Savannah River National Laboratory™

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We put science to work.™

An Overview of SRNL Tritium Activities

Greg Staack

Hydrogen Processing Group

Workshop on Tritium Control and Capture

October 27-28, 2015

SRNL-STI-2015-00582, Rev. 0

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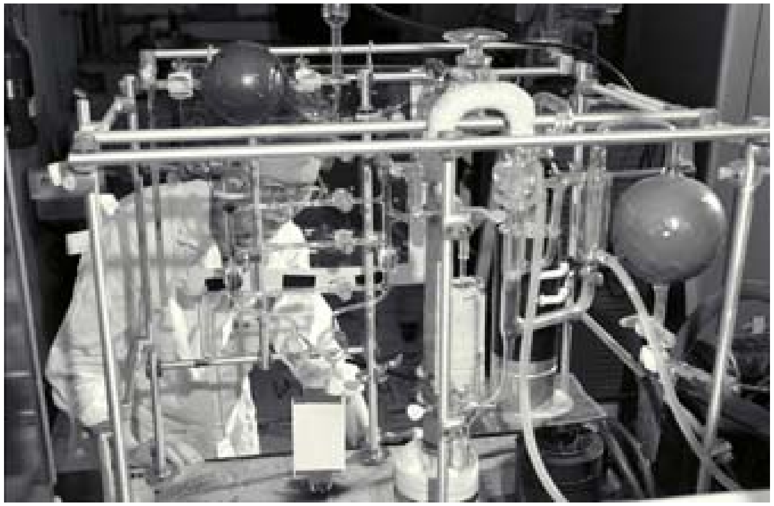
Savannah River Site

Savannah River Site - Production Years



SRS produced about 36 metric tons of plutonium from 1953-1988

- Began operation in 1953
- Produce and recover nuclear materials
  - Tritium
  - U, Pu
  - Cf, Np, Cm, Am,…
- Facilities
  - Heavy water extraction plant
  - Nuclear fuel and target fabrication facility
  - Five reactors
  - Two chemical separations plants
  - Naval Fuels
  - Waste management facilities
  - Tritium

Savannah River Tritium Enterprise Mission

Fill Tritium Reservoirs in support of the Nation's Nuclear Stockpile

- Extract new tritium from TPBARs irradiated in TVA reactors
- Recycle gas from returned bottles
  - Receive
  - Unload
  - Reprocess
  - Mix
  - Load
  - Ship
- Minimize environmental impact
- Provide a SAFE working environment
- One objective of the SRNL Hydrogen Processing Group is to provide technical support to the Facilities

Overview – Tritium Processing Research and Development Focus Areas

Remove impurities

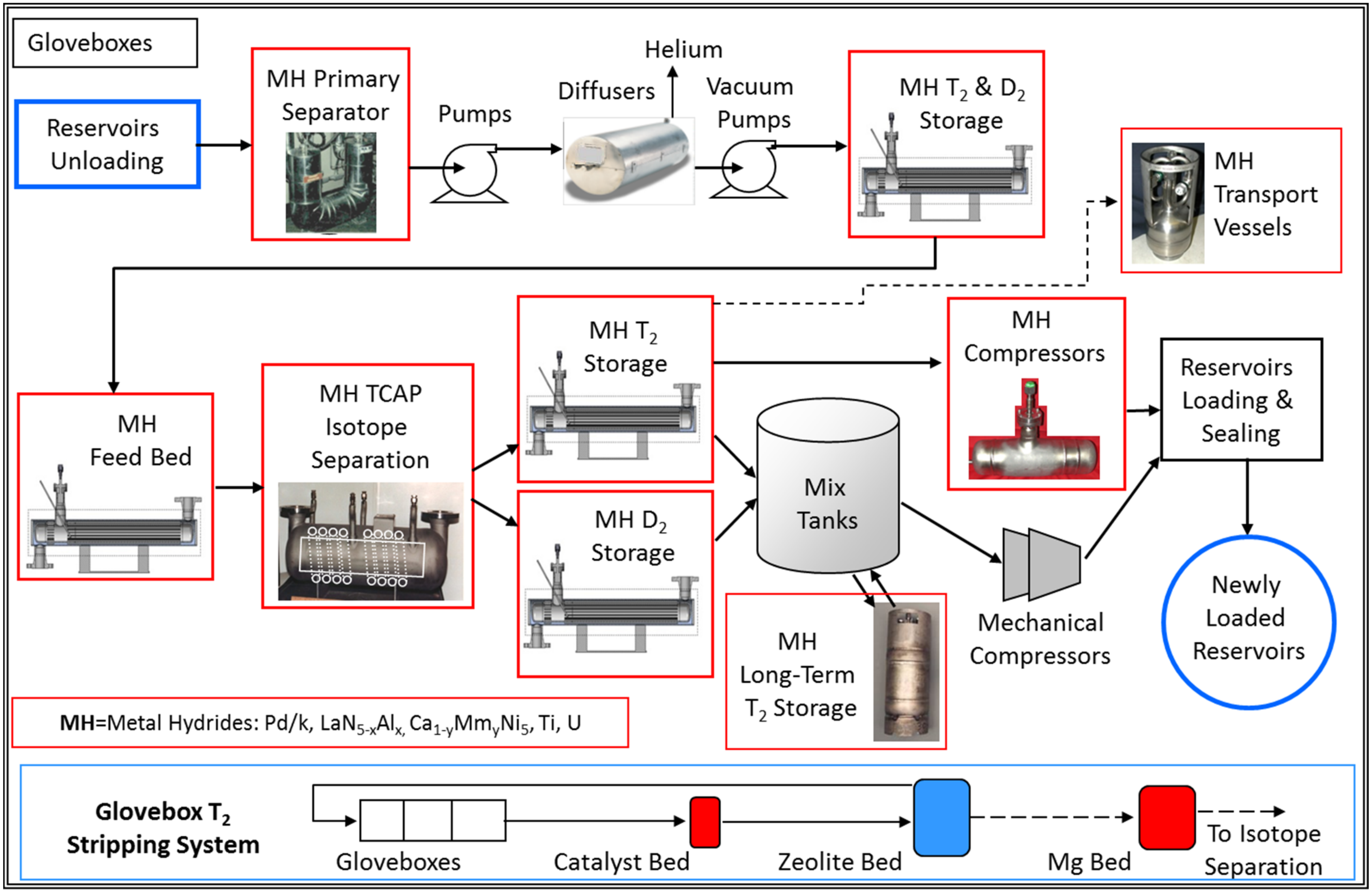
Separate hydrogen isotopes and Store

Clean up tritium-containing gases

- Impurity Removal
  - Hydrogen Separation from Impurities
  - He-3 cleanup
- Storage (Metal Hydride Beds / Tanks)
  - Materials options based on temperature/pressure/purity requirements
  - In-bed calorimetry
- Hydrogen Isotope Separation
  - Thermal Cycling Absorption Process (TCAP)
- Secondary Confinement
- Tritiated Water Processing / Detritiation Systems
  - Glovebox Atmosphere Detritiation (Stripper Systems)
  - Tritium Recovery from water (Z-Bed Recovery)
  - Detritiation of process effluent gases (TPS)
- Pumping (Evacuation/Circulation)
- Tritium Effects on materials

