

Tritium Permeation and Extraction Issues for the ARIES-CS

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Presentation Overview

- Present tritium inventory and permeation safety limits
- Discuss the factors that impact tritium inventories and permeation in ARIES-CS
- Describe three tritium extraction methods that are being proposed for PbLi systems and present the performance of the most applicable system for Dual Cooled Lead Lithium (DCLL) blankets
- Present preliminary inventory and permeation results of ARIES-CS
- Conclude with a summary

Tritium Inventory and Permeation Safety Limits

- The DOE site boundary dose limit during accidents is 10 mSv; because tritium will not be the only radioactive material released during accidents it is assumed that half of the dose will be due to tritium, which translates into an allowable tritium release of ~140 g-T as HTO if the release is stacked, or ~15 g-T as HTO if release is at ground level
- The factor of 10 between ground versus stacked release limits illustrates the need to maintain confinement building integrity during helium pressurization events so that all releases can be stacked
- As a comparison, ITER's in-vessel mobile tritium inventory is 450 g, most of which ends up in ITER's pressure suppression system during accidents
- Total facility airborne operation release to the environment is limited to 1 g-T/a as HTO, or ~30 Ci/d; in-facility releases can be higher if an air detritiation system is available
- The allowed public dose from routine release of radionuclides into community drinking water is 0.04 mSv/year (40 CFR 141.66), which translates into a tritium concentration of 20,000 pCi/l for drinking water

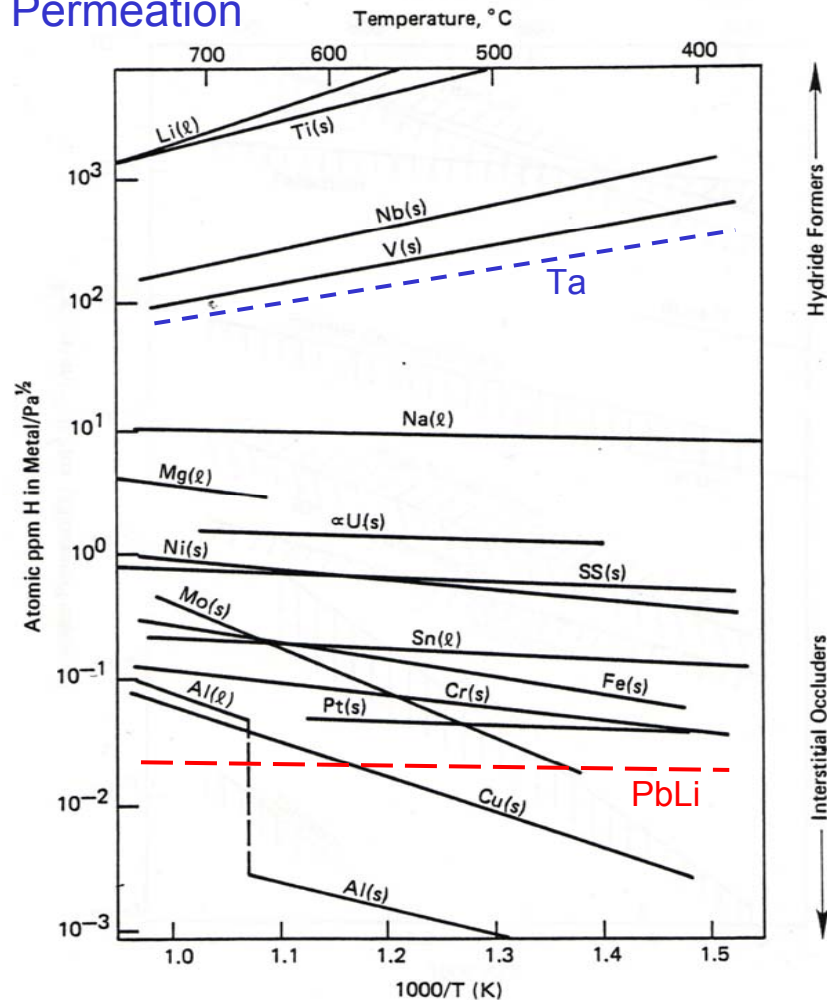
Factors That Impact Tritium Inventories and Permeation

- Material compatibility dictates material choice and the selected material determines tritium inventories and permeation rates
 - Clever design approaches have allowed the use of reduced activation ferritic steel (RAFS) as blanket and piping structural material ($T < 550^{\circ}\text{C}$)
 - RAFS is not an effective tritium barrier at the adopted ARIES-CS operating temperature range ($400^{\circ}\text{C} < T < 550^{\circ}\text{C}$)
 - However, the high PbLi blanket outlet temperature ($T = 700^{\circ}\text{C}$) requires that the heat exchanger (HX) structural material be a refractory metal alloy (Nb, Ta, W) or SiC
 - Niobium and tantalum getter tritium, requiring exceptionally low PbLi tritium concentrations to avoid high tritium inventories
- The breeding material processing rate needed to minimize system tritium inventory and permeation rate, along with breeder temperature, will determine the tritium extraction method needed for ARIES-CS
 - The International Fusion Community has been investigating three methods for extracting tritium from PbLi: bubble columns, permeators, and alkaline metal heat exchanger (HX) traps

