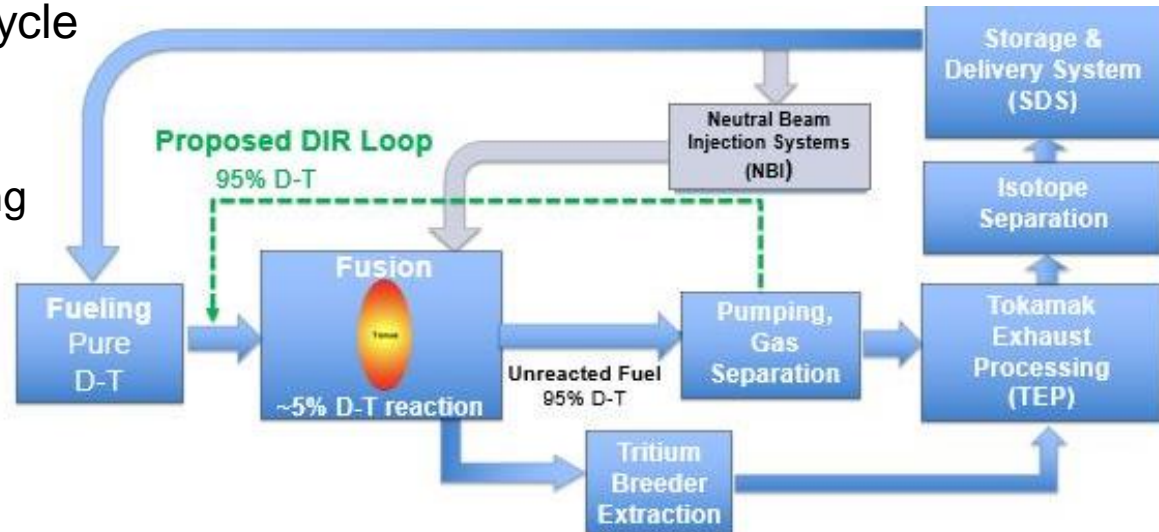
Fuel cycle design: Outstanding challenges

- Closing the fuel cycle
 - Minimizing any tritium losses from and holdup within the system
 - Cost of tritium (\$30,000/gm US – € 27,800)
 - Scarcity
 - Safety concerns
 - Improving process definition & increasing technical readiness level (TRL)
 - Low plasma burnup = high available recycle fraction
- Tritium inventory management, operations with Q₂ (H/D/T)
- Tritium breeding – *highly needed!!!!*

More detail is needed for an eventual FPP/DEMO design, and overall tritium inventory is no small factor!

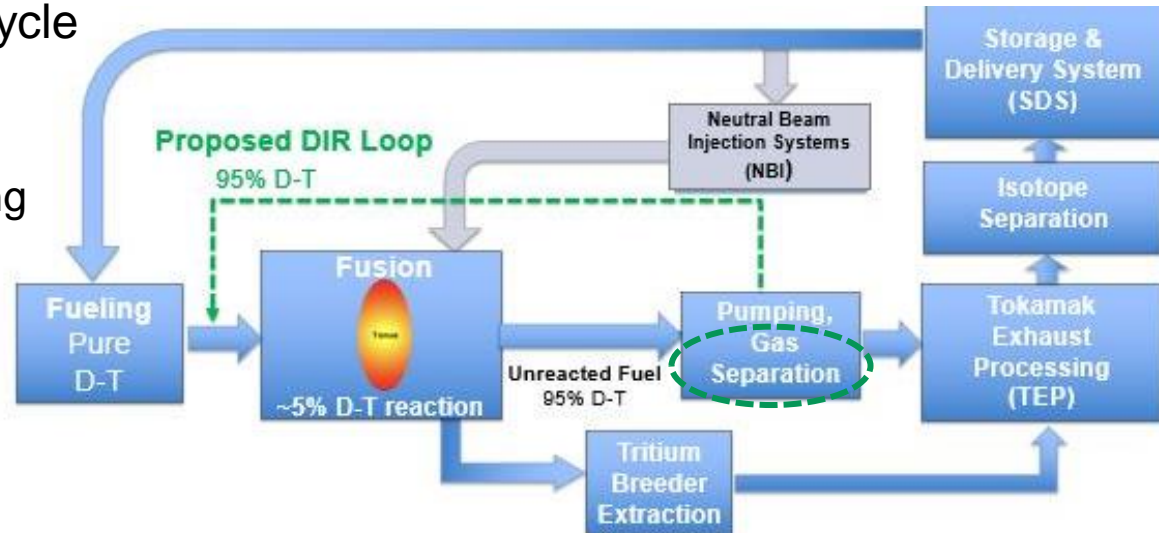
Direct Internal Recycle Concept (DIR)