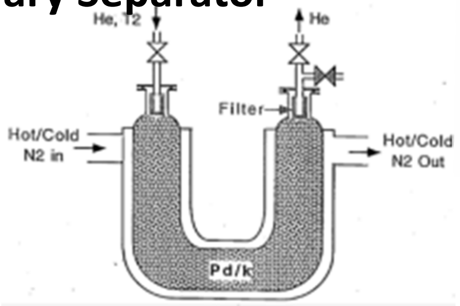
SRNL R&D provided current metal hydride-based Tritium Processes. Current research efforts are underway to improve efficiency, decrease operation costs, and to modularize processes

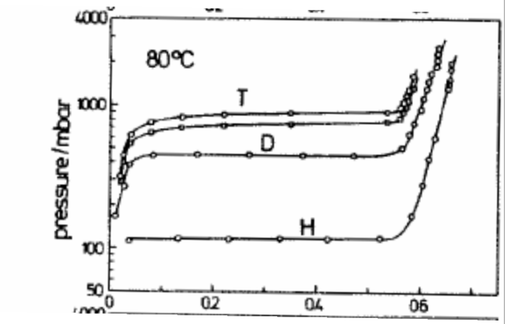
Tritium Processing at SRS – The Largest MH Based Facility in the World

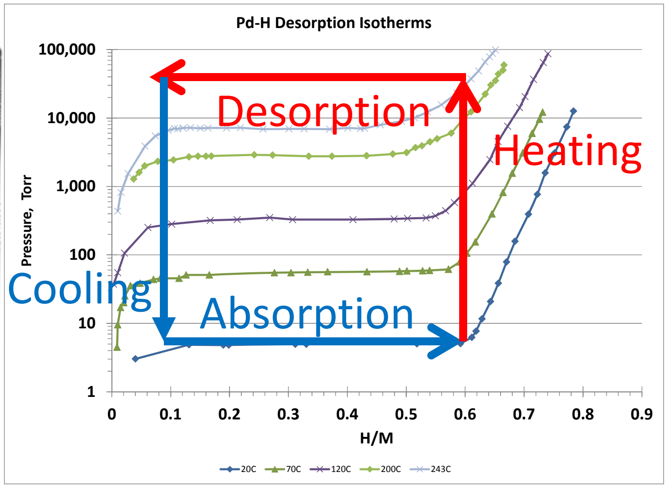


Impurity Removal by Bulk Separation

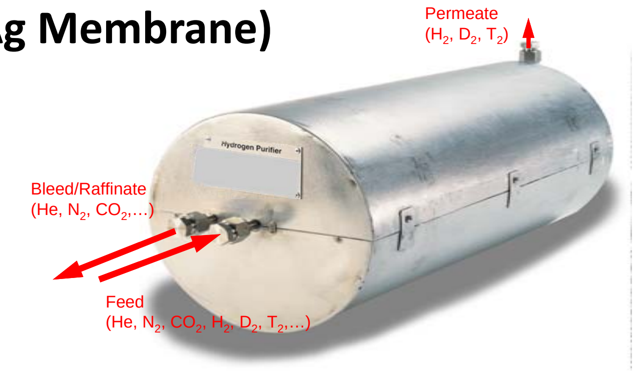
Flow-Through Bed/ Primary Separator

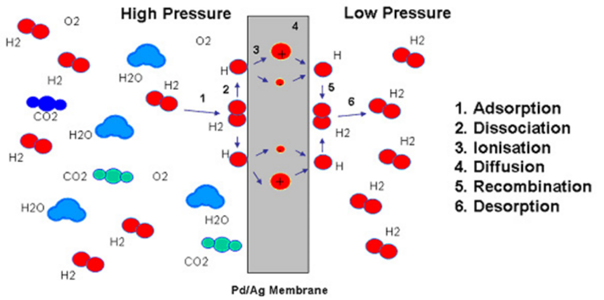


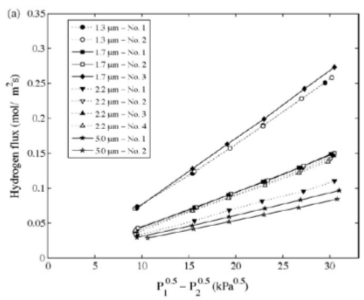


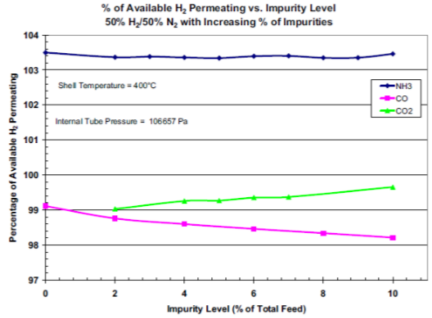


Diffuser/Permeator (Pd-Ag Membrane)