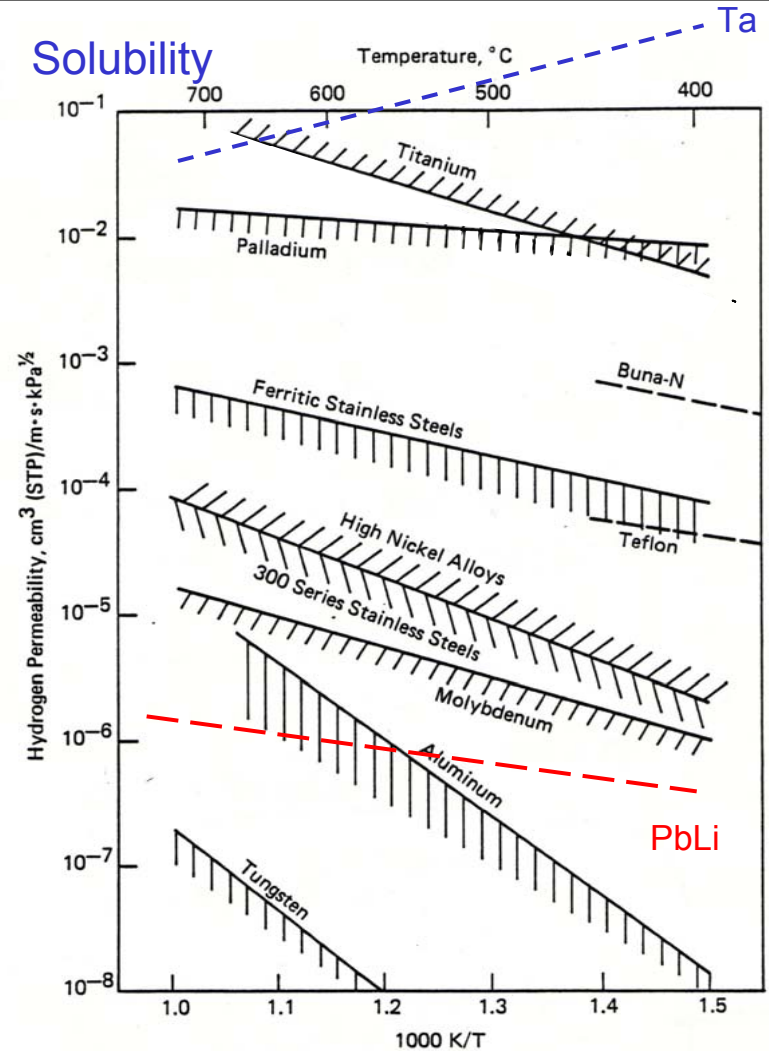
Tritium Behavior in Metals

(Nb is ~3 times higher than Ta)

Permeation



Solubility



Bottom line: Tritium is slow to diffuse through PbLi, but once it reaches a PbLi/gas or PbLi/metal surface, then it would rather reside in almost any material other than PbLi (especially in Nb or Ta)

Heat Exchanger Tritium Inventory*

PbLi T2 Pressure (Pa)	Niobium Tubes			Tantalum Tubes		
	Reactor (g)	Two Field Periods Four cooling loops (g/loop)	Three Field Periods Six cooling loops (g/loop)	Reactor (g)	Two Field Periods Four cooling loops (g/loop)	Three Field Periods Six cooling loops (g/loop)
1.0	2660	665	443	1010	253	168
0.45	1785	446	298	678	170	113
0.2	1190	298	198	450	113	75
.16	1065	266	177	403	101	67
.1	840	210	140	321	80	54
.07	703	176	117	269	67	45
.05	595	149	99	225	56	38
.04	532	133	89	200	50	33

Inventory based on heat exchanger surface area of 20,000 m², tube wall thickness of 1 mm, and temperature averaged solubility. Color scheme: **red** - larger inventory than ITER, **light blue** - less than ITER but greater than allowable stacked release, **green** - less than allowable stacked release but greater than allowable ground release

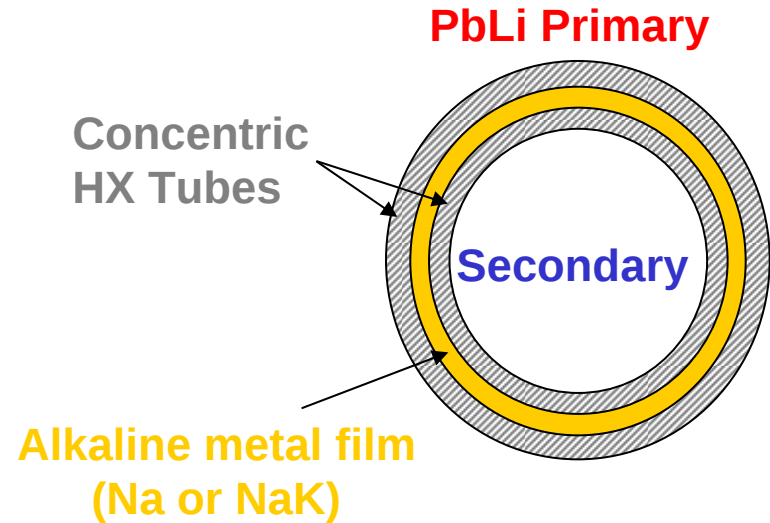
Extraction System PbLi Processing Flow Rates

