



**Savannah River
National Laboratory®**

Closing the fuel cycle

2020 Introduction to Fusion Energy and Plasma Physics Course

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Savannah River National Laboratory

Public information from:

2010 Tritium Conference Tutorial Lectures, Nara, Japan , 2010.10.29, five tutorial lectures taught by T. Tanabe (Kyushu university, Japan), M. Glugla (ITER), T. Yamanishi (JAEA), S. Willms (LANL, now with ITER), S. Konishi (Kyoto University, Japan);

Various public presentations with credits to: K. Heroux, G. Staack, M. Morgan, B. Garcia-Diaz, J. Klein, and D. Babineau at SRNL; Th. Giegerich, Chr. Day, R. Knitter, N. Osman and F. Cismondi at KIT; I. Castillo at AECL EACL; C. Forsberg at MIT.

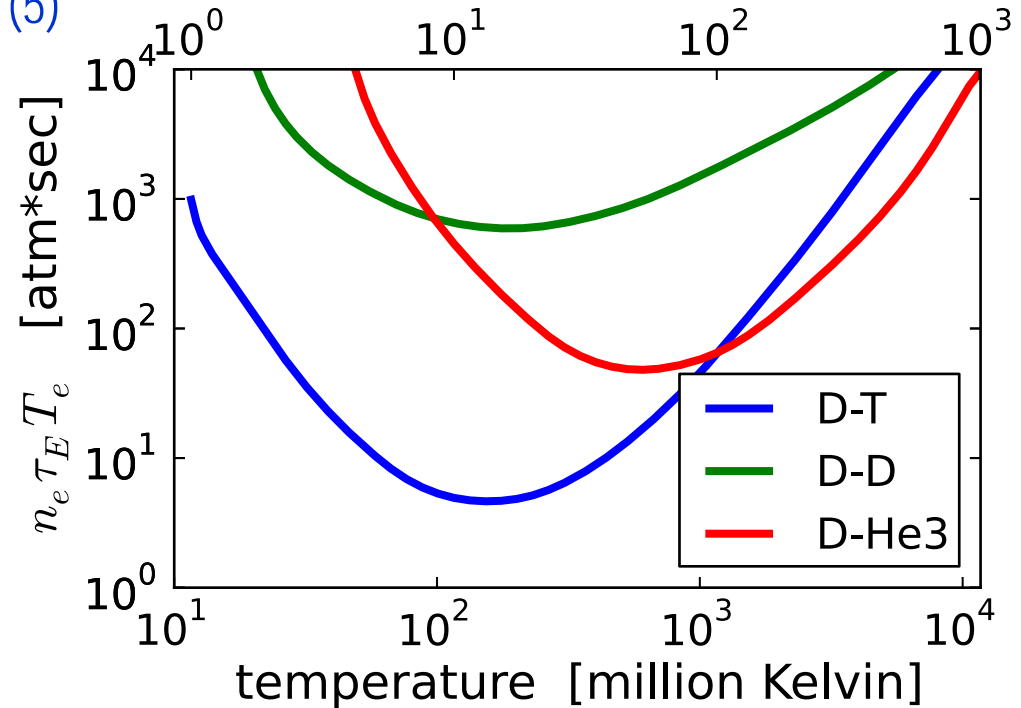
T and D as fuels for fusion reactions



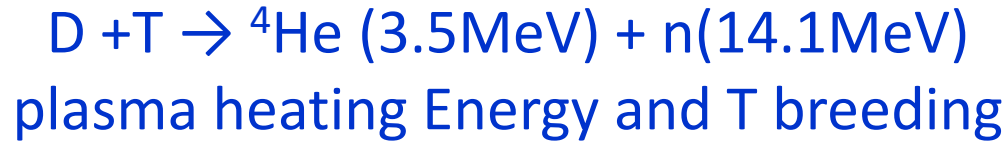
DT fusion (1) is the most suitable fusion reactions

The D + ${}^3\text{He}$ reaction is attractive for no neutron production, though accompanying DD reactions do produce it.

The fusion triple product condition for three fusion reactions.
temperature [keV]



Tritium resource is very limited → need breeding



- Deuterium: Vienna Standard Mean Ocean Water (VSMOW) is 155.76 ppm
- Tritium must be produced or bred internally from lithium
 - 56 kg tritium per GW year (thermal) of fusion power
 - About 100 g tritium produced per year in a standard CANDU fission reactor
 - 20 to 25 kg tritium available (mainly in Canada) for ITER startup
 - Tritium must be bred by reactions in blanket systems



- Overall breeding ratio is expected to be above ~1.1 (not easy to achieve)



Deuterium

Property	D ₂ O (Heavy water)	H ₂ O (Light water)
Freezing point (°C)	3.82	0.0
Boiling point (°C)	101.4	100.0
Density at STP(g/mL)	1.1056	0.9982
Temp. of maximum density (°C)	11.6	4.0
Viscosity (at 20°C, mPa·s)	1.25	1.005
Surface tension (at 25°C, μJ)	7.193	7.197
Heat of melting (cal/mol)	1,515	1,436
Heat of vaporisation (cal/mol)	10,864	10,515

Source: in seawater at a D/H ratio of 156 ppm

Used: in nuclear energy (e.g. D₂O in CANDU reactors)

Production methods (D₂O): e.g. Girdler-Sulfide Process (isotope exchange column) + vacuum distillation

Estimated earth availability: 5×10^{16} kg (in oceans)

Sufficient for several billion years !!

F. Cismondi, Basics of breeding blanket technology, KIT

^6Li Enrichment

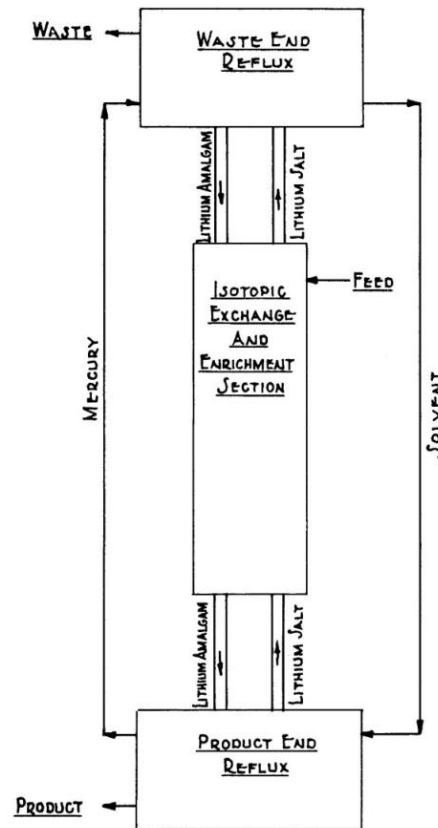
COLEX process

- Counter-current flow of a LiOH solution (OREX: LiCl in PDA) and lithium amalgam, ^6Li accumulates in the amalgam phase
- Production between 1955 and 1963 in the Y12 plant in Oak Ridge, Tennessee.

Other concepts:

- Displacement chromatography
- Ion exchange methods
- Intercalation methods
- Electrolysis
- Electrophoresis
- Electromigration
- Crown ether complex
- Liquid ammonia methods
- Electromagnetic separation
- Laser based separation methods

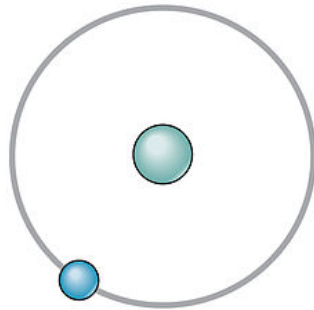
SIMPLIFIED CHEMICAL REFLUX SYSTEM FOR LITHIUM ISOTOPIC OPERATIONS



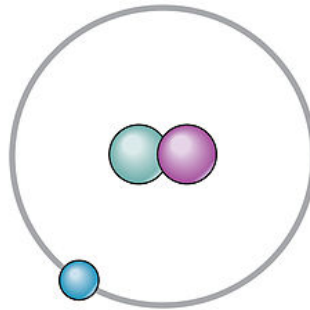
Source: *Separation Science and Technology*, 20 (9-10), 633–651 (1985).

Reference: Th. Giegerich, Chr. Day, R. Knitter, N. Osman, *Lithium enrichment issues in the sustainable supply chain of future fusion reactors*, KIT/ITEP, 05 May 2016

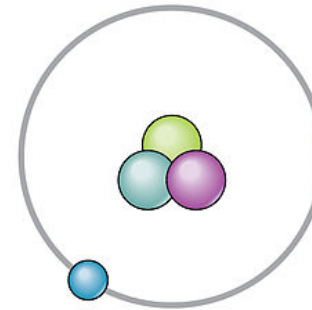
Tritium, one of the three hydrogen isotopes



Protium (^1H)



Deuterium (^2H)



Tritium (^3H)

– Tritium is the radioactive hydrogen isotope

- decay: $^3\text{T} \rightarrow ^3\text{He} + \beta \text{ electron} + \text{antineutrino}$
- 18.6 keV total (average 5.7 keV kinetic, + nearly undetectable antineutrino)
- decay heat: 324 mW/g
- half life: 12.32 years (loss $\sim 5.5\%$ per year)
- isotope mass: 3.0160492 u
- Shielding of tritium radiation is not really a issue (Except direct exposure of organs)
- HTO is $> 10,000$ times hazard than HT gas
- 9,650 Ci/g (3.57×10^{14} Bq/g)
- EPA drinking water standard: < 20 pCi/cc