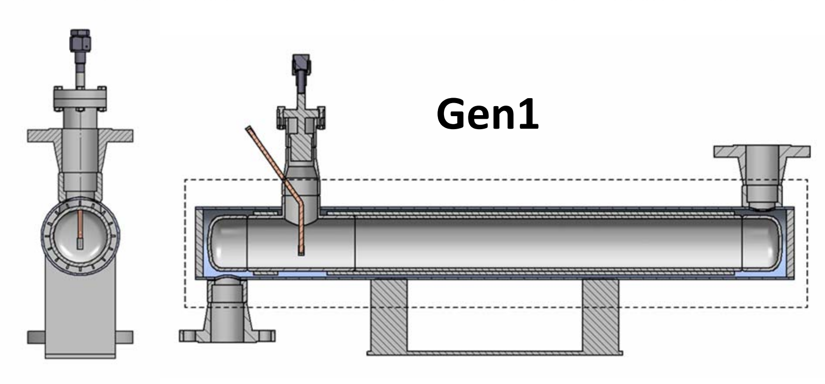




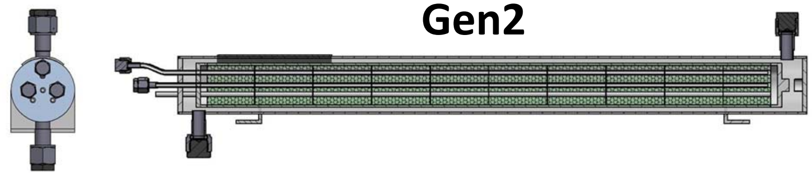


Tritium Storage: La-Ni-Al Metal Hydride for “He-3 Free” Gas Delivery

Gen1



Gen2



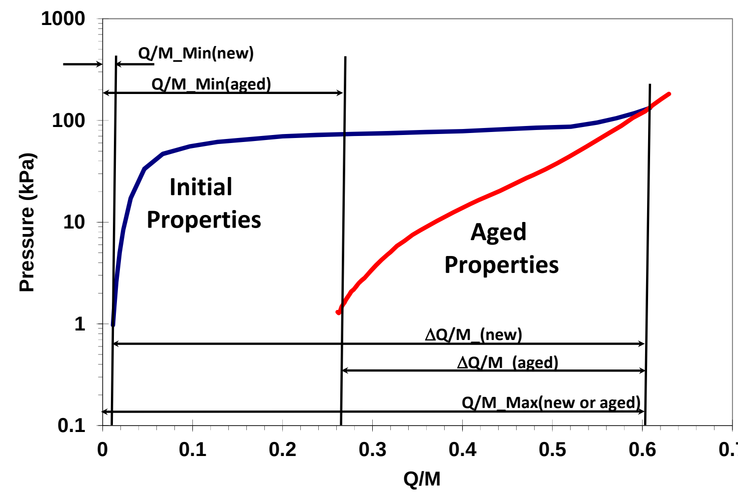
Gen3

LANA properties change with tritium aging.

- Formation of “heel”
- Decreased plateau pressure
- Eventual loss of plateau
- Eventual weeping of He-3

Limited life components

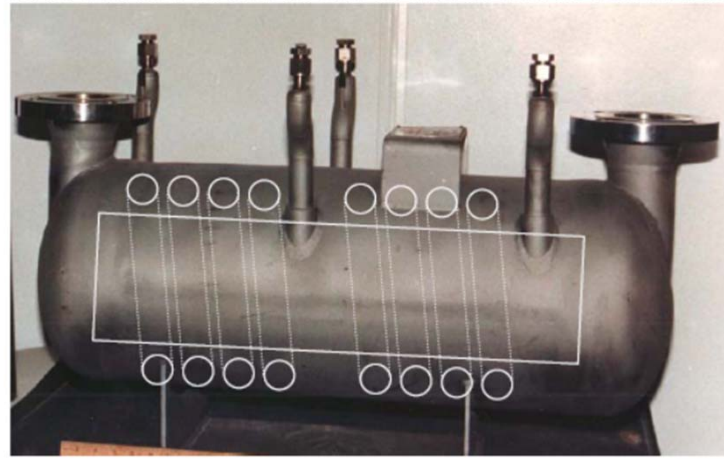
He-3 recovery from retired beds is being pursued.



Three Generations of Metal Hydride Storage Bed Development

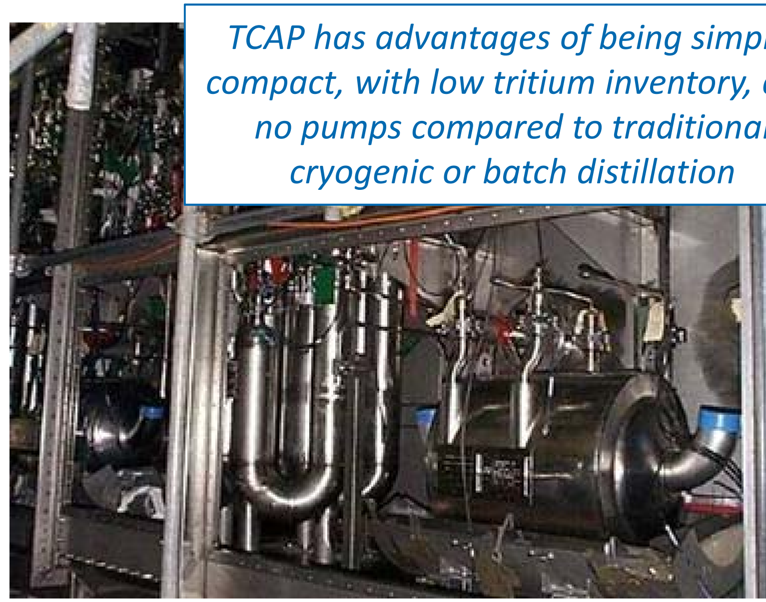
Thermal Cycling Absorption Process - TCAP

- "Heart" of the Tritium Facilities
- 4th generation of isotope separation at SRS
  - *Thermal Diffusion (1955-1986)*
  - *Fractional Absorption (1964-1968)*
  - *Cryogenic Distillation (1967- 2004)*
- Semi continuous TCAP invented at SRS in 1981. Originally coupled with a Plug Flow Reverser (PFR).
- Uses Pd/k to separate hydrogen isotopes
  - *Recover tritium (T_2)*
  - *Stack protium (H_2)*
- Hot and cold N_2 for thermal swing
- Feed, product, and raffinate gases stored on LANA beds



Original plant TCAP (1994)

TCAP has advantages of being simple, compact, with low tritium inventory, and no pumps compared to traditional cryogenic or batch distillation