- Lead lithium flow rates in the different blanket systems (at 2000 MW):
 - Helium Cooled Lead Lithium (HCLL) blankets = 300 kg/s ($3.4 \times 10^{-2} \text{ m}^3/\text{s}$)
 - Dual Coolant Lead Lithium (DCLL) blankets = 28000 kg/s ($3.2 \text{ m}^3/\text{s}$)
 - Self Cooled Lead Lithium (SCLL) blankets = 44000 kg/s ($5.0 \text{ m}^3/\text{s}$)
- For a fusion power of 2000 MW, the tritium production rate is $\sim 3.9 \text{ mg/s}$; as a result the PbLi tritium concentration after a single coolant pass is:
 - HCLL blankets = $2.3 \times 10^{22} \text{ T/m}^3$ (1468 Pa)
 - DCLL blankets = $2.5 \times 10^{20} \text{ T/m}^3$ (0.17 Pa)
 - SCLL blankets = $1.6 \times 10^{20} \text{ T/m}^3$ (0.07 Pa)
- The problem is that the PbLi in DCLL blankets can achieve tritium concentrations in a single pass through the blanket that result in exceeding tritium inventory limits for Nb tube HXs; this means that the total PbLi outlet stream must be processed prior to flowing into the HXs.

An Alkaline Metal Heat Exchanger Trap may not a Good Extraction Method for DCLL Blankets (Option 3)

Concept: Tritium permeating concentric HX tubes is chemically trapped by an alkaline metal film, forming NaT. The alkaline metal is subsequently processed batch-wise by introducing the metal to a “cold trap”.

Alkaline Metal Heat Exchanger Trap



- This concept has primarily been considered for RANKINE power cycles when the primary temperature is $< 500^{\circ}\text{C}$
- Application to DCLL would require Nb HX tubes and there is some concern that the HX tritium inventory would be too high, however some additional analyses would be required to confirm this concern

Membrane Loop for a Gas Bubble Extraction Column (Option 2)

(Option 2)

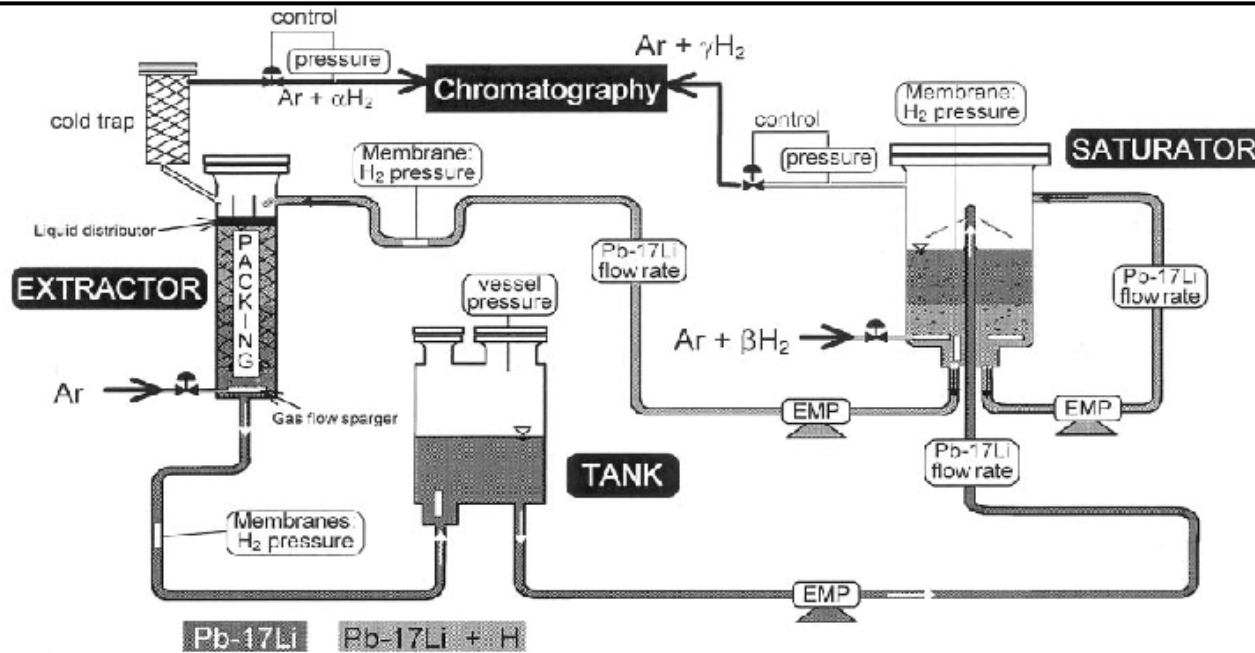


Fig. 1. Melodie loop (extractor in an immersed hydraulic configuration).