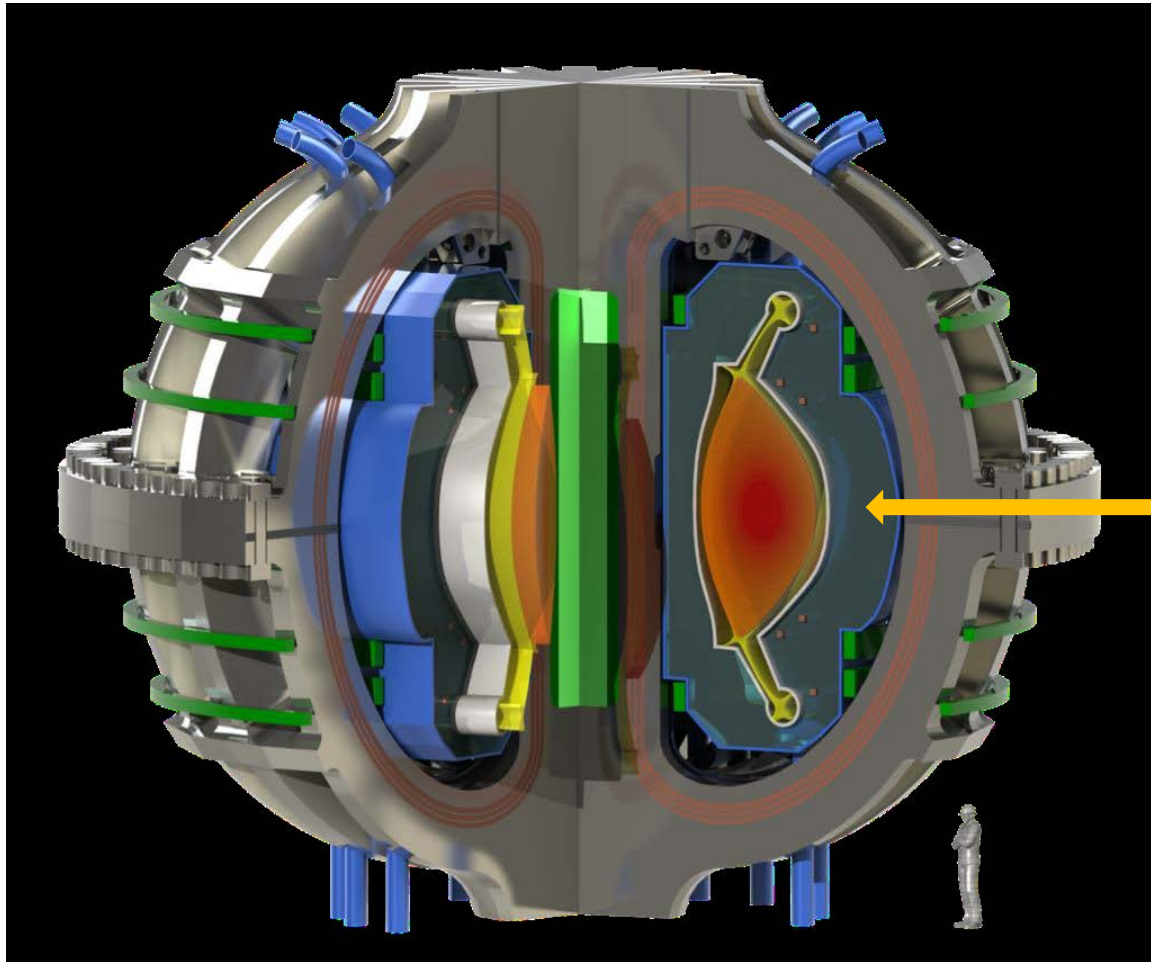
Discovery

- deuterium 1931
- tritium 1934

Most of valves are not compatible with tritium because of polymer packing



Breeding blanket integrated with fusion reactor (example with ARC)



Fusion reactor



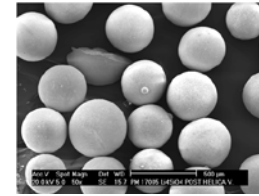
Breeding blanket

Source: Charles Forsberg, Molten Salt Liquid Blanket Integrated Validation Plan, Massachusetts Institute of Technology, Department of Nuclear Science and Engineering, December 6, 2019

Tritium breeding materials

Multiple solid and liquid breed concepts. Parts of these concepts have been tested. No realistic, integrated tests have been performed

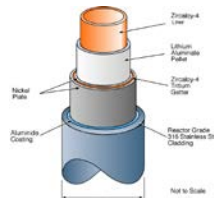
Li_4SiO_4 pebbles (FZK) 0.2- 0.4 mm



Li_2TiO_3 pebbles (CEA) 0.6 – 0.8 mm



LiAlO_2 pellet

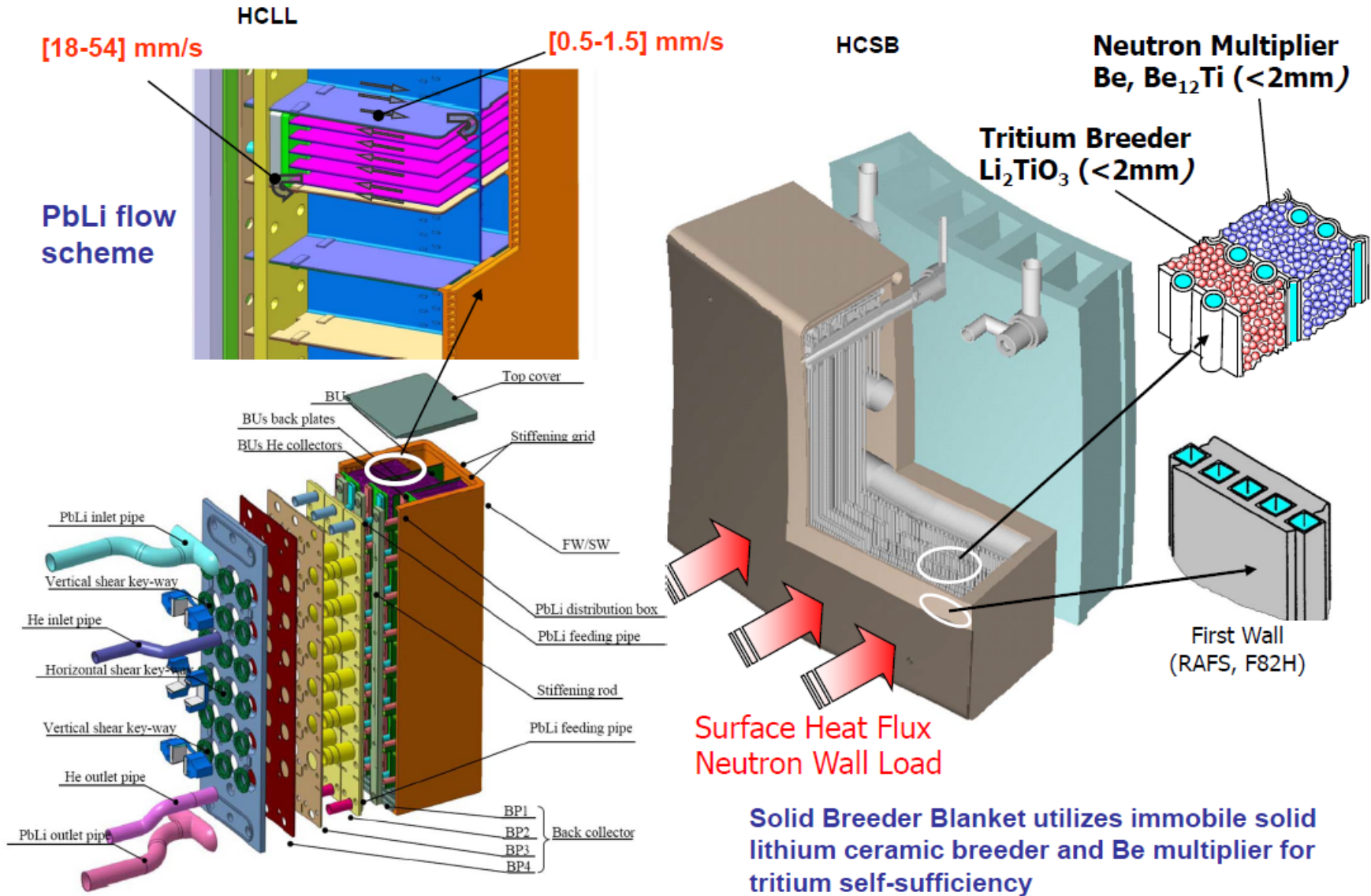


PbLi, work at 300-500°C

Li_2BeF_4 (Flibe), m.p., 459°C



Blanket systems are complex and have many integrated functions, materials, and interfaces



S. Willms, Tritium Science and Technology in the Future, 2010 Tritium Conference Tutorial Lectures, Nara, Japan, 2010.10.29

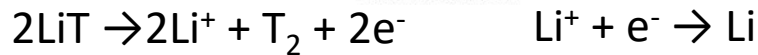
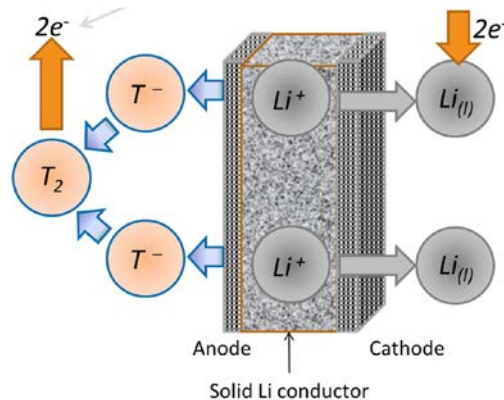
Tritium breeding technical challenges

- Tritium breeding blanket materials and configurations
- Blanket structural materials
- Blanket operations and control
- Blanket maintenance and disposal
- Blanket diagnostics
 - ❖ – Example:
 - For liquid breeding material, what characterizes the flow channel for the coolant blanket application, and how to maintain its function throughout blanket lifetime?
 - For solid breeding material: What radiation resistant properties should the solid breeder pebble have?

