Closing the fuel cycle

2020 Introduction to Fusion Energy and Plasma Physics Course

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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

\[ D + T \rightarrow ^4\text{He} + n + 17.6\text{MeV} \quad (1) \]
\[ D + D \rightarrow T + H + 3.98M \quad (2) \]
\[ D + D \rightarrow ^3\text{He} + n + 3.25\text{MeV} \quad (3) \]
\[ T + T \rightarrow ^4\text{He} + 2n + 11.3\text{MeV} \quad (4) \]
\[ D + ^3\text{He} \rightarrow ^4\text{He} + H + 18.3\text{MeV} \quad (5) \]

DT fusion (1) is the most suitable fusion reactions. The D + ^3He reaction is attractive for no neutron production, though accompanying DD reactions do produce it.

The fusion triple product condition for three fusion reactions.

\[ n_e T_E T_e \]

\[ \text{temperature [million Kelvin]} \]

\[ \text{temperature [keV]} \]

\[ 10^{-1} \quad 10^{0} \quad 10^{1} \quad 10^{2} \quad 10^{3} \]

\[ 10^{0} \quad 10^{1} \quad 10^{2} \quad 10^{3} \]
Tritium resource is very limited → need breeding

\[
D + T \rightarrow ^{4}\text{He} (3.5\text{MeV}) + n(14.1\text{MeV})
\]
plasma heating Energy and T 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

\[
\begin{align*}
^6\text{Li} + n & \rightarrow T + ^4\text{He} + 4.8\text{MeV} \\
^7\text{Li} + n & \rightarrow T + ^4\text{He} + n \rightarrow 2.5 \text{ MeV} \\
^9\text{Be} + n & \rightarrow 2n + ^2\text{He} \rightarrow 2.5 \text{ MeV} \\
a\text{Pb} + n & \rightarrow 2n + ^a-1\text{Pb} \rightarrow 7 \text{ MeV}
\end{align*}
\]

• Overall breeding ratio is expected to be above \(~1.1\) (not easy to achieve)
Deuterium

**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:** $5 \times 10^{16}$ kg (in oceans)

**Sufficient for several billion years !!**

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<table>
<thead>
<tr>
<th>Property</th>
<th>D₂O (Heavy water)</th>
<th>H₂O (Light water)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Freezing point (°C)</td>
<td>3.82</td>
<td>0.0</td>
</tr>
<tr>
<td>Boiling point (°C)</td>
<td>101.4</td>
<td>100.0</td>
</tr>
<tr>
<td>Density at STP(g/mL)</td>
<td>1.1056</td>
<td>0.9982</td>
</tr>
<tr>
<td>Temp. of maximum density (°C)</td>
<td>11.6</td>
<td>4.0</td>
</tr>
<tr>
<td>Viscosity (at 20°C, mPa·s)</td>
<td>1.25</td>
<td>1.005</td>
</tr>
<tr>
<td>Surface tension (at 25°C, μJ)</td>
<td>7.193</td>
<td>7.197</td>
</tr>
<tr>
<td>Heat of melting (cal/mol)</td>
<td>1,515</td>
<td>1,436</td>
</tr>
<tr>
<td>Heat of vaporisation (cal/mol)</td>
<td>10,864</td>
<td>10,515</td>
</tr>
</tbody>
</table>

*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, $^6$Li accumulates in the amalgam phase

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

Tritium, one of the three hydrogen isotopes

- Tritium is the radioactive hydrogen isotope
  - decay: $^3\text{T} \rightarrow ^3\text{He} + \beta$ electron + antineutrino
  - 18.6 keV total (average 5.7 keV kinetic, + nearly undetectable antineutrino)
  - decay heat: 324 mW/g
  - half life: 12.32 years (loss ~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)

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

\[
\text{Li}_4\text{SiO}_4 \text{ pebbles (FZK) 0.2 - 0.4 mm}
\]

\[
\text{Li}_2\text{TiO}_3 \text{ pebbles (CEA) 0.6 – 0.8 mm}
\]

\[
\text{LiAlO}_2 \text{ pellet}
\]

\[
\text{PbLi, work at 300-500°C}
\]

\[
\text{Li}_2\text{BeF}_4(\text{Flibe}), \text{ m.p., 459°C}
\]
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

Solid Breeder Blanket utilizes immobile solid lithium ceramic breeder and Be multiplier for tritium self-sufficiency.
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

\[ 2\text{LiT} \rightarrow 2\text{Li}^+ + \text{T}_2 + 2e^- \]
\[ \text{Li}^+ + e^- \rightarrow \text{Li} \]

\[ E^0_{400^\circ C} = -2.82 \text{ V} \]
\[ E^0_{400^\circ C} = -3.22 \text{ V} \]
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%)
Diffusion vs. Permeation