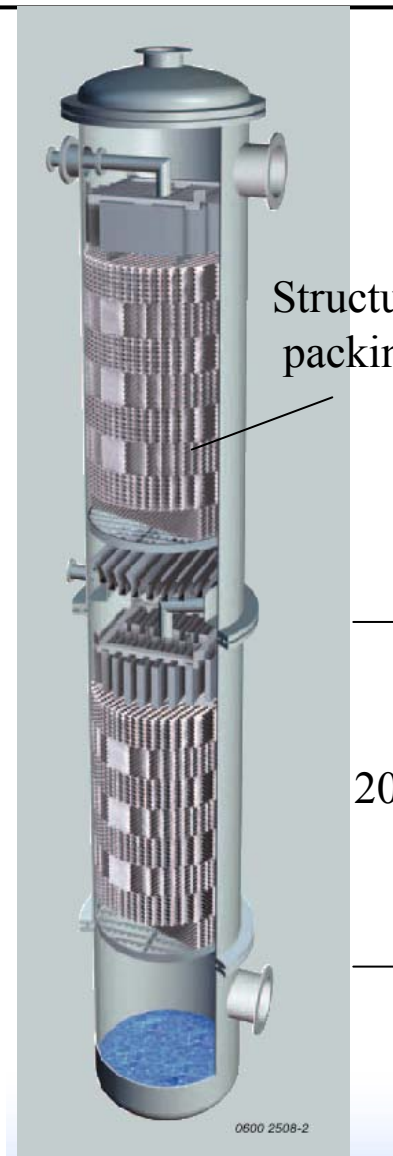
Table 1

Extraction tests in a non-immersed hydraulic configuration
($T = 673$ K)

Test No.	L (l h ⁻¹)	G (Ncm ³ min ⁻¹)	$P_{H_2,IE}$ (Pa)	η (%)
1	70–90	100	1200–1350	20–22
2	30–50	100	1000–1100	29–31
3	30–50	500	975–1000	29–31
4	30–50	100	450–475	23–25
5	30–50	100	220–230	23–25