Tritium extraction

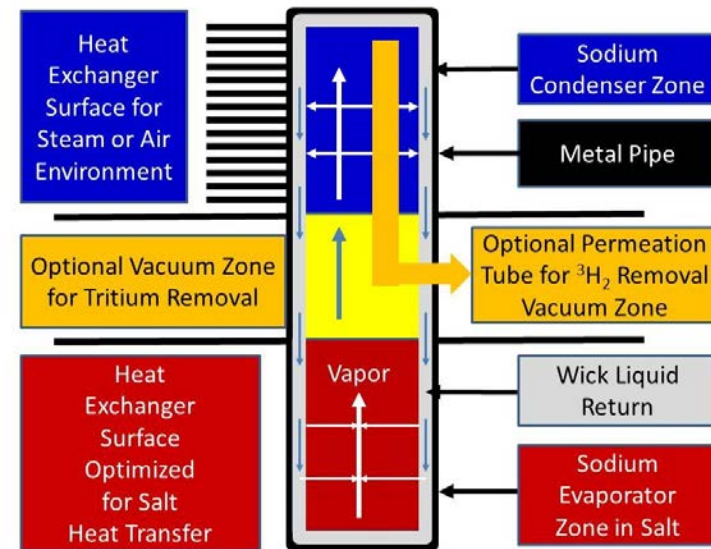
- Tritium extraction from breeding materials
- Tritium extraction from blanket coolants
- Tritium extraction diagnostics
- Blanket systems tritium handling and containment

Electrochemical concept by SRNL



$$E^0_{400^\circ\text{C}} = -2.82 \text{ V}$$

$$E^0_{400^\circ\text{C}} = -3.22 \text{ V}$$



Thermal concept by
ARC (MIT)

Tritium fuel processing

ITER tritium fuel cycle

Fuel cleanup

Isotope separation

Tritium storage and delivery

Water detritiation

Tritium pumping

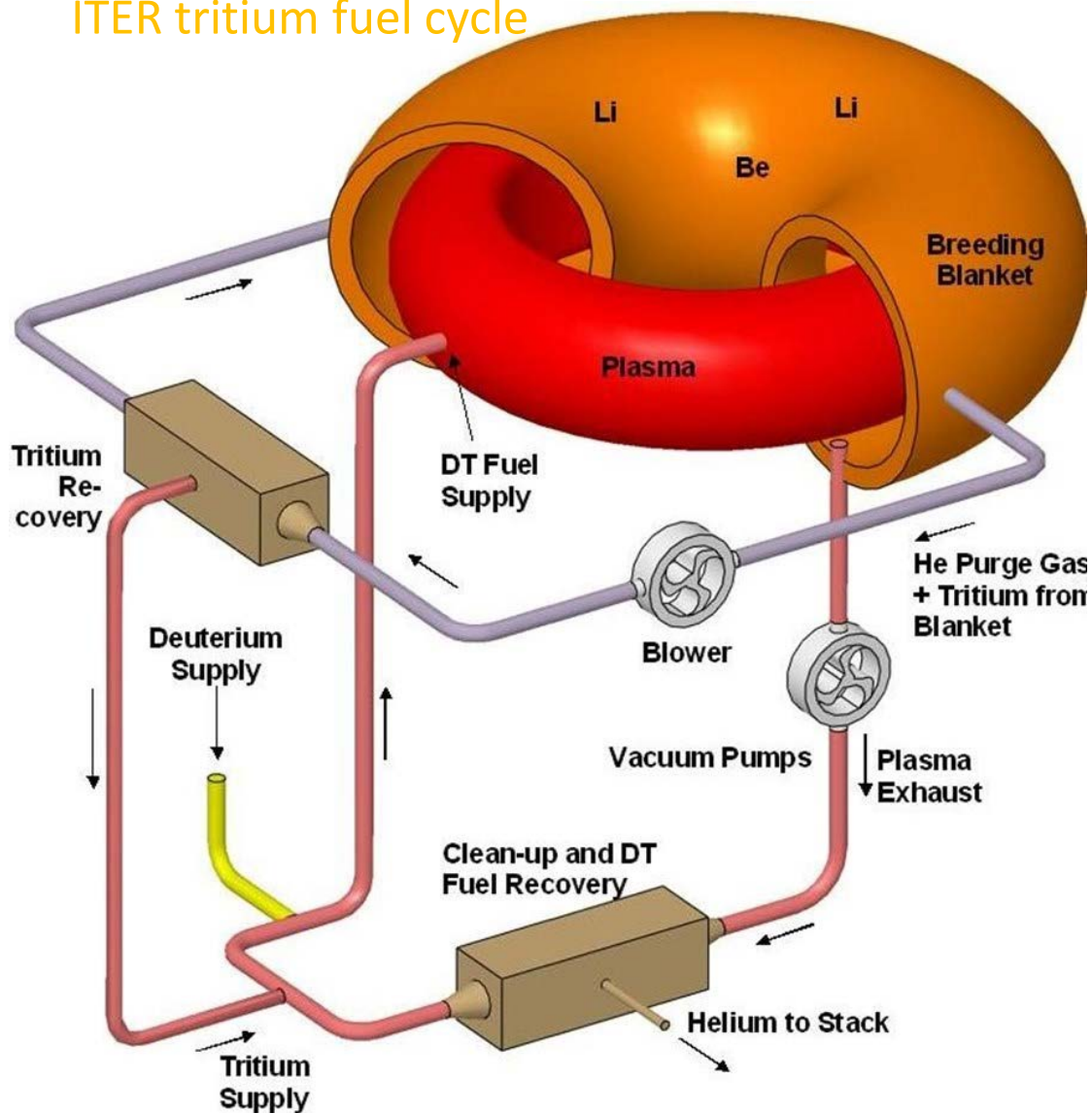
Confinement & detritiation

- Process
- Glovebox
- Air

Gas analysis

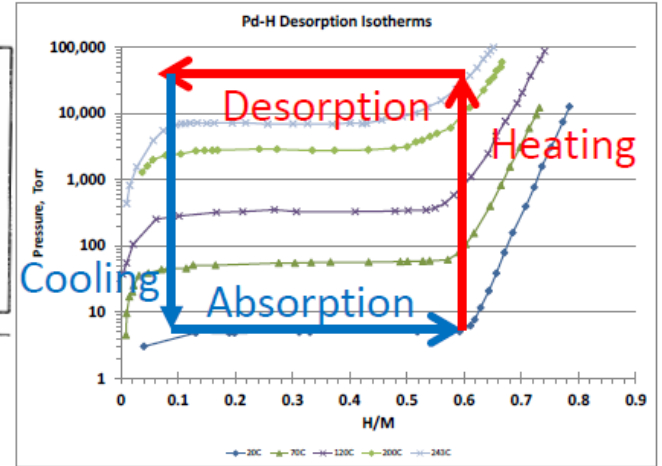
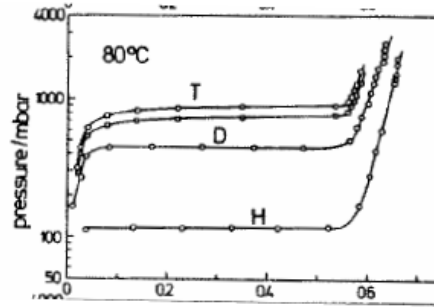
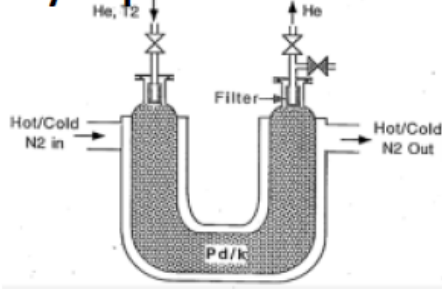
Process control

Tritium accountability

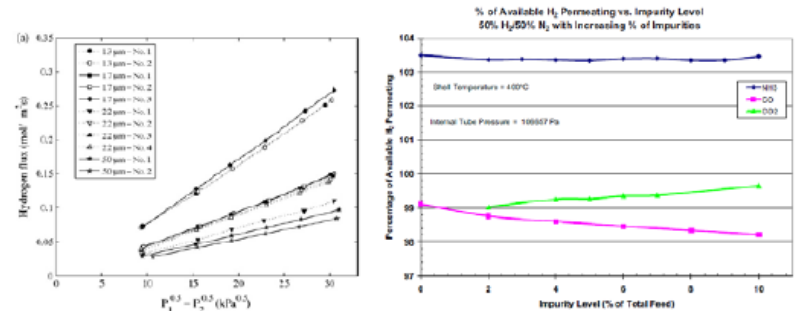
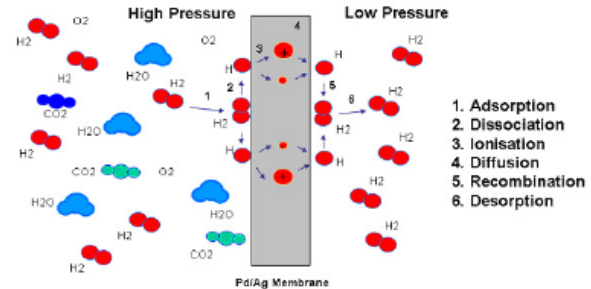


Impurity removal from bulk hydrogen isotopes (e.g., > 95%)

Flow-Through Bed/ Primary Separator



Diffuser/Permeator (Pd-Ag Membrane)



Diffusion vs. Permeation

Diffusion

Average velocity of gas molecules by Maxwell-Boltzmann's distribution

$$v = \sqrt{\frac{8kT}{\pi m}} = \sqrt{\frac{8RT}{\pi M}}; \quad v_H/v_D = \sqrt{2}; \quad v_H/v_T = \sqrt{3}; \quad v_D/v_T = \sqrt{3/2}$$

Molecular kinetics gives incident flux to wall surface under pressure P

$$J = nv = \frac{P}{\sqrt{2\pi m k T}}; \quad J_H/J_D = \sqrt{2}; \quad J_H/J_T = \sqrt{3}; \quad J_D/J_T = \sqrt{3/2}$$

Permeation

Sieverts' law - a rule to predict the solubility of gases in metals

$$H_2 \text{ (molecular gas)} \leftrightarrow 2 H \text{ (dissolved atoms)}; \quad K = \frac{c_{at}^2}{P_{mol}}; \quad c_{at} = \sqrt{K P_{mol}}$$

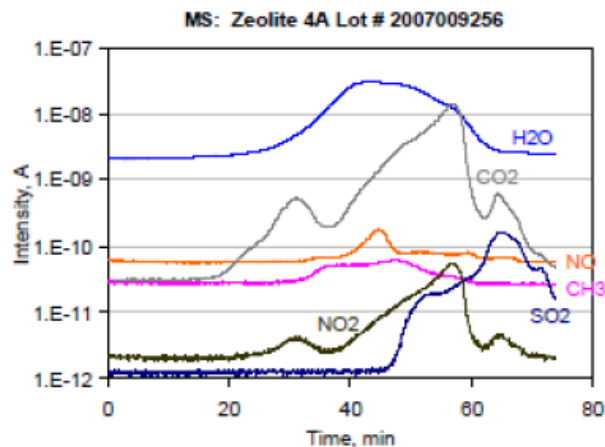
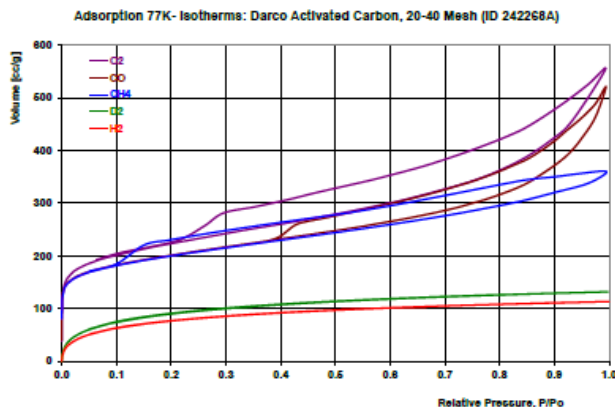
Permeation flux

$$F = \phi \frac{\sqrt{P_{up}} - \sqrt{P_{down}}}{\delta} A; \quad \phi_{HD}/\phi_{H_2} = 0.9; \quad \phi_{HD}/\phi_{D_2} = 1.1$$