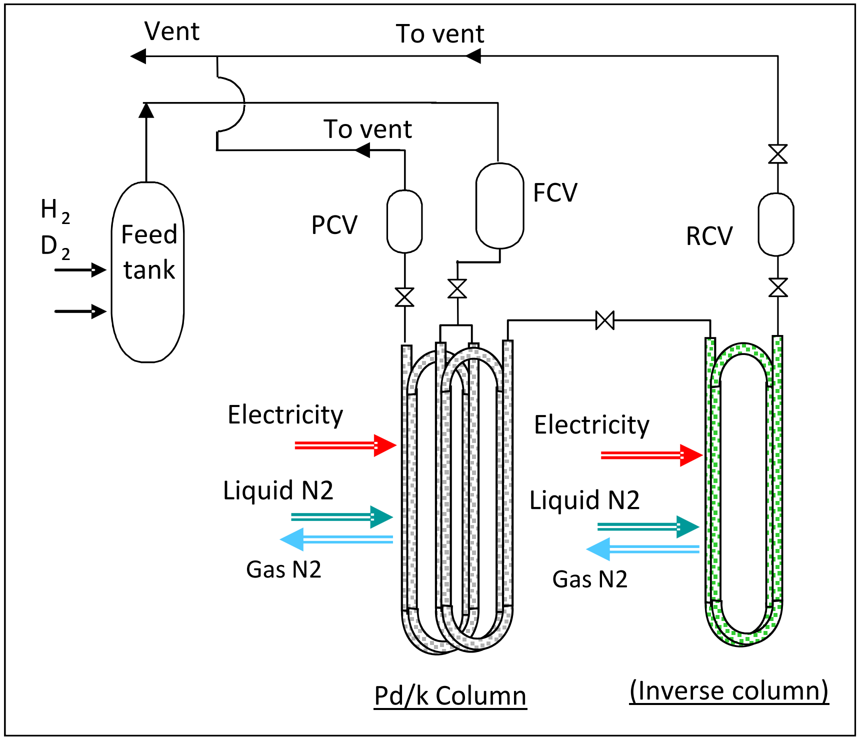


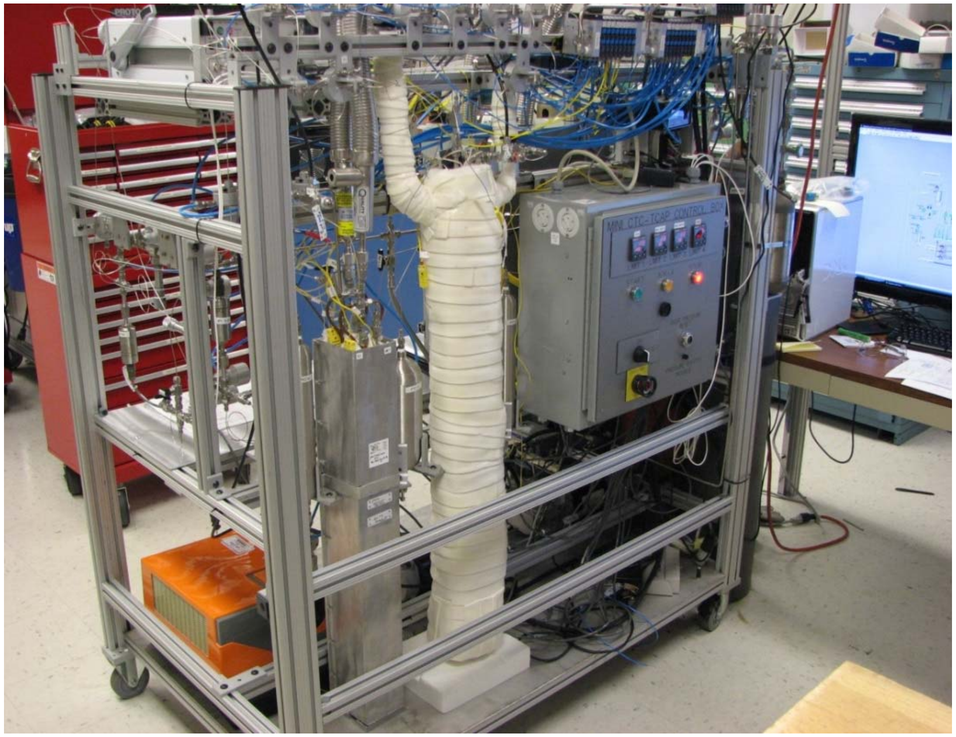
HT-TCAP installed in Tritium Facility (2004)

Advances in Hydrogen Isotope Separation Technology

TCAP (Thermal Cycling Absorption Process) advanced from

- "Pd/k – passive Plug Flow Reverser" to "Pd/k – Active Inverse Column"
- Results: Increased throughput and improved product purity
- Thermal Cycling with Hot and Cold N_2 to Electric Heating and once through LN_2 Cooling
- Results: Decreased cycle time