$$\eta = (\sqrt{P_{H_2,IE}} - \sqrt{P_{H_2,OE}}) / \sqrt{P_{H_2,IE}}$$

N. Alpy, et al., FED, 49-50
(2000) 775-780.

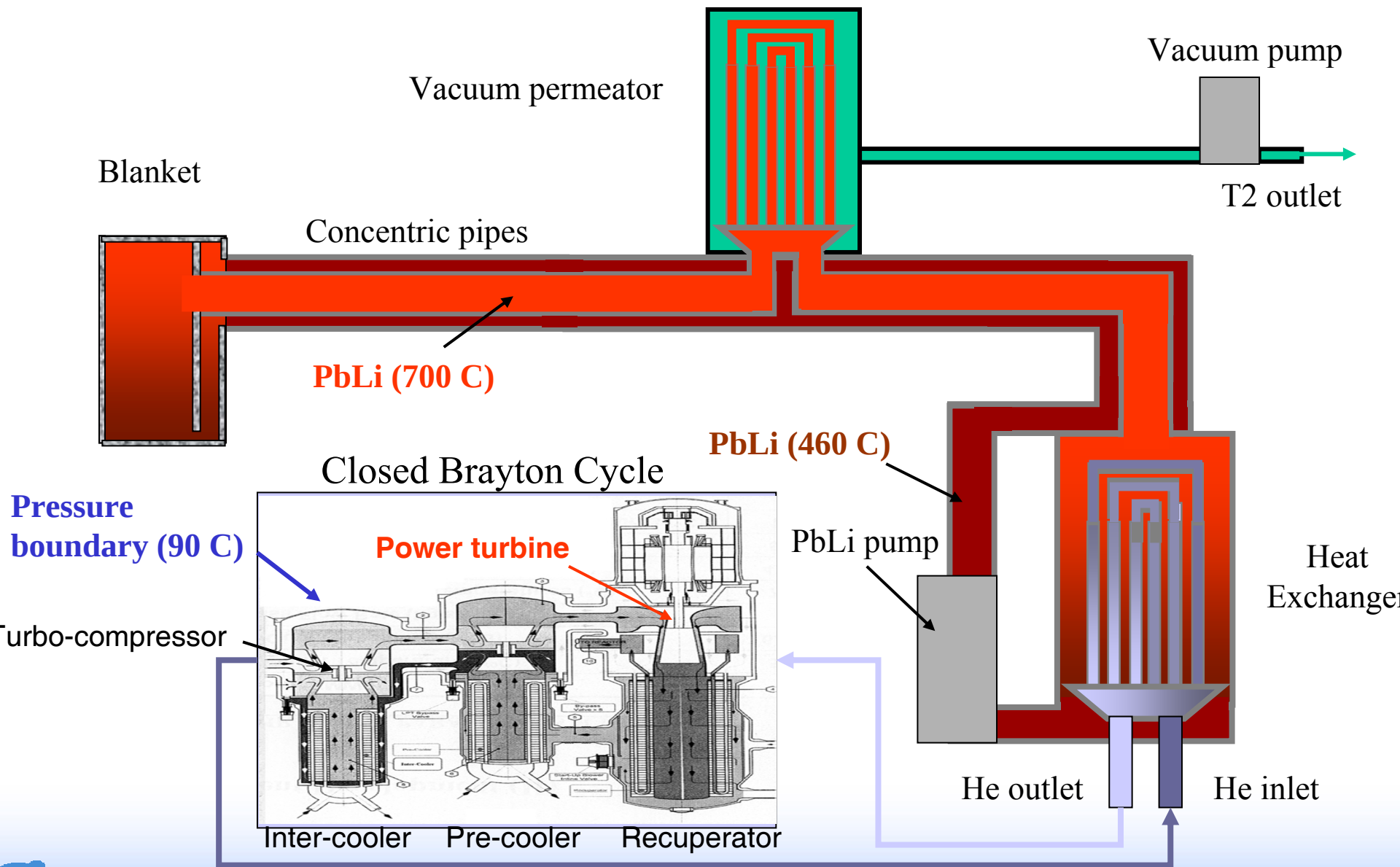


Sulzer Column

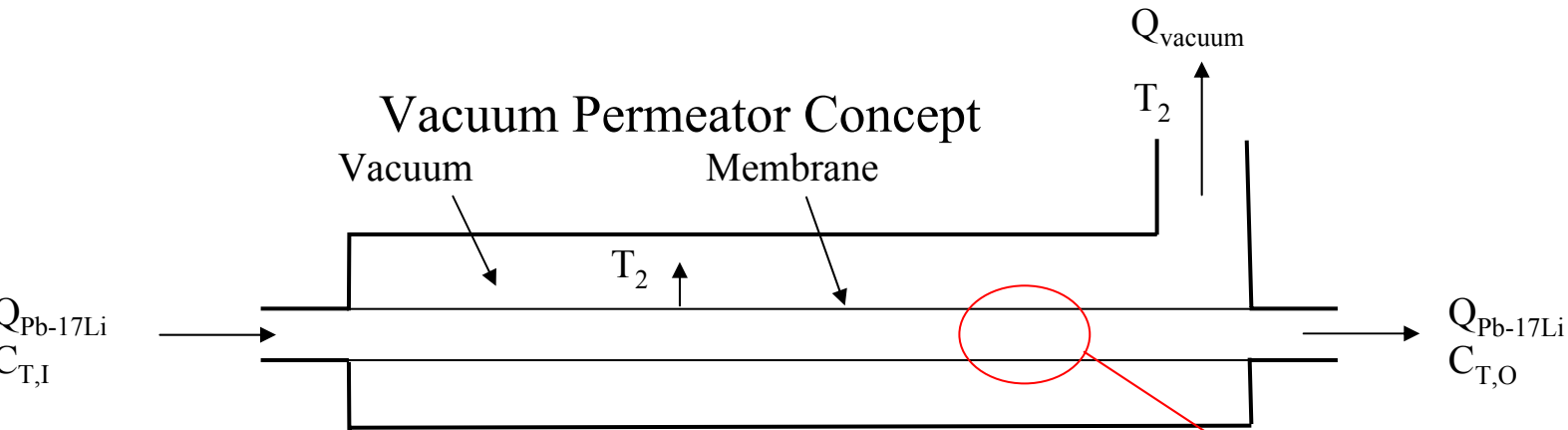
Extraction Columns are not a Good Option for DCLL Blankets

- Because the DCLL blanket concept requires an effective extraction system in the PbLi outlet steam where the temperature is 700°C, the columns will have to be constructed of either tungsten or SiC to deal with this high temperature and to reduce any tritium inventory buildup in the column packing material.
- Since the entire coolant stream must be processed each pass, the number of extraction columns needed is ~240,000 at a column flow rate of 50 l/hr (columns need to be 2.4 m long to achieve required cleanup efficiency of 60 %).
- Because these columns operate at pressures less than that of the DCLL PbLi coolant, the entire primary coolant stream will have to be de-pressurized and re-pressurized per coolant cycle.
- There is some concern that bubble columns will not be as efficient at tritium partial pressures below 100 Pa.
- In order to make extraction columns work for the DCLL blanket, permeation barriers would have to be placed inside the blanket to keep tritium out of the helium cooling stream and the HXs would have to be made of W, however:
 - There is a concern in the materials community that permeation barriers are ineffective in a radiation environment
 - Fabricating HXs from W seems problematic in comparison to using Nb
- This is why extraction columns are better suited for HCLL blankets

Primary Side PbLi Vacuum Permeator (Option 1)



Permeator Tritium Transport Overview



Scott Willms of the Los Alamos National Laboratory (LANL) examined 10 mass transport correlations for ITER TBM design work, and proposed adopting the following correlation:

$$\frac{K_m D_{\text{tube}}}{D_{T,\text{Pb-17Li}}} = 0.0096 \text{Re}^{0.913} \text{Sc}^{0.346}$$