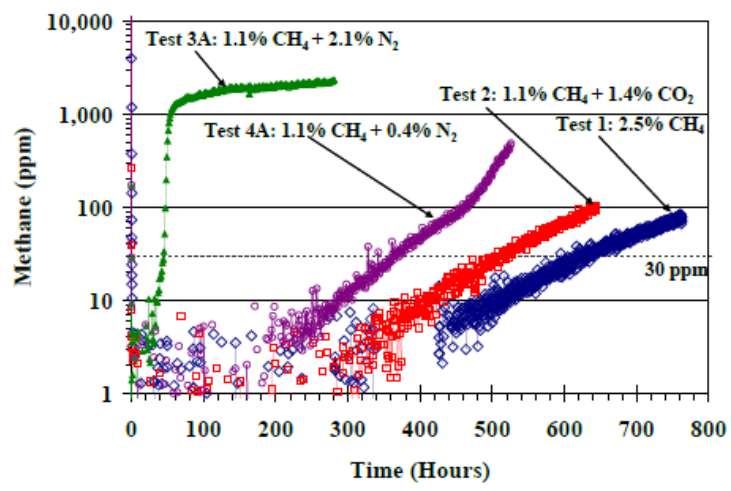
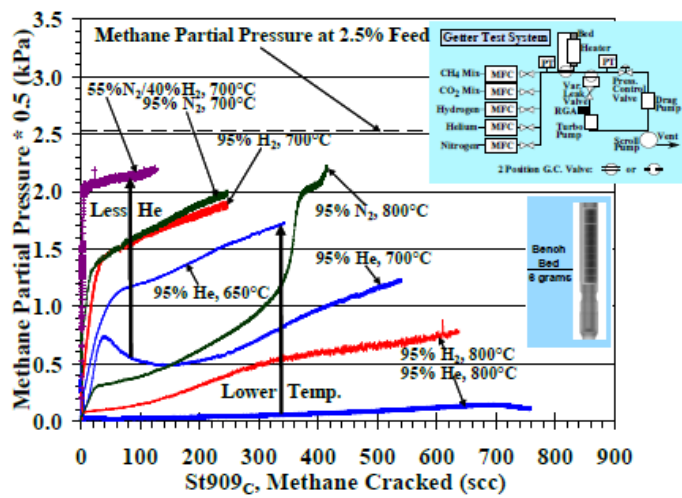


Tritium recovery from bulk impurities (e.g., > 95%)

Activated Carbon/ Molecular Sieve



Non-Evaporative Getters (NEG)



Isotope separation

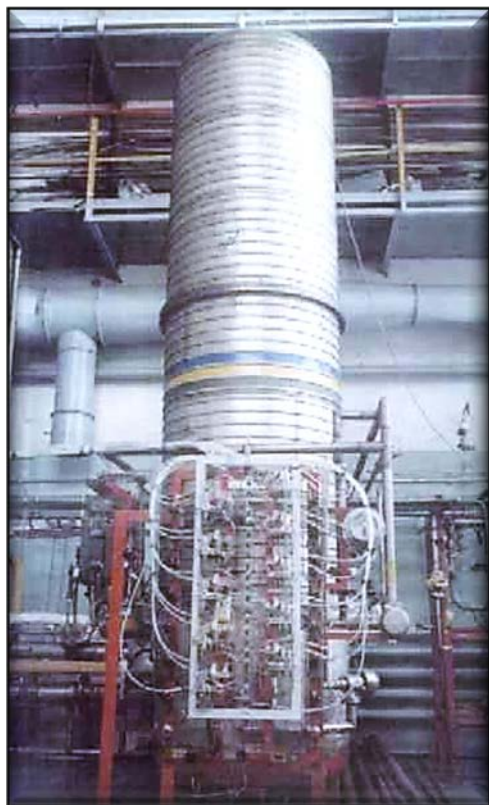
H_2, D_2, T_2

Hydrogen Isotope Separation Process

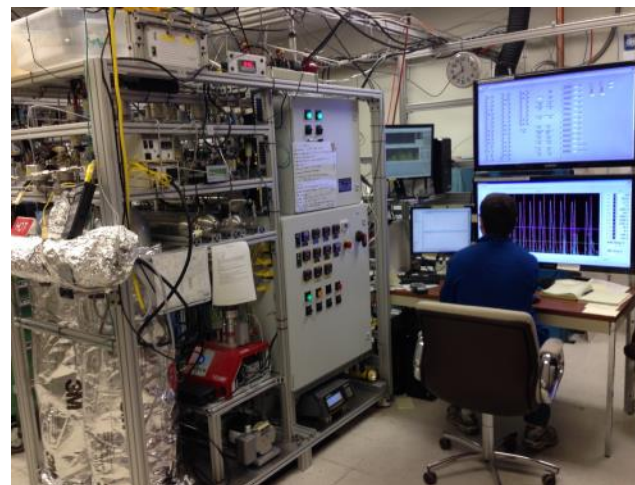
H_2
 D_2
 T_2

- Six species with different boiling points

Species	H_2	HD	HT	D_2	DT	T_2
Boiling Point, Kelvin	20.7	22.1	23.5	23.8	25	25.5



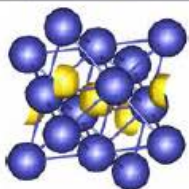
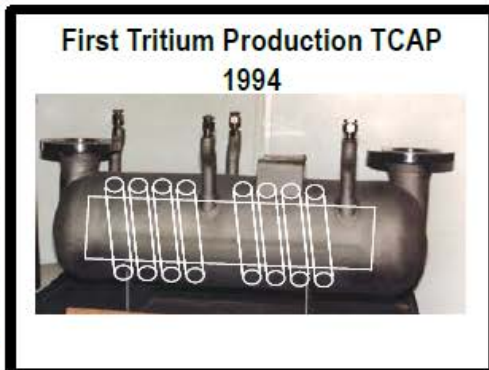
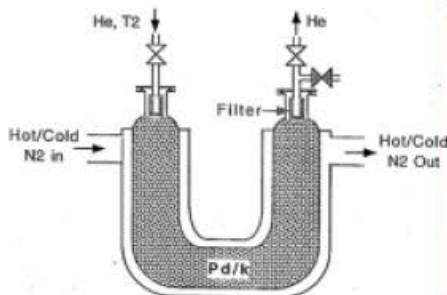
Cryogenic Distillation



TCAP - Thermal Cycling Absorption Process
(palladium has the highest isotopic effect)



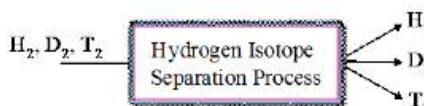
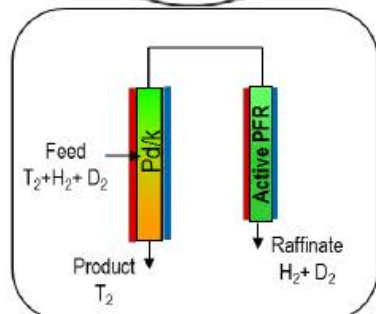
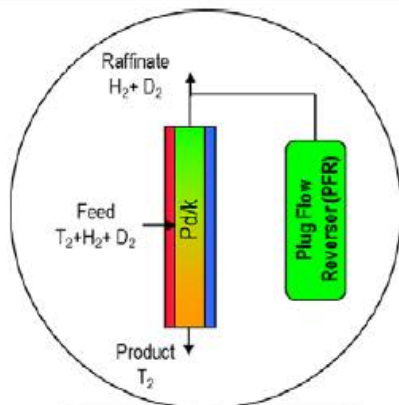
TCAP development for isotope separation



Palladium favors lighter hydrogen isotope, especially at low temperatures



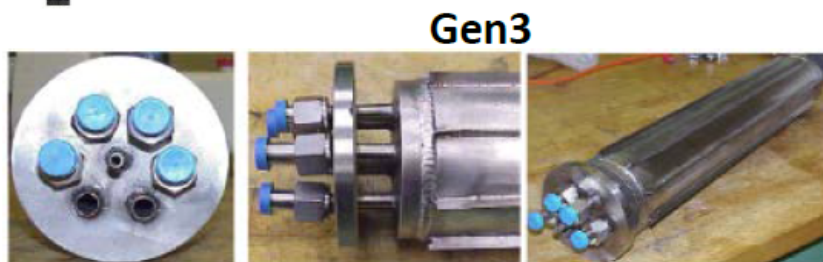
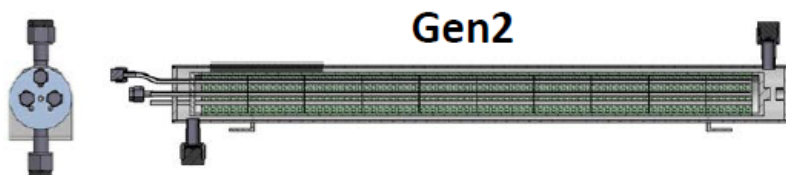
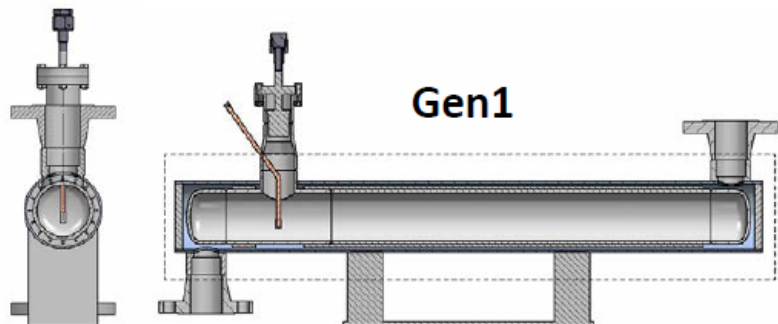
Molecular Sieve favors heavier hydrogen isotope (active PFR)