- Minimizing processing steps to reintroduce DT exhaust to fueling
- Reduces Q_2 inventory in steps like isotope separation, storage
 - Higher throughput = higher fraction of total inventory in use
 - Could reduce size & complexity of larger, more expensive systems such as ISS
- Select highest-volume, highest- Q_2 streams for immediate recycle
- Some proposed recycle concepts exhibit isotope selectivity
 - Increases processing time



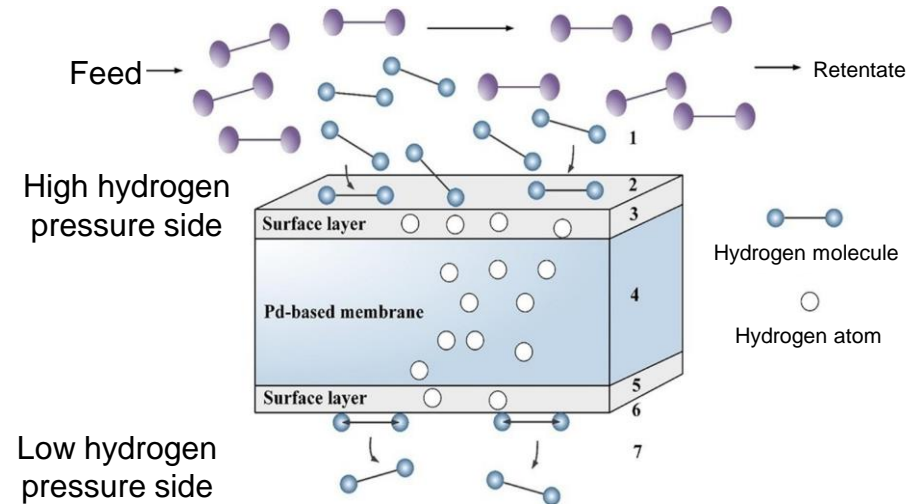
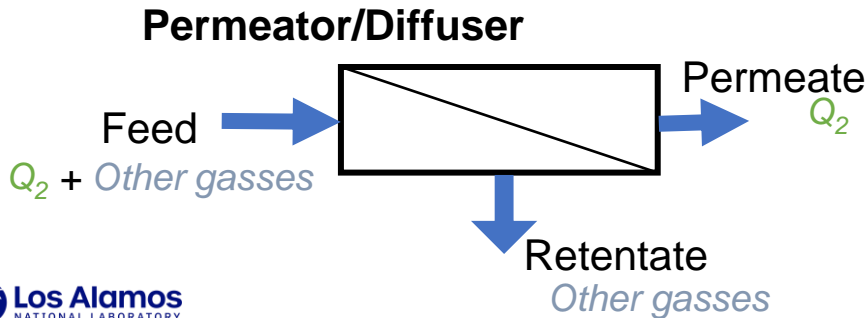
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Permeation Through Q_2 Specific Membranes

- Permeation can accomplish the major function of DIR: hydrogen isotope separation
 - Removes non- Q_2 gasses, leaving pure Q_2 stream
 - Doping may be needed for 50:50 DT for fueling
 - High TRL at large scale
 - Commercially available
 - Fast response time
 - Continuous process
 - Scaleup: high throughputs possible

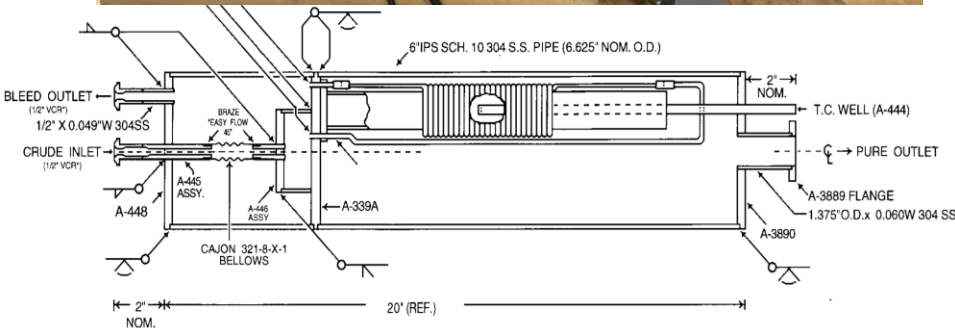


<https://doi.org/10.1016/j.cep.2017.07.021>

LANL permeator & experiment design



- **RSI model RS-3870 Permeator**
- PdAg membrane (0.47 m²)
- Operated at 350 °C
- Testing between 15-95% H₂ in Argon
 - Broad range of burnup fractions
 - Permeation of remaining hydrogen for direct recycle to fueling
- Tested with tritium-compatible pumps
 - Metal Bellows
 - All-Metal Scroll (Normetex)
- Data generated to validate permeator models for DIR calculations