-- Harriot and Hamilton, Chem Engr Sci, 20 (1965) 1073

Pb-17Li mass transport

$$\Gamma_T = K_m (C_{T,\text{Bulk}} - C_{T,S1})$$

$C_{T,\text{Bulk}}$

$Q_{\text{Pb-17Li}}$

$C_{T,S1}$

Membrane diffusion

$$\Gamma_T = -D_T \frac{\partial C_T}{\partial x}$$

$C_{T,S2}$

$C_{T,S3} = K_s \sqrt{P}$

Or $C_{T,S3}$ based on molecular recombination

$$\Gamma_{T2} = \alpha_r C_{T,S3}^2$$

$$\frac{C_{T,S2}}{C_{T,S1}} = \frac{K_{S,\text{Nb}}}{K_{S,\text{Pb-17Li}}}$$

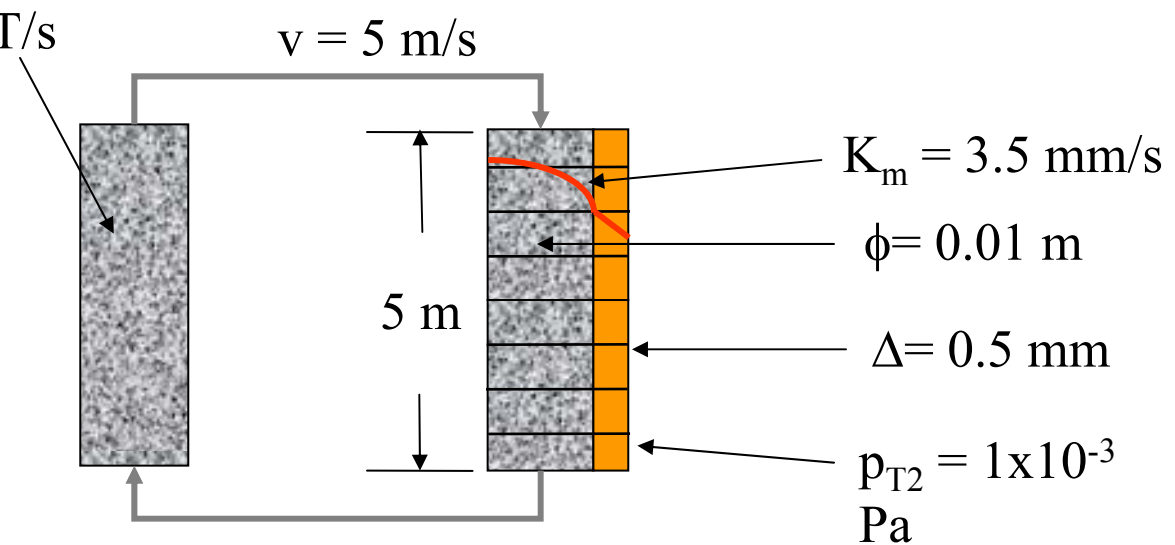
Development Issues Associated With Vacuum Permeators

- Permeators using a Pd-Ag alloy membrane have been developed by LANL for gaseous applications; but permeators for liquid metal systems have not been developed
- Measurements of tritium mass transport coefficients in PbLi for turbulent flow in tubes have not be made. This is a key parameter in assessing the viability of this option since the major resistance to extraction of the tritium is permeation of tritium through the PbLi.
- Material compatibility (corrosion) measurements have not be made for PbLi and Nb, although, in general, refractory materials are thought to be compatible with PbLi based on tests up to 1000°C, like the PbBi-W&Mo test conducted in the LECOR loop*
- At high temperatures Nb will rapidly oxidize, requiring a very high vacuum during operation or a surface layer of Pd which is more oxide resistant. This issue is a very serious recovery concern after loss-of-vacuum accidents and may require housing the permeator in an inert gas environment.

*C. Fazio, J. Nucl. Mater., 318 (2003) 325-332

Tritium Migration Analysis Program (TMAP) Permeator Model

$\Gamma_T = 5 \times 10^{-7} \text{ g-T/s}$
 7,690 tubes are required to process
 entire reactor primary flow giving
 3.46 kg/s per tube (5 m/s)
 Model is based on a single tube –
 tritium production is 3.9 mg-T/s
 divided by 7,690 tubes, or $5 \times 10^{-7} \text{ g-T/s-tube}$
 PbLi pressure drop $\sim 1.5 \text{ atm/m}$
 tube length



Mass transport

$$\frac{D_{\text{tube}}}{\mu_{\text{Pb-17Li}}} = 0.0096 \text{Re}^{0.913} \text{Sc}^{0.346} \Rightarrow K_m = 3.46 \frac{\text{mm}}{\text{s}}$$

$$\mu = \frac{\mu}{\rho D}; \text{Re} = \frac{\rho L v}{\mu}$$

$$7.4 \times 10^{-4} \frac{\text{kg}}{\text{m-s}}; c_p = 186 \frac{\text{J}}{\text{kg-K}}$$

Carrier and Hamilton, Chem Engr Sci, 20
 (1955) 1073

PbLi properties

$$^* K_s = 2.32 \times 10^{-8} n_{\text{PbLi}} \exp(-1350/RT) \frac{T}{\text{m}^3 - \text{Pa}^{1/2}}$$

$$^{**} D = 2.5 \times 10^{-7} \exp(-27000/RT) \frac{\text{m}^2}{\text{s}}$$

$$n_{\text{PbLi}} = \rho_{\text{PbLi}} \frac{A}{M} = 3.07 \times 10^{28} \frac{\text{a}}{\text{m}^3}$$

$$R = 8.31 \frac{\text{J}}{\text{mol-K}}; \rho_{\text{PbLi}} = 8813 \frac{\text{kg}}{\text{m}^3}; M = 0.173 \frac{\text{kg}}{\text{mol}}$$

^{*}Reiter, FED 14 (1991) 207-211

^{**}Teria, J. Nucl. Mater. 187 (1992) 247-253

Nb properties

$$^! K_s = 2.83 \times 10^{-6} n_{\text{Nb}} \exp(35567/RT) \frac{T}{\text{m}^3 - \text{Pa}^{1/2}}$$

$$^{!!} D = 4.625 \times 10^{-8} \exp(-6233/RT) \sqrt{\frac{1}{3}} \frac{\text{m}^2}{\text{s}}$$