Rochester LLE TCAP - 2014



Hydride bed design for tritium storage



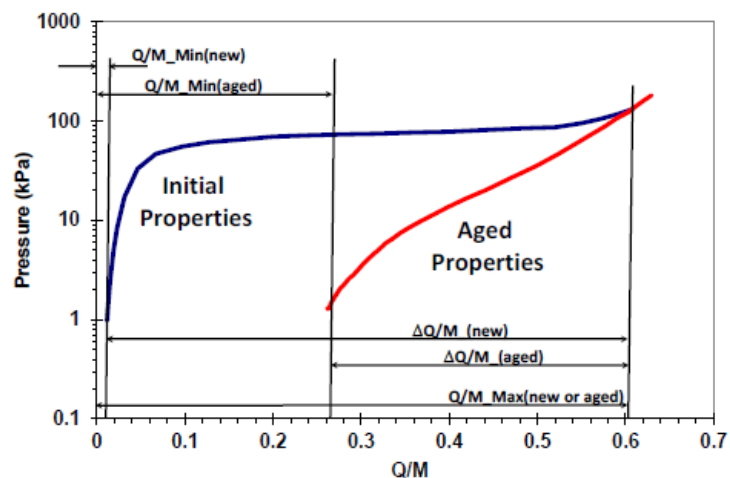
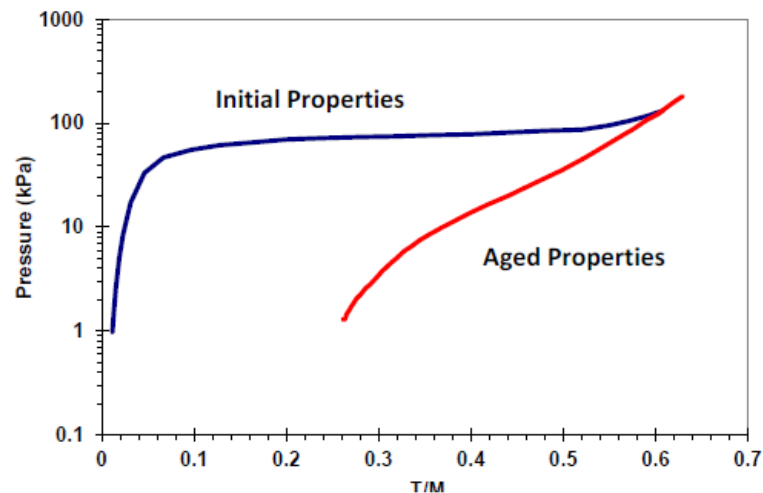
Fully Assembled End View

Fully Assembled End Cap detail

Fully Assembled General View



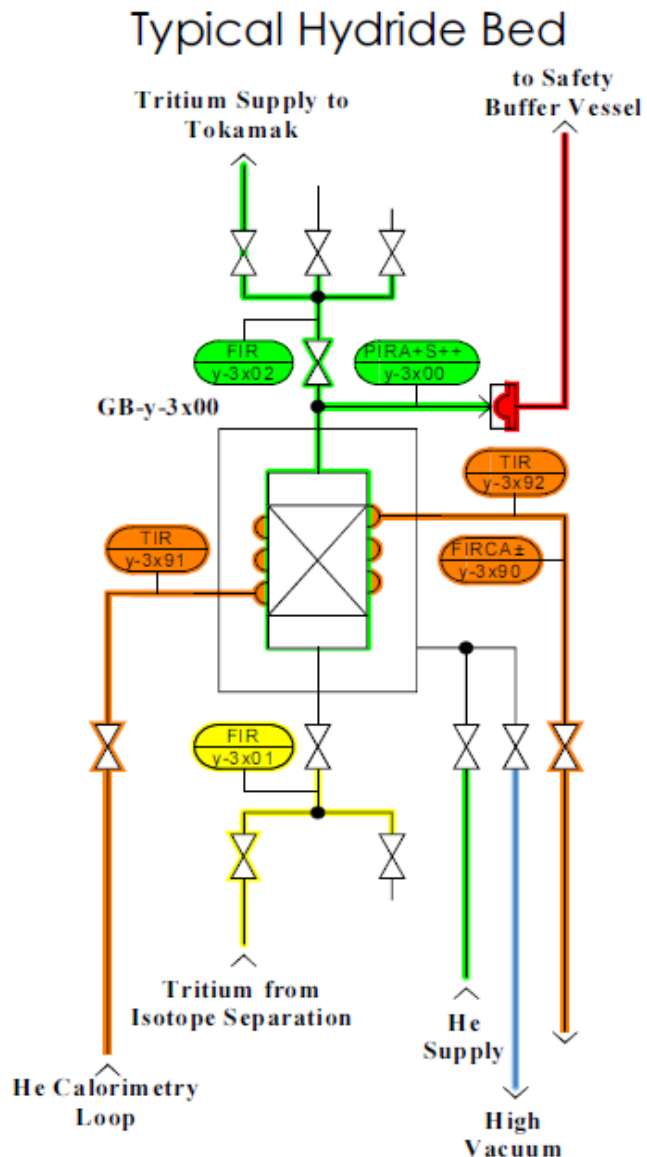
Fully Assembled Process Vessel Side View



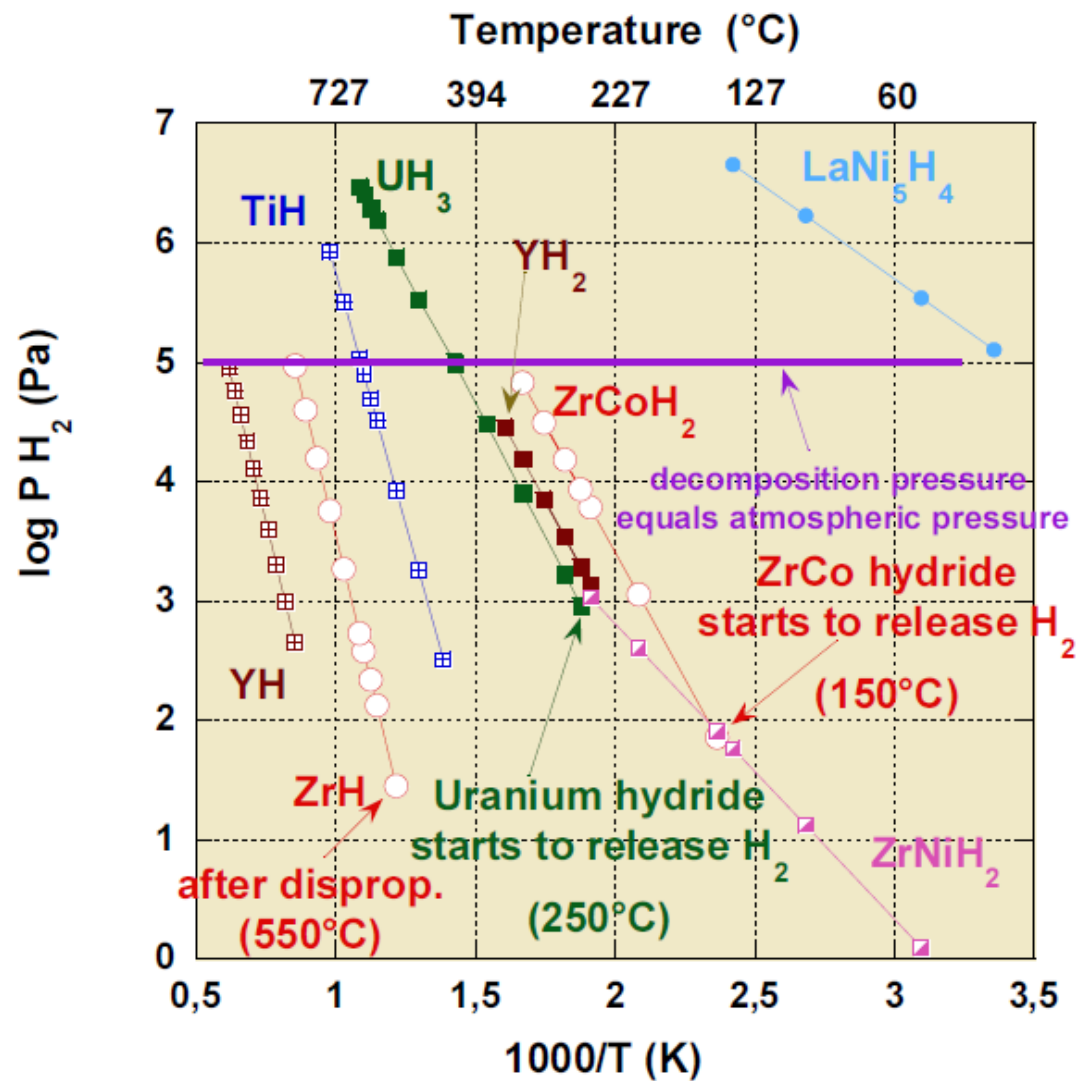
Three Generations of Metal Hydride Storage Bed Development