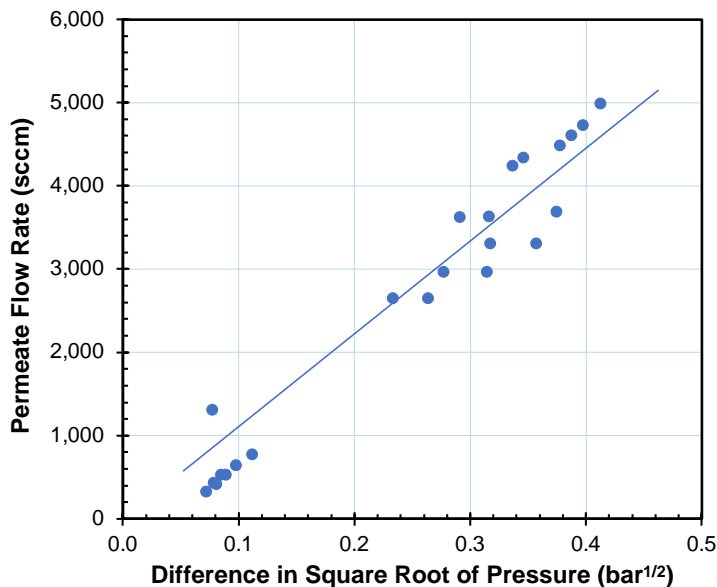


Our modeling approach

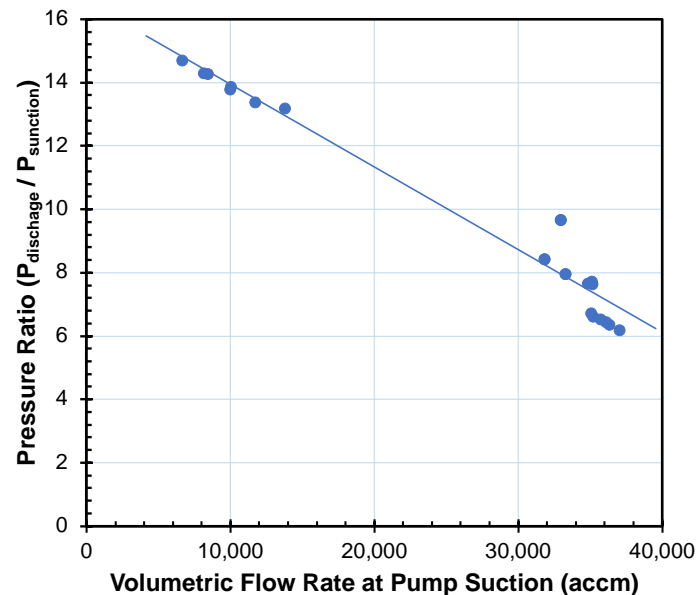
- **Goal Unit operations simulation for fuel cycle design**
 - Physics-based chemical engineering models of each operation
 - Generate flexible software tools for user-specific design
 - Generate models that are readily accessible to businesses and universities
 - HPL laboratory capability/permeators able to test/validate/refine models
- **Permeator modeling**
 - Emphasizes on steady-state model permeator design
 - Explicit treatment of hydrogen isotopes and disproportionation reactions
 - Includes retentate pressure drop and heat transfer
 - Define the relationship between permeator geometry & performance
 - Coupling permeator & pump models
 - Predicts pump train performance from individual pump models
 - Predicts permeate pressures based on pump models
 - Using CAPE-Open standard for inter-platform compatibility

Data Summary

Permeate Flow Rate as a Function of the Difference in Square-Root of Hydrogen Partial Pressures