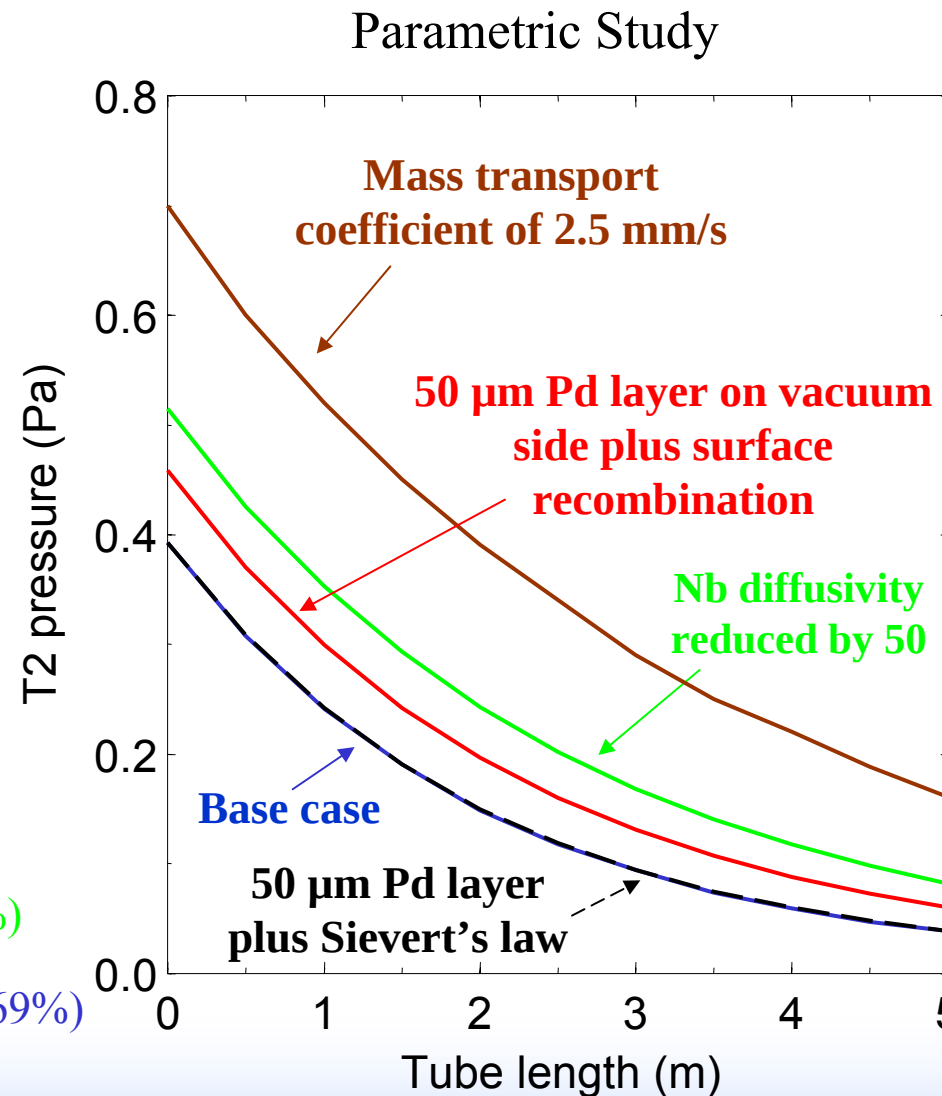
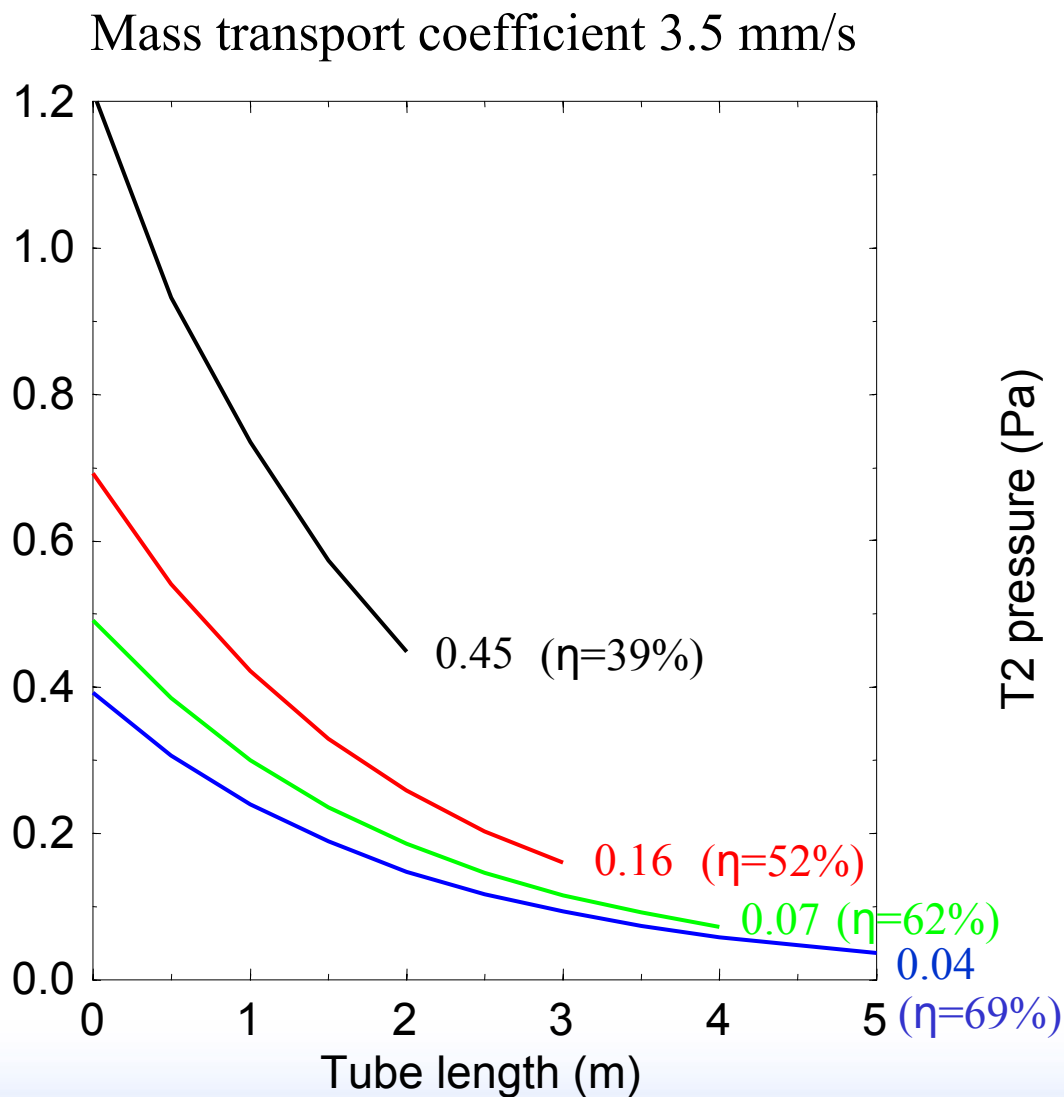
$$n_{\text{Nb}} = \rho_{\text{Nb}} \frac{A}{M} = 5.543 \times 10^{28} \frac{\text{a}}{\text{m}^3}$$