Example - Storage and Delivery - Jacketed Vessels, Hydride Selection

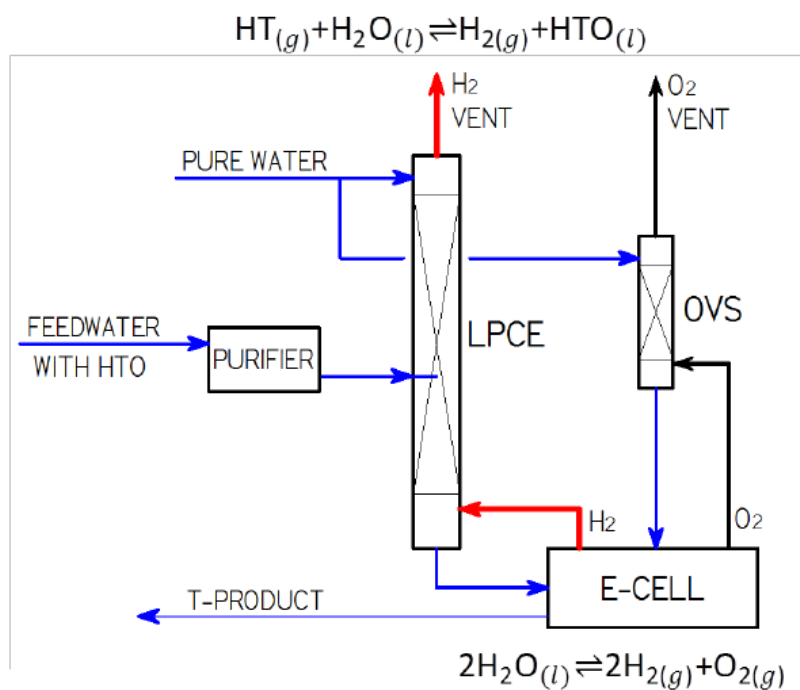


Van't Hoff Diagram for Various Hydrides



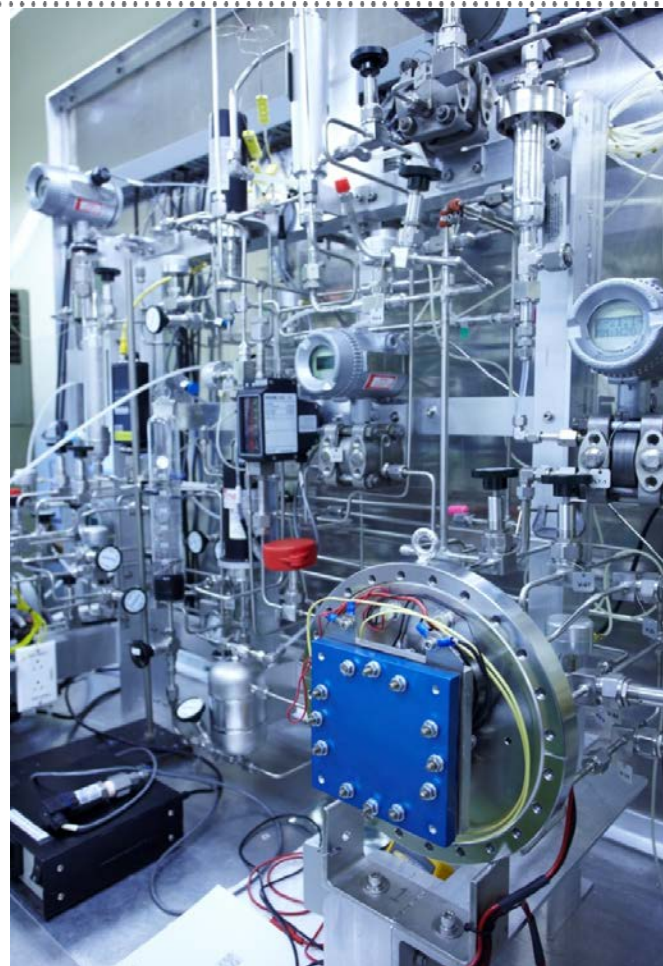
David Babineau, et al., Review of the ITER Fuel Cycle, 23rd IAEA FEC, 2010.

Water Detritiation System



H. Boniface, A Practical Process for Light Water Detritiation at Large Scales, Pacific Basin Nuclear Conference, 2014

Information from: Ian Castillo, Overview of AECL's Tritium Compatible Electrolyser Program, Tritium Focus Group Meeting, Idaho National Lab, 2014 Sep 24-25



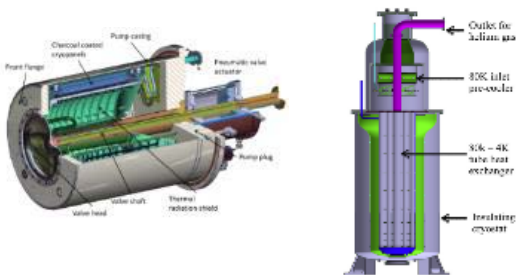
Small CECE Process at AECL

C. Muirhead, et al., Advanced Combined Electrolysis & Catalytic Exchange (CECE) Process for Hydrogen Isotope Separation in Water Decontamination Applications, 64th Canadian Chem. Eng. Conf. Niagara Falls, 2014

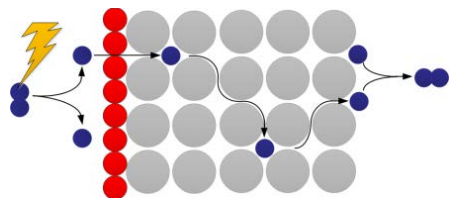


Vacuum and pumps

Cryosorption pumps

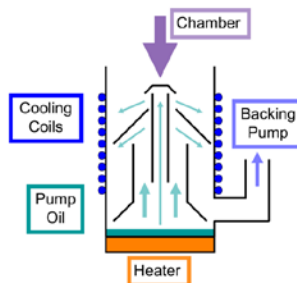


Snail pump under test at LANL.

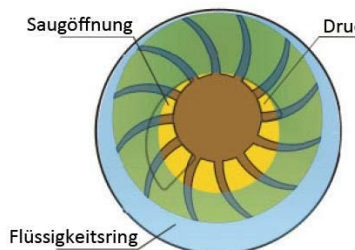


Metal Foil Pump (MFP)

Vapor diffusion pump



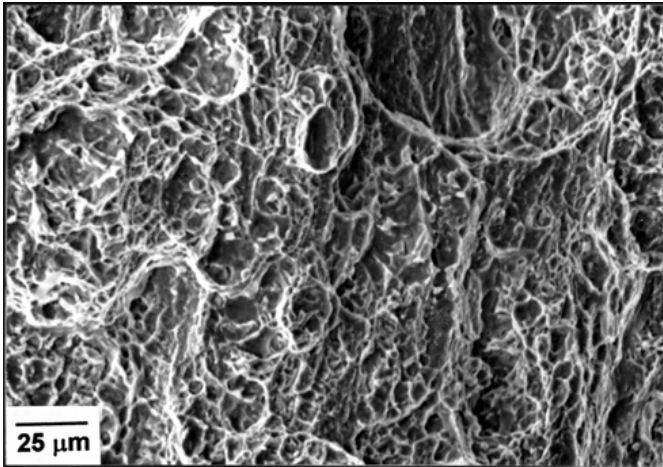
discontinued Normetex – complete dry and fluid tight vacuum pump



Liquid ring pumps (LRPs)

Th. Giegerich, Chr. Day, Tritium processing technology developments at KIT for nuclear fusion reactor, KIT/ITEP, 15 May 2018
 S. Willms, Tritium Science and Technology in the Future, 2010 Tritium Conference Tutorial Lectures, Nara, Japan, 2010.10.29

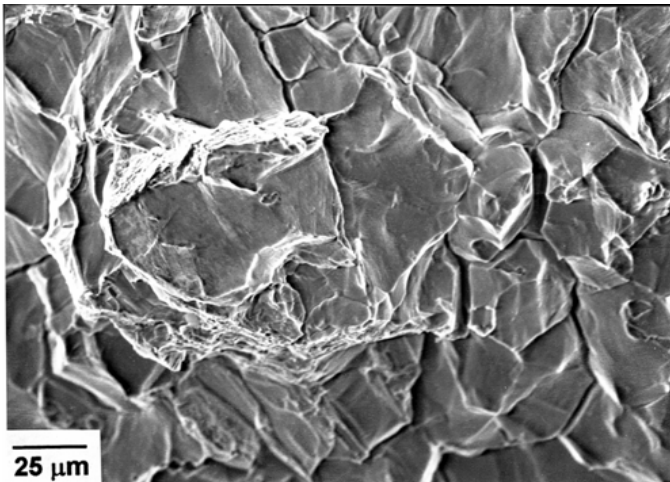
Tritium material compatibility evaluation



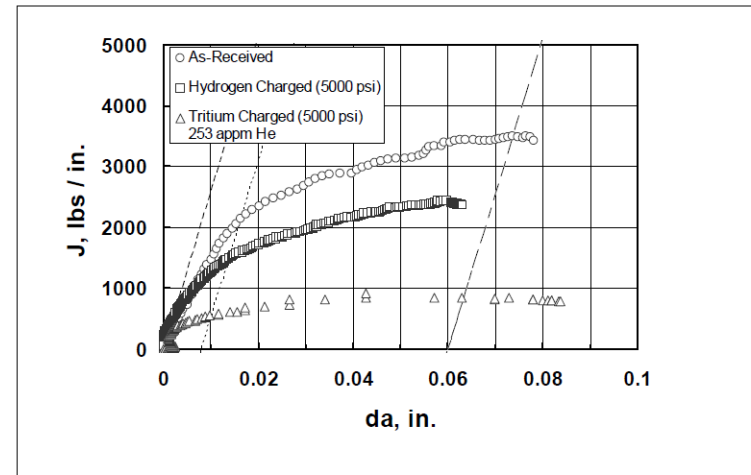
Unexposed stainless steel



Helium Hardened Microstructure



Tritium-Exposed & Aged



Conventionally Forged Type 21-6-9 Stainless Steel

Michael Morgan, "Effect of Forging Process on Tritium Compatibility of Stainless Steel", *Hydrogen and Helium Isotopes in Materials*, February 6-7, 2008, Sandia National Laboratory, Albuquerque, NM