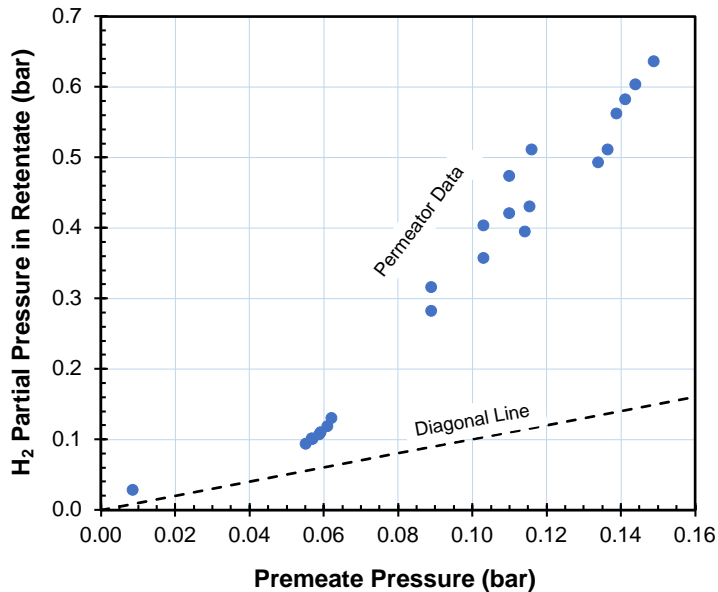


Experimental Pump Curve for MB-601 Metal Bellows Pump

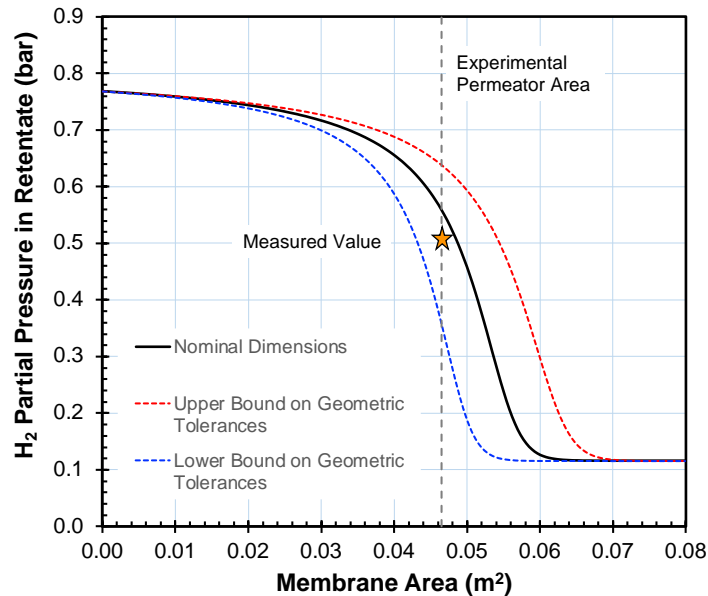


Experiments Conducted Under Breakthrough Conditions

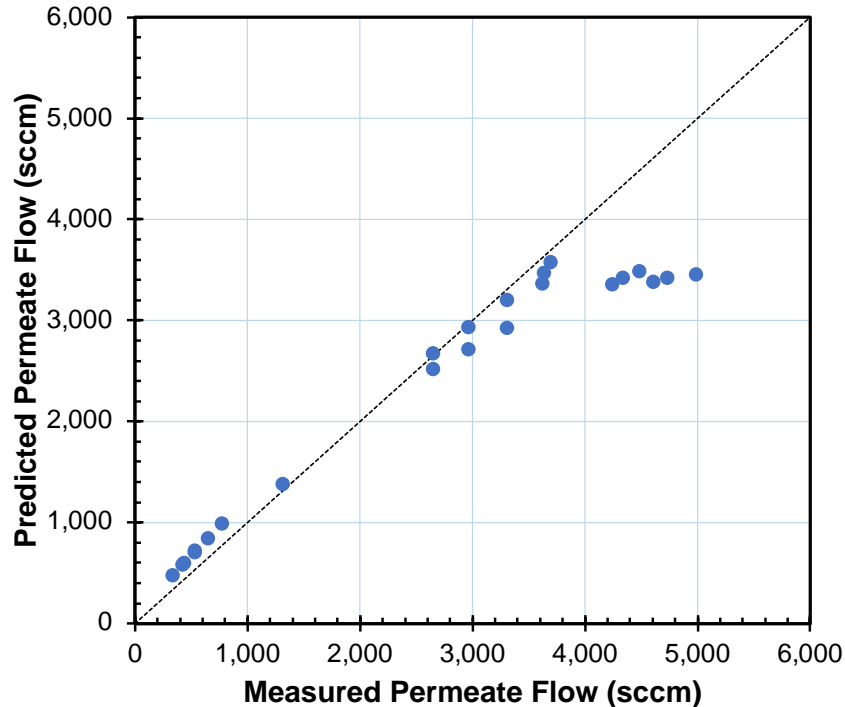
Hydrogen Partial Pressure in the Retentate as a Function of Permeate Pressure