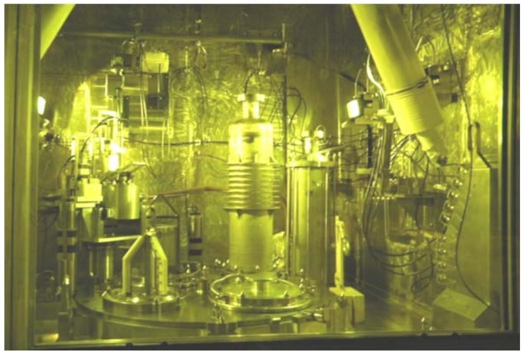


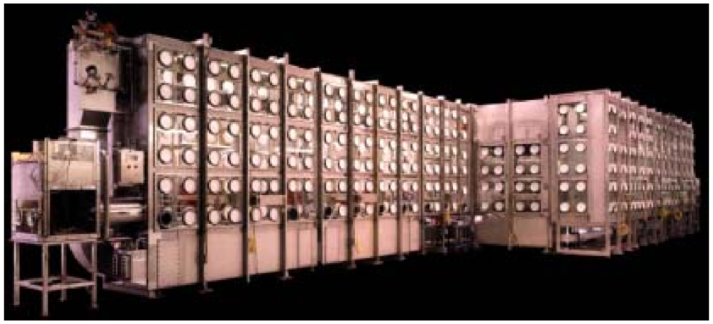


Glovebox Confinement Systems with Tritium Stripping

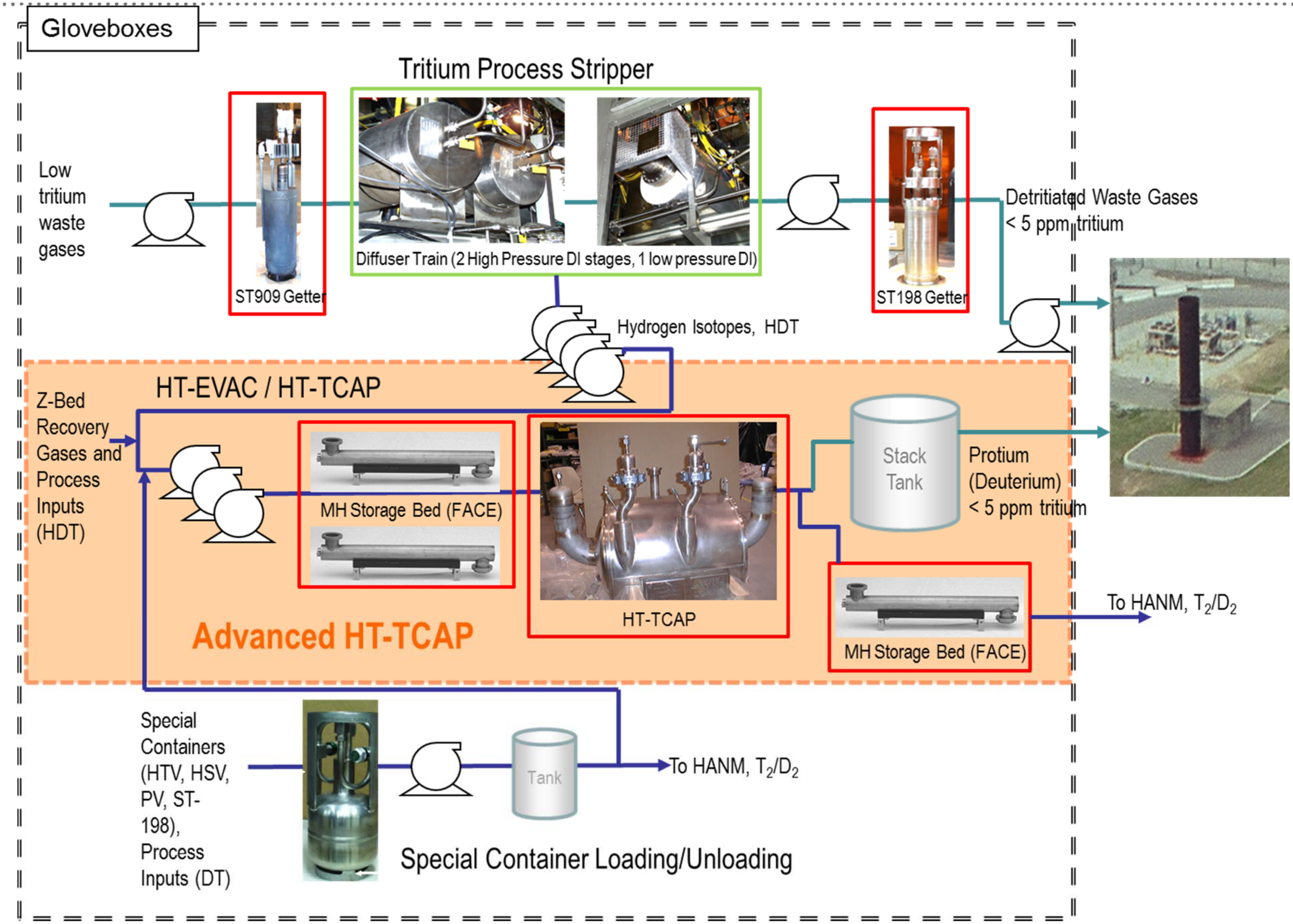
Glovebox/Secondary Confinement vs Fresh Air Hoods

- Legacy facilities used once through fresh air hoods – allowed product to escape
- In order to stem releases, closed loop glovebox system employed (RTF & TEF)
- Equipped with a secondary stripper for high activity
- Emissions were reduced from 100's kCi/year average (60's – 80's) to 10's of kCi/year average now
- Closed glovebox cleanup loop requires processing of byproducts – impact not fully realized during the early days of RTF (mid 90's)





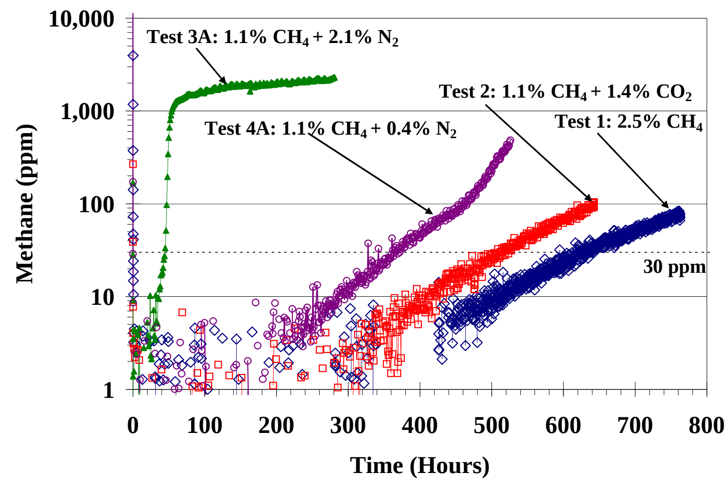
H-Area New Manufacturing – Tritiated Waste Gas Processing



Tritium Process Impurity Removal

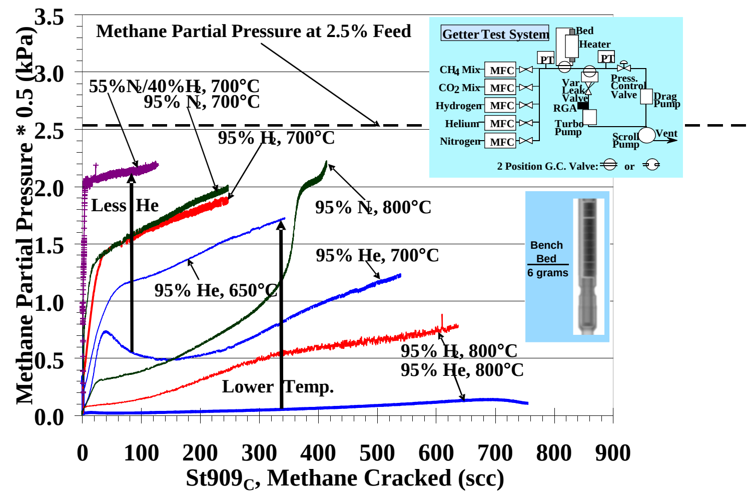
Molecular Sieve

- Large temperature swings
- May require upstream catalyst (e.g. water processing)
- Typically desorbed to regenerate bed, transferring impurities to a different process stream (Z-Bed Recovery or He-3 cleanup)



Non-Evaporative Getters

- Selected to "getter" various species, with or without decomposition
- Usually at elevated temperatures
- St909 crack methane, absorb carbon (not regenerated)
- St198 absorb tritium (regenerated)



Tritium Effects on Materials –Hydrides & Polymers

Hydrides:

– Pd based operations – (TCAP) long service life with clean gas

– LANA operations – limited service life due to He-3 formation

– Getters – depend on service conditions

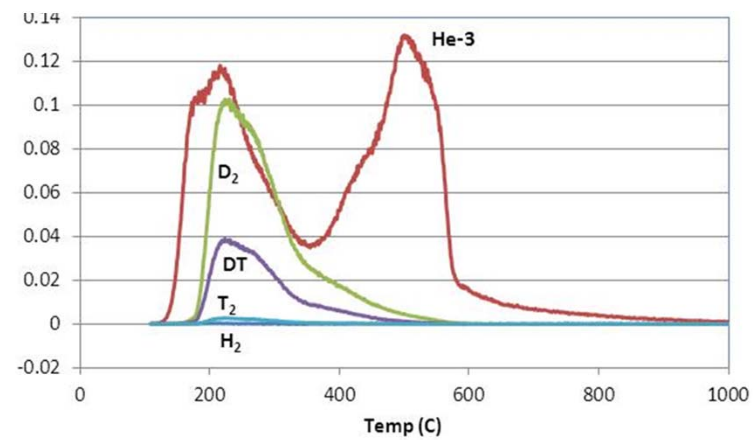
Polymers - a necessary compromise:

– Sometimes unavoidable in tritium systems (seals, lubrication, packing, etc.)