Tritium Interaction with Metal Surface

H₂O always exists in H₂

Any “clean” hydrogen includes more than a few ppm of H₂O

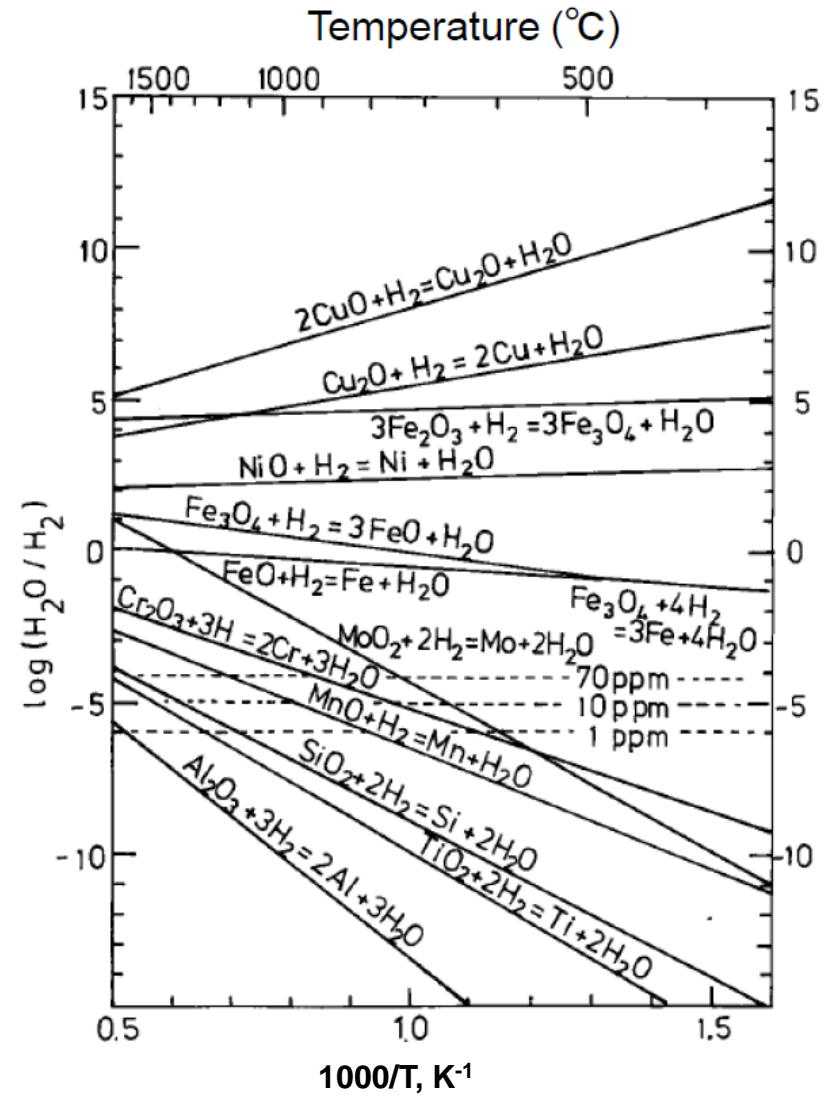
Oxidation and reduction on metal surface



$$\Delta G = -RT \ln K$$

$$= -RT \left[\frac{P(H_2O)}{P(H_2)} \frac{\alpha(M)}{\alpha(MO)} \right]$$

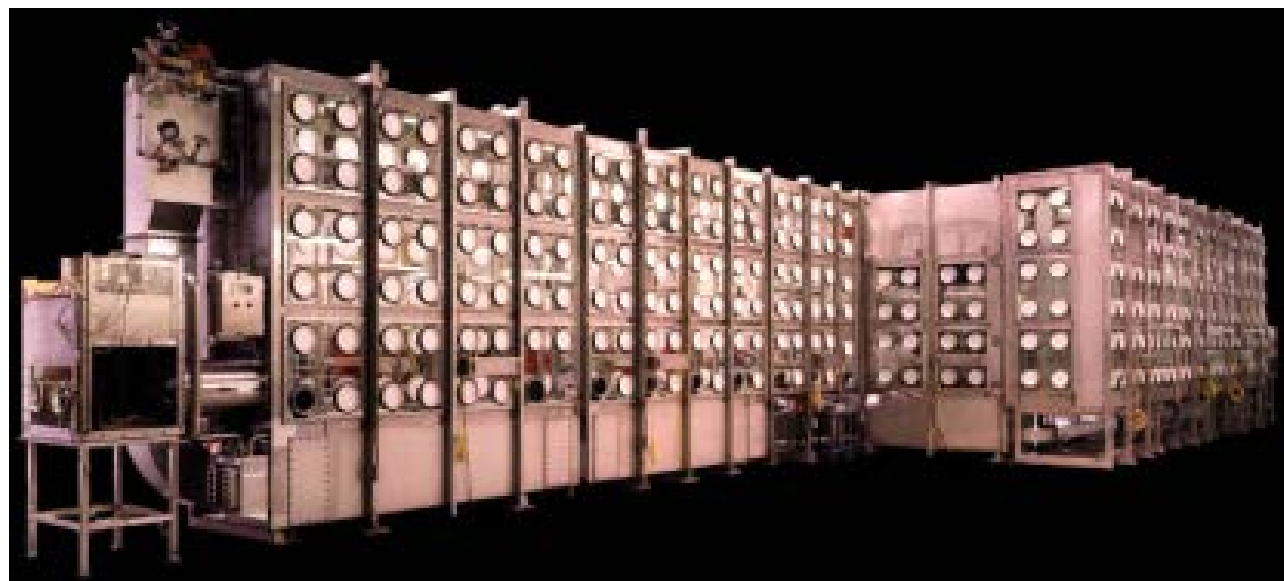
$$\approx -RT \left[\frac{P(H_2O)}{P(H_2)} \right]$$



T. Tanabe, What is tritium, 2010 Tritium Conference Tutorial Lectures, Nara, Japan , 2010.10.29

Tritium confinement and radiological safety

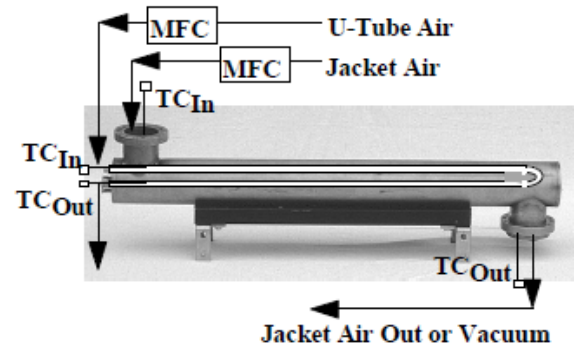
- Primary, secondary and tertiary confinement
- Area monitor and personnel surveillance
- Permeation barriers
- Occupational and environmental tritium monitoring
- Maintenance systems
- Waste handling, characterization and processing
- Decontamination and decommissioning
- Personnel protection equipment
- Best practice and safety culture



Tritium Accountability

State-of-the-Art

- – P-V-T Composition: 2% or less
- – Dedicated calorimeters: $\pm 0.25\%$ in 6-8 hours
- – In-bed accountability (IBA): 1-2%



Non-destructive Detection of Hydrogen in Solids

- Direct: Nuclear reaction, Ion beam analysis, diffraction (neutron, ion, X-ray, electron) (Only for hydrogen in near surface layers)
- Electron transition: EELS, UPS, LIF
- Vibrational and rotational transition: IR, Raman, Low energy EELS (Limited to surface or transparent materials)
- Diffraction, Channeling: Neutron, Ion, Electron
- Magnetic moment: NMR, ESR
- Indirect: Elastic energy (Internal Friction, Gorsky effect)
- Electronic and Mechanical property change
- All are not applicable at higher temperatures
- And few methods can be applied in vessel tritium analysis

State-of-the-Art

- – (see Safety and Environmental presentation)

Demo Function/Requirements

- – Some type of on-site or permitted disposal site will be needed

Gaps

- – Assurance that fusion will not suffer from “Yucca Mountain syndrome”
- – Recycle option from “Safety and Environmental” presentation needs to include tritium recovery functionality in project scope

S. Willms, Tritium Science and Technology in the Future, 2010 Tritium Conference Tutorial Lectures, Nara, Japan , 2010.10.29



Areas to further improve / develop

- Fuel Cleanup: Technology improvement
- Isotope Separation: Tritium inventory and technology improvement
- Tritium Storage and Delivery: Technology and assaying improvement
- Water Detritiation: Technology improvement. Need low-level tritiated water processing system.
- Pumping: Need larger capacity pumps
- Effluent Detritiation: Would benefit from system which does not produce water
- Gas Analysis: Technology improvement
- Process Control: Duty cycle and flowrate will require better control
- Modeling: Accurate, easy-to-use models will be essential

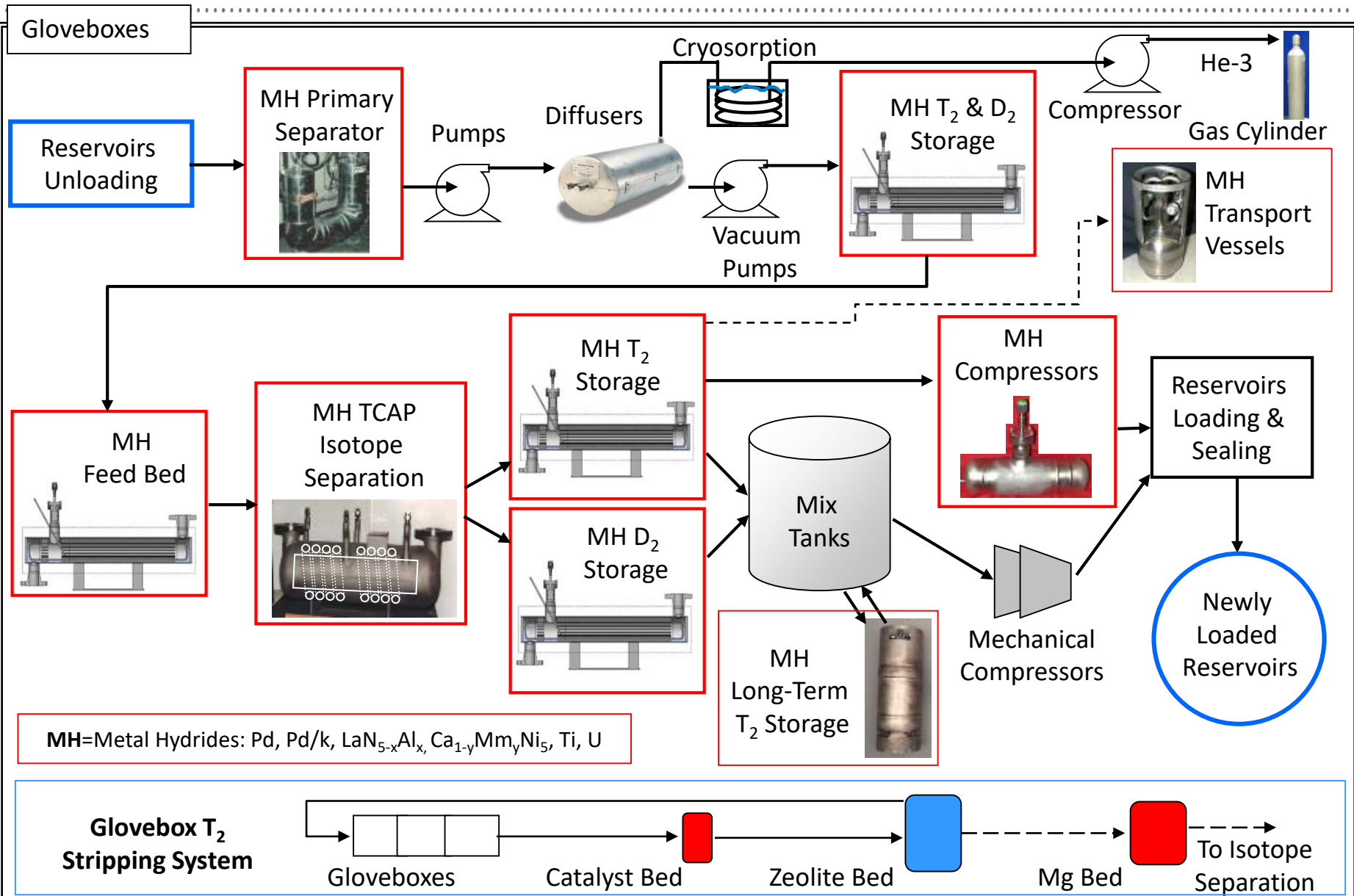
S. Willms, Tritium Science and Technology in the Future, 2010 Tritium Conference Tutorial Lectures, Nara, Japan , 2010.10.29