A Sample Comparison of Experimental Data to Model Predictions



Comparison of Model Prediction to Experimental Results

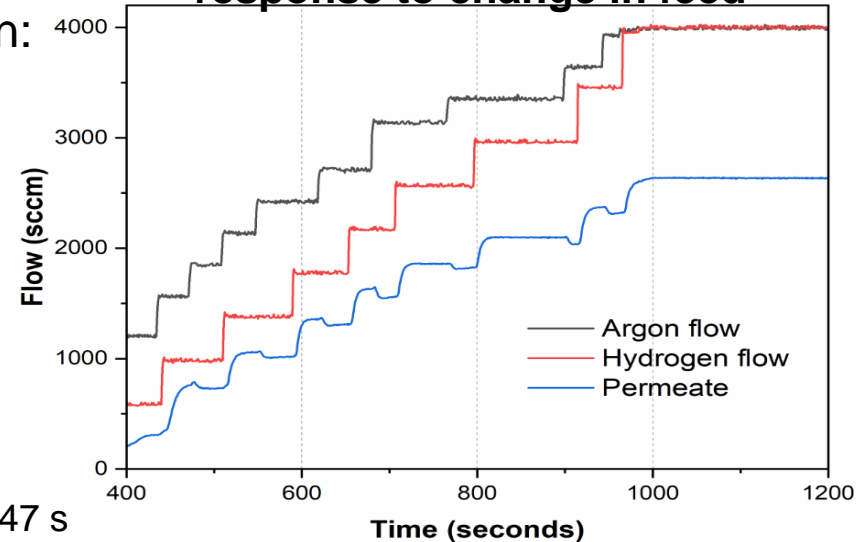


- Data obtained with flows greater than breakthrough flow most useful for evaluating models
 - Flows less than the are insensitive to permeability
 - Excessively large flow rates do not test ability of model to predict breakthrough
- Model predicts permeate flow rate for over 70% of the data
 - Still need to resolve differences for high permeate flow data

Dynamic behavior

- Observed permeate dynamics depend on:
 - Lag between flow controllers and permeator
 - Volume of the permeator inlet plenum
 - Dynamics of the membrane
 - Pump-down time of permeate plenum
- Estimated time response for tests
 - Permeator feed lag: 0.5 – 0.9 s
 - Inlet plenum response: 2.5 – 5 s
 - Permeate plenum pump down: 7 – 41 s
 - Overall response less membrane response: 11 – 47 s
- Membrane response on the order of a few seconds
 - Membrane response time increase with square of thickness
 - Increasing membrane unlikely to impact permeator response time

Change in permeation in response to change in feed

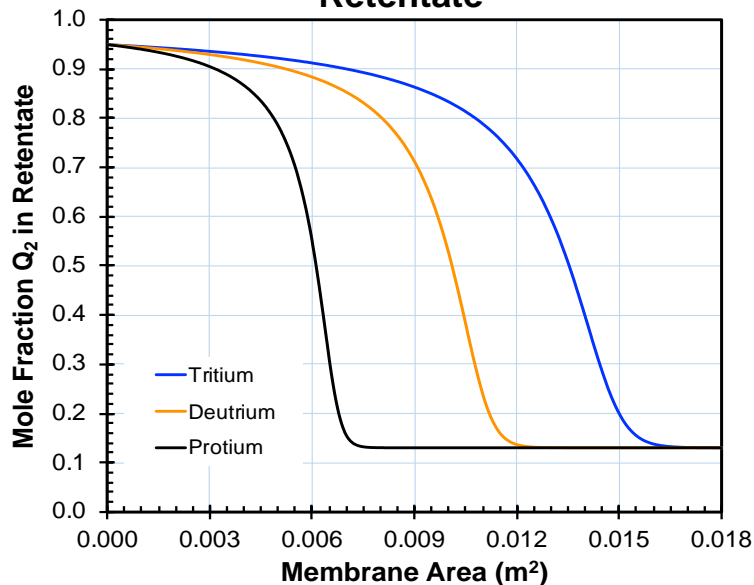


$$\tau = \frac{D_Q \times t}{x^2}$$

τ – time constant
 D_Q – diffusion coefficient
 x – membrane thickness
 t – time

Validity of Experiments with Protium

Typical Q₂ Concentration Profiles in the Retentate



Dimensionless Permeator Equation

$$\frac{d\Pi}{dx} = -\Gamma \cdot (\sqrt{\Pi} - \sqrt{\Pi_{perm}}) \cdot (1 - \Pi)$$