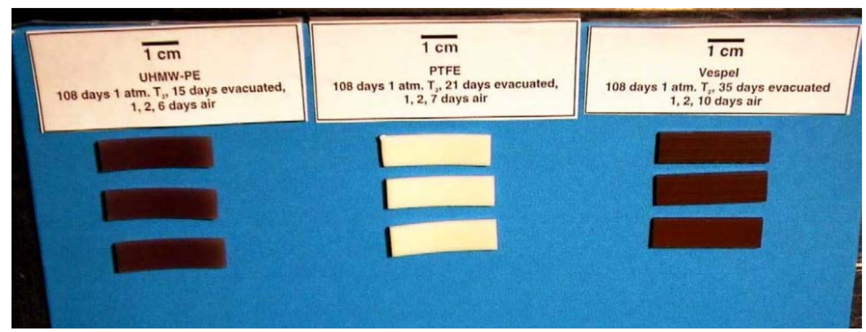
– Polyethylene can release hydrogen, methane and other hydrocarbons

– PTFE (Teflon®) can react to form TF

– EPDM and Vespel™ polyimide – OK



Gas release from tritium aged LANA.75



Tritium aged samples of UHMW-PE, PTFE, and Vespel

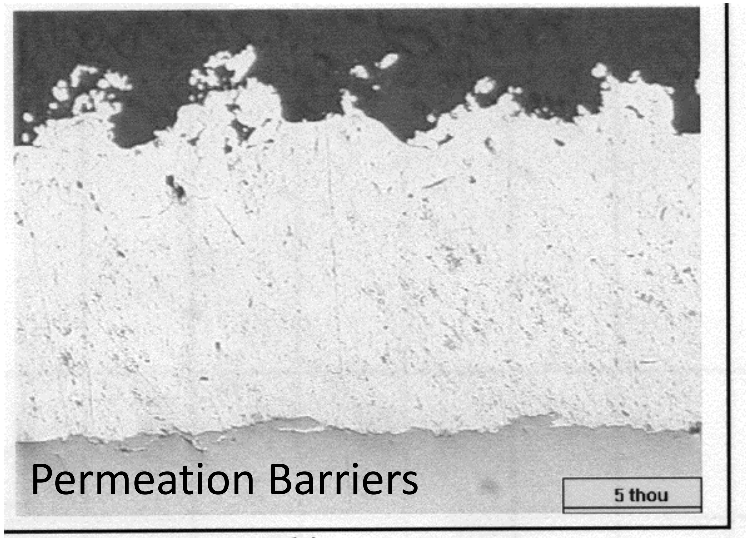
Tritium Effects on Materials –Steels & Coatings

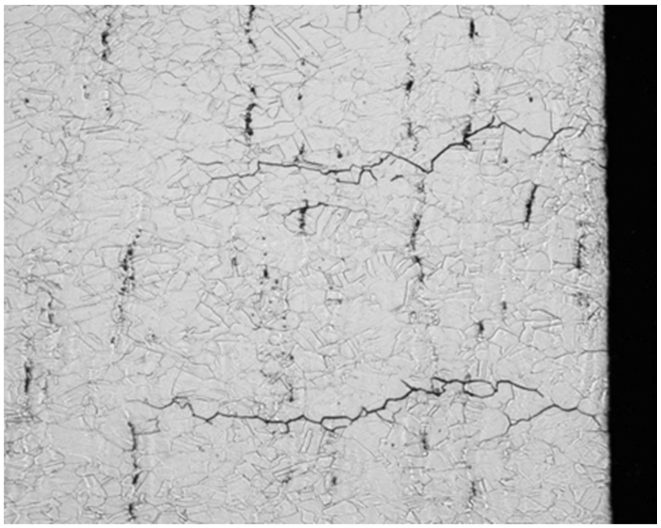
Hydrogen embrittlement of containment materials exacerbated by decay to He-3:

– Use Austenitic SS
– Do not use Ferritic, Martensitic, or Precipitation Hardened SS
– Cleanliness and Surface Coatings Are Important

Permeation Barriers – must maintain integrity with temperature fluctuations:

– Tritium wetted face
  – Additional protection for equipment
  – Manufacturing challenges
  – Must be inert to process stream
– Glovebox face
  – Reduces stripper load
  – Easier to deposit





Intergranular Cracking -characteristic of hydrogen embrittlement

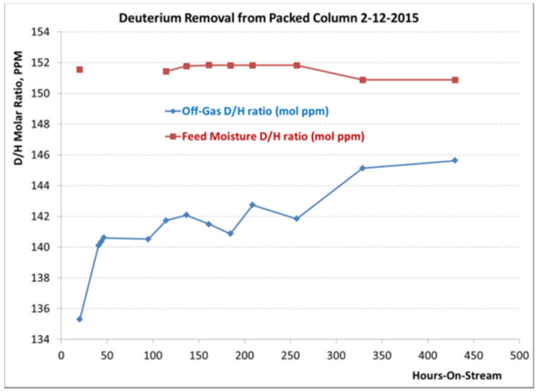
Application of 60+ Years of Tritium Experience to Other Areas

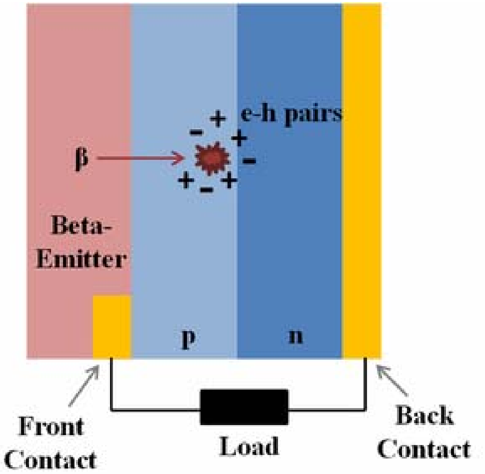
Current Endeavors:

- He-3 recovery from process materials
- Medical isotope production
- Fusion Support
  - ITER
  - LLE

Potential Endeavors:

- Water detritiation
- Betavoltaics
- Electron Capture Detectors
- Analytical capabilities
- T_2 Recovery from breeder materials





SRNL Support for SHINE

SHINE Medical Technologies, Inc. in Monona, WI

- Producer of medical isotopes, primarily Mo-99
- Currently no domestic producers
- NRU in Canada, the only North American producer and single largest producer in the world, is scheduled for shutdown in 2018
- Production technique favorable from a nonproliferation standpoint

SRNL leading design process for tritium system including:

- Accelerator interface
- Impurity removal
- Isotope separation
- Hydrogen storage and delivery
- Shutdown/startup protocols

*Image from SHINE Medical Technologies™ website

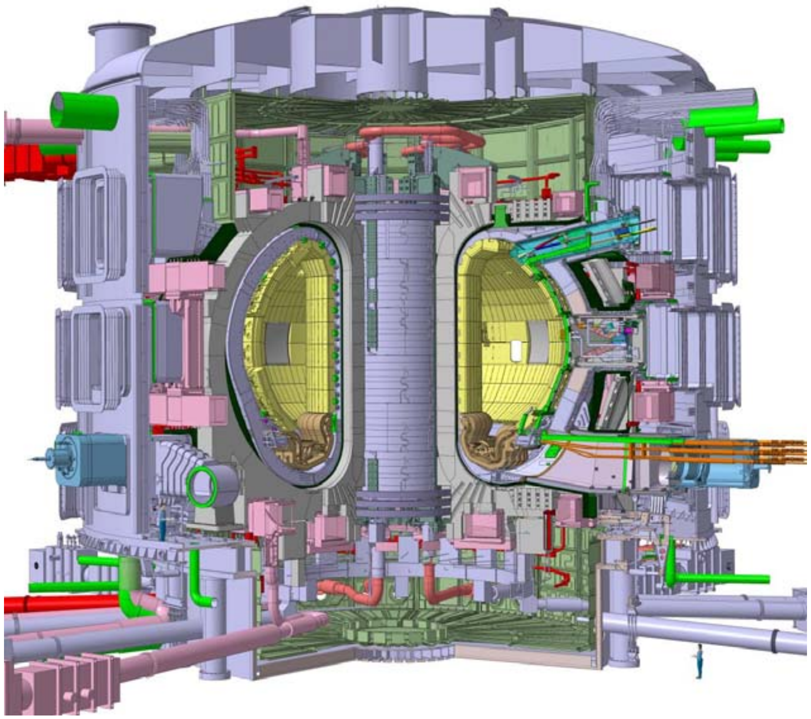
SRNL Support for ITER

– US ITER Project Office

SRNL/SRNS is the Design Authority/Design Agent for the Tokamak Exhaust Processing System which is one of 16 Procurement Packages within the ITER Fuel Cycle

– ITER International Organization Direct Support

- Tritium Analytical System Conceptual Design
- Highly Tritiated Water processing development and design verification
- Tritium Process Control System Conceptual Design
- Tritium Accountability and Tracking Program Development (MC&A)
- Tritium Storage and Delivery System Hydride Storage Bed Design Support



SRNL Support for LLE

Laboratory for Laser Energetics in Rochester, NY

- NNSA Funded for Stockpile Stewardship
- "established in 1970 as a center for the investigation of the interaction of intense radiation with matter"
- Direct drive inertial confinement fusion
- ~1mm DT "ice" target placed within 10 μm of target

The OMEGA EP Laser System bathed in its own light as laser beams ignite during a target shot
*Image from LLE Website

SRNL involvement

- Fabricated micro -TCAP for batch isotope separation
- Provided technical guidance for startup
- Improved test results through better gas control of gas mixtures

Summary

- Tritium Processing is a complex chemical process – containment, separations, unexpected reactions, materials compatibility, and accountability.
- Tritium Processing R&D at SRNL is ongoing to ensure that both:
  - the needs of the Savannah River Tritium Enterprise are met, AND
  - the continuously evolving requirements of tomorrow's fusion machines are met.

Thank You

Backup Slides

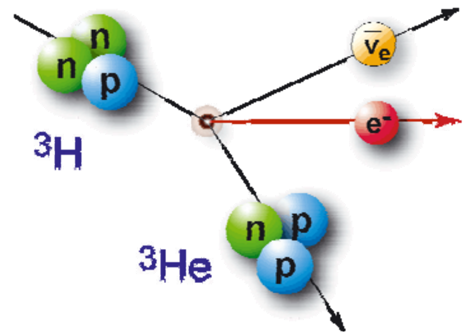
Radiochemistry of Tritium and Impurities (1/2) *

- Self-radiolysis of T_2

$$T_2 \rightarrow T_2^+ + others$$

$$T_2^+ + T_2 \rightarrow T_3^+ + T$$

$$T_3^+ + e^- \rightarrow 3T$$

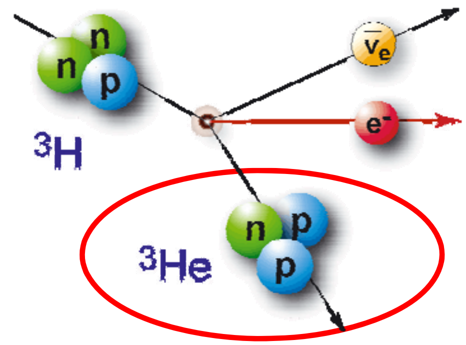


- Self-radiolysis of T_2O (liquid T_2O is self heating)
  - β - radiation causes the formation of T_2O^+ ions, T_3O^+ ions, $T^•$ radicals, $OT^•$ radicals, and T_2O_2 via consecutive chemical reactions
    - *Due to the primary self-radiolysis products of T_2O it is highly corrosive*
  - Gas phase above T_2O becomes pressurized
    - *Radiochemical equilibrium above T_2O at 159 kPa (~1200 torr)*
      - Rate of gas production balanced by rate of (radiochemical) recombination

* Glugla, Fusion Reactor Fuel Cycle INSTN Lecture, February 6, 2009

Radiochemistry of Tritium and Impurities (2/2) *

- Reaction of T_2 with oxygen, nitrogen and air



$$2T_2 + O_2 \rightarrow 2T_2O$$

$$3T_2 + N_2 \rightarrow 2NT_3$$

$$T_2 + air \rightarrow T_2O + T_2O_2 + NO + NO_2$$

$$CO_X + T_2 \rightarrow CT_4 + T_2O$$ as well as other heavier hydrocarbons

  - Reaction products are subject to (self-) radiolysis
  - Radiochemical equilibrium composition depends on tritium concentration (ratio of T/ΣH) and presence of metal catalysts (especially precious metals such as Pt, Pd)