Diffusion

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

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

Molecular kinetics gives incident flux to wall surface under pressure P

\[ J = nv = \frac{P}{\sqrt{2\pi mkT}}; \quad \frac{J_H}{J_D} = \sqrt{2}; \quad \frac{J_H}{J_T} = \sqrt{3}; \quad \frac{J_D}{J_T} = \sqrt{3/2} \]

Permeation

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

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

Permeation flux

\[ F = \phi \frac{\sqrt{P_{up}} - \sqrt{P_{down}}}{\delta} A; \quad \frac{\phi_{HD}}{\phi_{H_2}} = 0.9; \quad \frac{\phi_{HD}}{\phi_{D_2}} = 1.1 \]
Tritium recovery from bulk impurities (e.g., > 95%)

Activated Carbon/Molecular Sieve

Non-Evaporative Getters (NEG)
Isotope separation

- Six species with different boiling points

<table>
<thead>
<tr>
<th>Species</th>
<th>H₂</th>
<th>HD</th>
<th>HT</th>
<th>D₂</th>
<th>DT</th>
<th>T₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Boiling Point, Kelvin</td>
<td>20.7</td>
<td>22.1</td>
<td>23.5</td>
<td>23.8</td>
<td>25</td>
<td>25.5</td>
</tr>
</tbody>
</table>

Cryogenic Distillation

TCAP - Thermal Cycling Absorption Process (palladium has the highest isotopic effect)
TCAP development for isotope separation

Metal Hydride Materials for Hydrogen Storage

Palladium favors lighter hydrogen isotope, especially at low temperatures

Molecular Sieve favors heavier hydrogen isotope (active PFR)

First Tritium Production TCAP 1994

Rochester LLE TCAP - 2014
Hydride bed design for tritium storage

Three Generations of Metal Hydride Storage Bed Development
Example - Storage and Delivery - Jacketed Vessels, Hydride Selection

**Water Detritiation System**

\[
HT(g) + H_2O(l) \rightleftharpoons H_2(g) + HTO(l)
\]

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

**Small CECE Process at AECL**

\[
2H_2O(l) \rightleftharpoons 2H_2(g) + O_2(g)
\]


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

Cryosorption pumps

Metal Foil Pump (MFP)

Vapor diffusion pump

Liquid ring pumps (LRPs)

Snail pump under test at LANL.

discontinued Normetex – complete dry and fluid tight vacuum pump

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

H₂O always exists in H₂

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

Oxidation and reduction on metal surface

\[ MO + H_2 \leftrightarrow M + H_2O - \Delta G \]

\[ \Delta G = -RT \ln K \]

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

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

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: ± 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

MH=Metal Hydrides: Pd, Pd/k, LaN_{5-x}Al_x, Ca_{1-y}Mm_yNi_5, Ti, U

Glovebox T₂ Stripping System
- Gloveboxes
- Catalyst Bed
- Zeolite Bed
- Mg Bed
- To Isotope Separation

MH=MH T₂ & D₂ Storage
- MH Primary Separator
- Pumps
- Diffusers
- Vacuum Pumps

Reservoirs Unloading
- MH Feed Bed
- MH TCAP Isotope Separation

MH T₂ Storage
- MH D₂ Storage

Mix Tanks
- Cylinder

MH Compressors
- He-3 Compressor

Cryosorption

Reservoirs Loading & Sealing
- Newly Loaded Reservoirs

Gloveboxes
- MH Transport Vessels
All tritium processing systems contain the same core fuel cycle functions.
Tritium and environment

1. Cosmic ray
   1-1.3 EBq

\[ ^{14}\text{N} + n \rightarrow ^{3}\text{H} + ^{12}\text{C} \]
\[ ^{16}\text{O} + n \rightarrow ^{3}\text{H} + ^{14}\text{N} \]

Natural T $\approx 2.7$ kg

2. Nuclear bomb
   240 EBq (185-240)

World inventory (2010)
   1-1.3 EBq (1) + 17 EBq (13-17)

3. Consumer products
   0.4 EBq y$^{-1}$
   (0.3-0.4)

3. Nuclear stations
   0.02 EBq y$^{-1}$
   (0.01-0.02)

4. Fusion reactor
   1000MW
   ($\sim$5kg)

EBq $= 10^{18}$

Source: S. Konishi, TRITIUM and Environment, 2010 Tritium Conference Tutorial Lectures, Nara, Japan, 2010.10.29
Useful References for Tritium System Design

- ASME B31.3 process piping code, ASME Boiler and Pressure Vessel Code Section VIII and Section II