Additional Issues

- Tritium recovery in waste components
 - ✓ ITER: high temperature bake out in tritium controlled hot cell
- Hold-up in hidden and cooler areas (gaps, ducts, behind in vessel components such as RF launchers, etc.)
 - ✓ – Maintain similar temperature in these components
- Wall conditioning produced tritium stream
 - ✓ – ITER: use available tritium exhaust and recovery system
- Impurities introduced into vessel due to abnormal or accidental conditions including from auxiliary systems such as NBI (SF_6 insulator)
 - ✓ – Limit material choices for in-vessel and auxiliary systems

S. Willms, Tritium Science and Technology in the Future, 2010 Tritium Conference Tutorial Lectures, Nara, Japan , 2010.10.29

Tritium Processing at SRS - The Largest Metal Hydride Based Tritium Facility in the World



The Fusion Fuel Cycle

All tritium processing systems contain the same core fuel cycle functions.

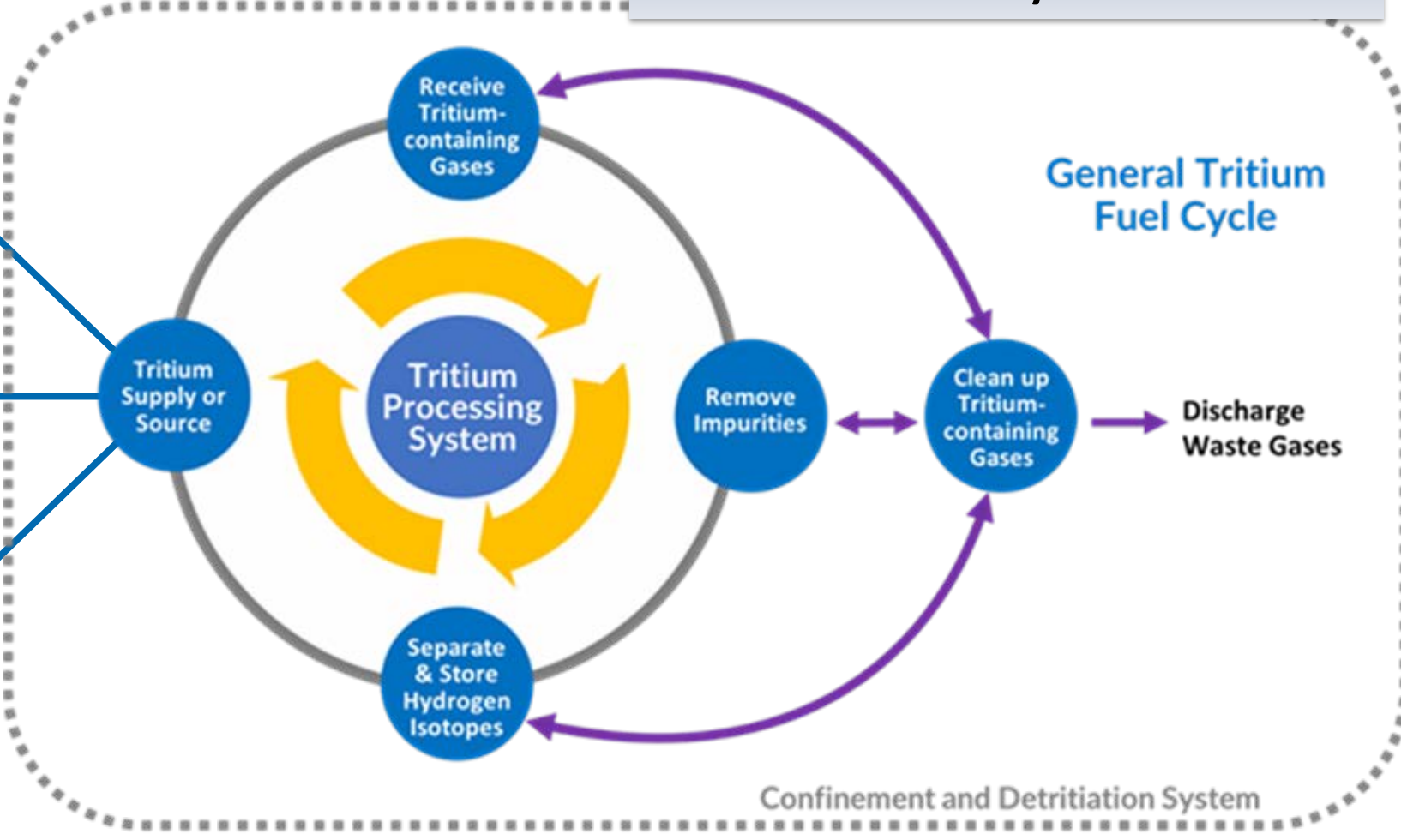


POWER



DEFENSE

COMMERCIAL



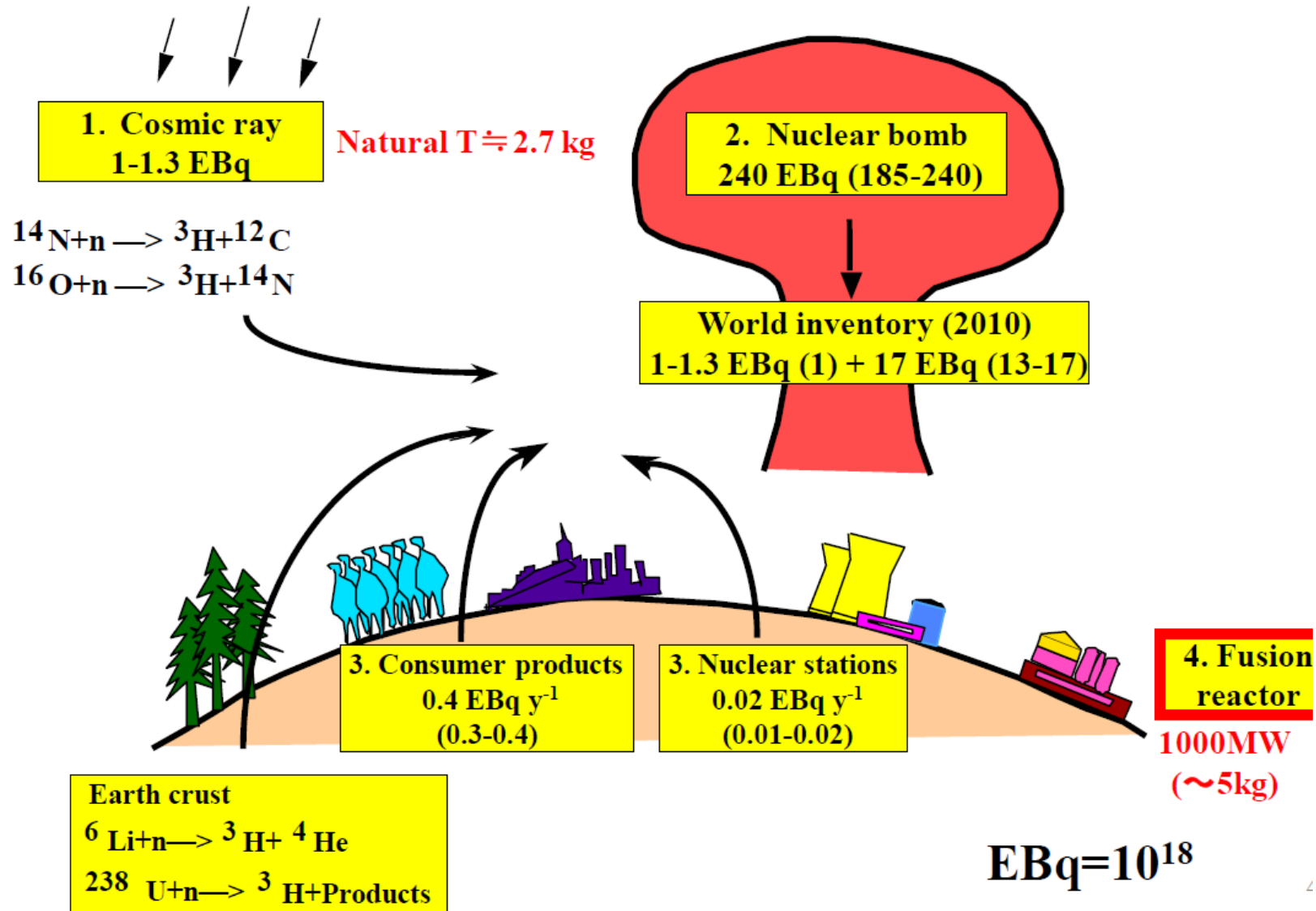
General Tritium Fuel Cycle

Discharge Waste Gases

Confinement and Detritiation System



Tritium and environment



Source: S. Konishi, TRITIUM and Environment, 2010 Tritium Conference Tutorial Lectures, Nara, Japan, 2010.10.29

Useful References for Tritium System Design

- DOE-STD-1129-2015 “DOE Handbook Tritium Handling and Safe Storage”, 2015
- William W Weaver et al, DOE/EH-0417, Technical Notice Issue No 94-01, “Guidelines for Valves in Tritium Service”, 1994
- F Mannone, Springer, Editor, "Safety in Tritium Handling Technology", Kluwer Academic Publishers, 1993
- International Atomic Energy Agency, "Safe Handling of Tritium: Review of Data and Experience", Vienna, 1991
- ASME B31.3 process piping code, ASME Boiler and Pressure Vessel Code Section VIII and Section II