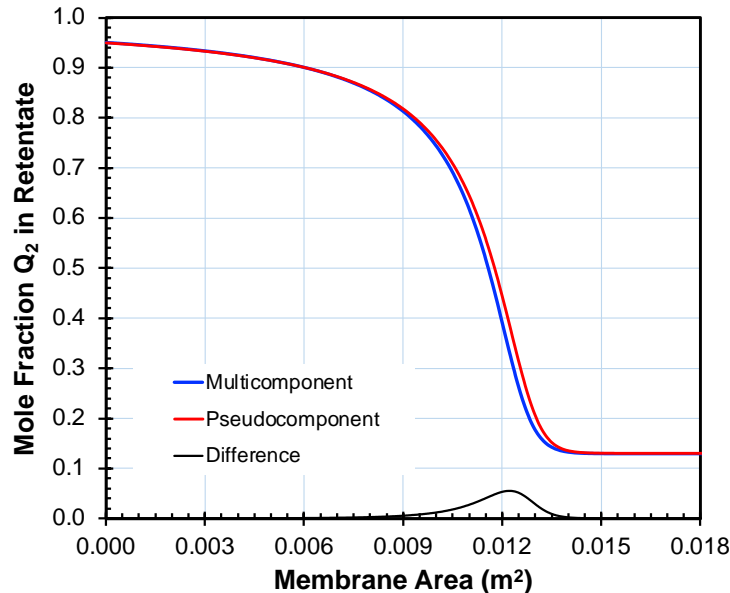
Dimensionless Parameters

- Q₂ partial pressure $\Pi = \frac{P_{Q_2}}{P_{ret}}$
- Membrane area $x = \frac{A}{A_{total}}$
- Permeability $\Gamma = \frac{k_p \cdot A_{total} \cdot \sqrt{P_{ret}}}{F_{inert} \cdot \delta}$
- Permeate pressure $\Pi_{perm} = \frac{P_{perm}}{P_{ret}}$

- **Smaller membrane area required for experiments with H₂**
- **H₂ representative D₂ and T₂ if dimensionless permeability is the same**

Applicability of Protium Experiments to Isotopic Mixtures

Q₂ Concentration Profiles in the Retentate for a 50/50 Deuterium Tritium Mixture



- Assuming disproportionation reactions are in equilibrium, mixture permeability can be estimated using a linear mixing rule

$$k_{p,mix} = k_{p,H} \cdot X_H + k_{p,D} \cdot X_D + k_{p,T} \cdot X_T$$

- A model representing a D-T mixture as a pseudo-component closely approximates an exact model
- Permeator experiments with protium are applicable to isotopic mixtures

Conclusions & Future work

- Permeation may be a promising solution for DIR if isotope resolution is not fully necessary
 - Higher TRL than alternatives
- Models generated for permeators predict performance for 70% of dataset
- A thorough understanding of operating conditions needed will give the most efficient, specific design
 - *True for all unit ops AND whole-plant design*
- Future work:
 - Goal: implement unit operations in system models to discuss broader impacts
 - Isotope effects: further investigation & validation
 - Change in permeate composition because of change in feed composition
 - Opportunity: enhanced membrane design

Acknowledgements



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Thank you!

How to reach out: hpl@lanl.gov

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LANL contributions to the Fusion
Fuel Cycle



Hydrogen storage in
uranium beds



Alternatives to Normetex:
Airsquared pump test report



The Fusion Fuel Cycle

- Major challenges for tritium use
 - Retention/permeation through materials at high temperature [DOE tritium standard]
 - β -radiation & decay
 - Safe storage & accountancy of large quantities of gas
 - Isotope exchange with protium (water, hydrocarbons)
 - $\text{H}_2\text{O} + \text{T}_2 \rightarrow \text{HTO}$
- Tritium handling best practices:
 - Pressure cascade
 - Secondary containment
 - Gloveboxes
 - Double-walled piping
 - Isolatable volume limits
 - Minimizing processing temp