$$\rho_{\text{Nb}} = 8850 \frac{\text{kg}}{\text{m}^3}; M = 0.0929 \frac{\text{kg}}{\text{mol}}$$

[!]Natesan, J. Nucl. Tech., 22 (1973) 1073

^{!!}Mound Report, MLM-3610 (1973)

Tritium Partial Pressure Versus Tube Length



Permeation Safety Concerns

- Sources of in-facility airborne operational release
 - Permeation through outer wall of concentric PbLi primary piping
 - Helium leakage from closed Brayton cycle
 - Permeation through pressure boundary of closed Brayton cycle
- PbLi piping wall permeation is ~ 3 g-T/a based on RAFS pipe walls at 400°C , area of 300 m^2 , thickness of 1 cm, and PbLi tritium pressure of 0.05 Pa
- The Brayton cycle helium leak rate is unknown. However, extrapolating the measured leak rate from Chinese HTR-10 (a helium cooled 10 MWth fission reactor leak tested at 3.9 MPa to be 1.7 cc/s) the estimated leak rate for ARIES-CS six closed Brayton cycles will be ~ 10 cc/s, which for a helium tritium pressure of 0.05 Pa gives ~ 7 mg-T/a
- Permeation from the Brayton cycle pressure vessel will be ~ 0.1 mg-T/a based on a tritium pressure 0.05 Pa, wall area of 3100 m^2 , thickness of 8.5 cm, and a temperature of 90°C
- The total is $\sim$ three times the allowed annual release to the environment, but building air detritiation (reduction of 100) and alumina primary pipe coatings (reduction of 10 to 1000 for $50\text{ }\mu\text{m}$ coating) can cut this release by a factor 1000

Permeation Safety Concerns (cont.)

- The major source of tritium into water is permeation through the inter-cooler pipe walls, but by using an alloy of aluminum for these pipes this permeation will be greatly reduced
- The permeation rate for the inter-coolers operating at 60°C, area of 30,000 m², and tube thickness of 2.5 mm is 0.63 mCi/day
- At this rate, and an assumed total water volume of 500 m³, the allowed community drinking water limit (20,000 pCi/l) is reached in less than 17 days of operation; therefore this water will have to eventually be processed since DOE's policy is retention and not dilution.
- Over the 30 year lifetime of the ARIES-CS this concentration will grow to 7 mCi/l at this rate of permeation
- As a point of comparison, processing of the Pickering CANDU reactor water occurred once the tritium concentration reached 500 mCi/l; therefore processing of ARIES-CS inter-cooler water during operation will probably not be required.
- Helium leak rates into inter-cooler water are also an unknown; however, even at a helium leak rate of 10 cc/s, after 30 years, the tritium concentration increase is only 4.4 mCi/l.

- A vacuum permeator appears to be the correct tritium extraction option for the DCLL blanket.
- For this extraction method, the tritium inventory in the primary heat exchangers can be less than the site boundary limit during accidents, provided the release is stacked
- Permeation rates into the ARIES-CS building and into the inter-cooler water should be acceptable
- It must be stressed that these results depend on the success of the vacuum permeator concept which needs additional R&D
 - Tritium mass transport coefficients in PbLi must be verified
 - PbLi corrosion of Nb must be determined
 - Nb oxidation must be prevented; hopefully a Pd surface layer will eliminate this concern
- However, detailed analyses must be performed to quantify tritium inventories and permeation rates once the ARIES-CS cooling system design evolves.