- Reaction of T_2 with methane
  - If deuterium is present 15 different labeled methanes appear in gas phase

$$T_2 + CH_4 \rightarrow CH_3T + CH_2T_2 + CHT_3 + CT_4$$

* Glugla, Fusion Reactor Fuel Cycle INSTN Lecture, February 6, 2009

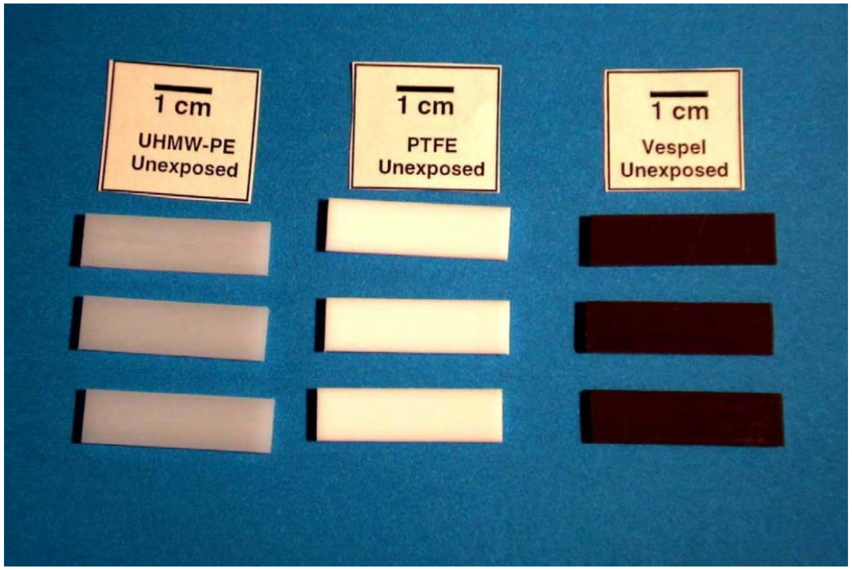
Tritium Effects on Polymeric Materials

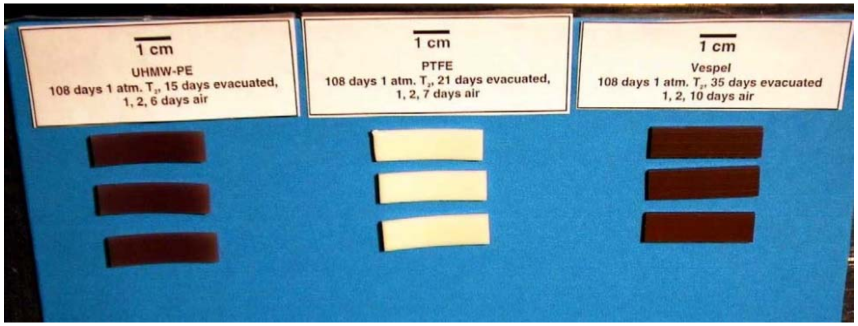
Polymers are a necessary compromise:

- Can't always avoid their use in tritium systems
- Needed for seals, lubrication, packing, etc.

SRNL and others have found that:

- When polymers are exposed to ionizing radiation (for example gamma rays, alpha particles, or beta particles from tritium decay), highly reactive "free radical" groups form when the energy of the ionizing photon or particle is absorbed *
- Polyethylene is known to emit hydrogen, methane and other hydrocarbon gases upon exposure to ionizing radiation *
- Tritium can react with PTFE (Teflon®) to form ^{3}HF (TF) *
- Crosslinked polymers contain sulfur which can be released in sulfur byproducts due to tritium decay – catalyst and permeation membrane poison

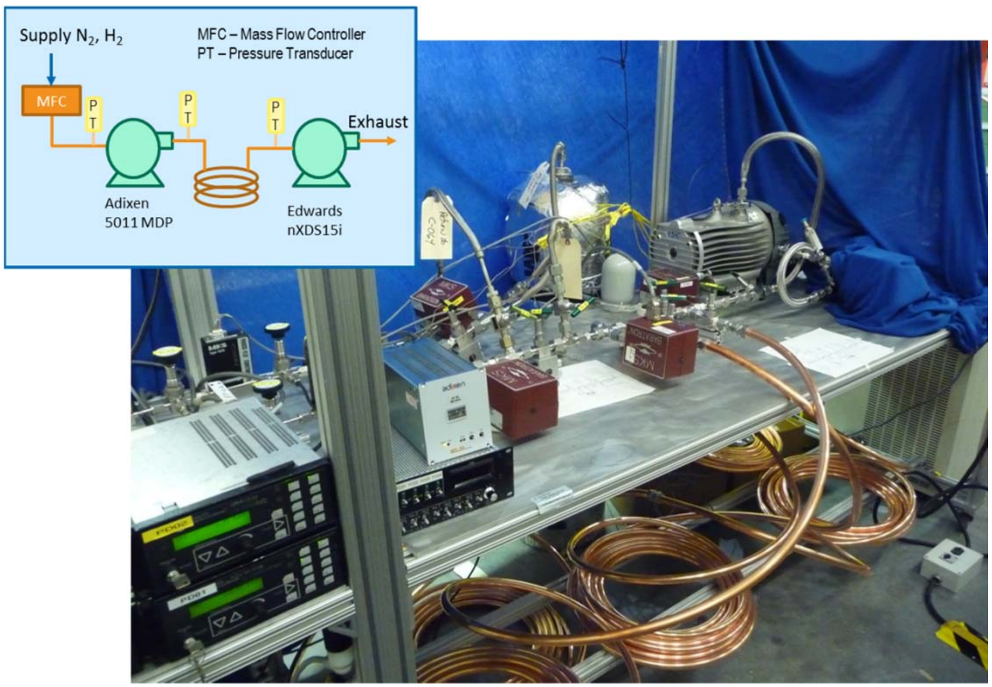




*** Effects of Tritium Exposure on UHMW-PE, PTFE, and Vespel® (U) – Clark and Shanahan – May 2006**

Tritium Fuel Cycle Technologies – Pumping (evacuation)

Evaluation of pumping technologies is necessary to move tritium-containing gases without the addition of impurities (from tritium exposure) – all metal construction is desired.



Candidate Technologies

- Normetex scroll pump and Metal Bellows pump
  - Normetex pumps unavailable
  - Alternate pump testing needed
- "Booster" pump to move gases
- High vacuum pumps (turbomolecular drag pumps)

Metal Bellows Pump MB-601

Edwards nXDS15i Dry Scroll Pump

Normetex (Eumeca) Scroll Pump

Adixen Molecular Drag Pump 5011