ITALIAN NATIONAL AGENCY FOR NEW TECHNOLOGIES, ENERGY AND SUSTAINABLE ECONOMIC DEVELOPMENT

Tritium control and handling

Major design issues

FUNFI-IT - Italian Meeting on Fusion Neutrons for Fission

Centro Ricerche Enrico Fermi, Roma –December 11th 2024

Silvano Tosti – Nuclear Deparment - ENEA

Summary

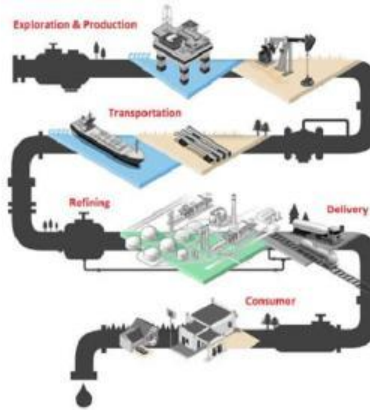
- **Introduction**
- **Tritium processing**
- **Tritium control**
  - **Antipermeation strategies**
  - **Coolant Purification System analysis**
  - **Tritium extraction from BB (Sieverts' constant uncertainties)**

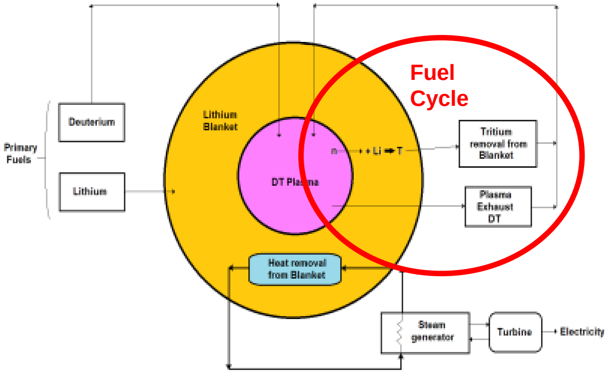
Introduction

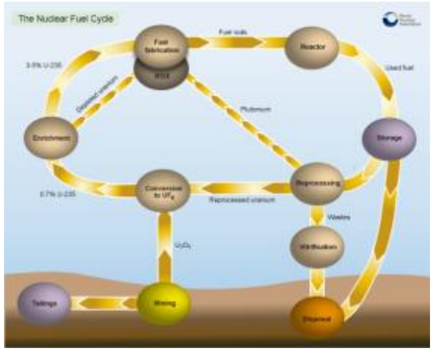
Singularity of the fusion fuel cycle

Neglecting D2 and Li production, all the fuel cycle units are inside the power plant

A fusion plant is inherently safer than other power systems (fission, oil, …)





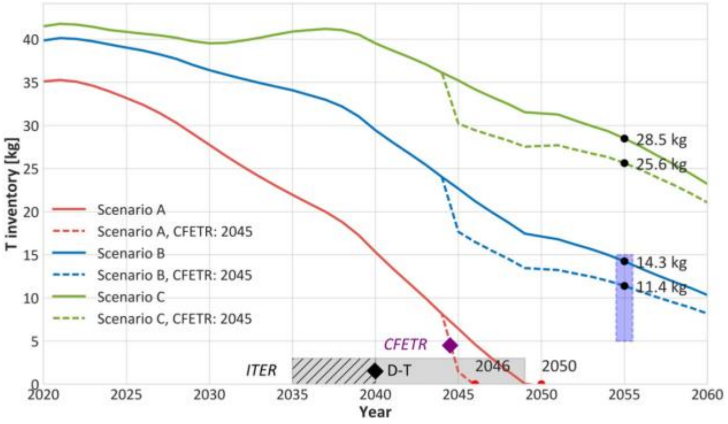


Introduction

Tritium availability for DEMO start-up

Future scenario: a “tritium window” (P. Rutherford) is not open indefinitely. Based on current estimates, it would be open until around 2050, after which it closes quite rapidly (unless the future of the CANDU reactor program turns out much more favorably than could presently be expected)

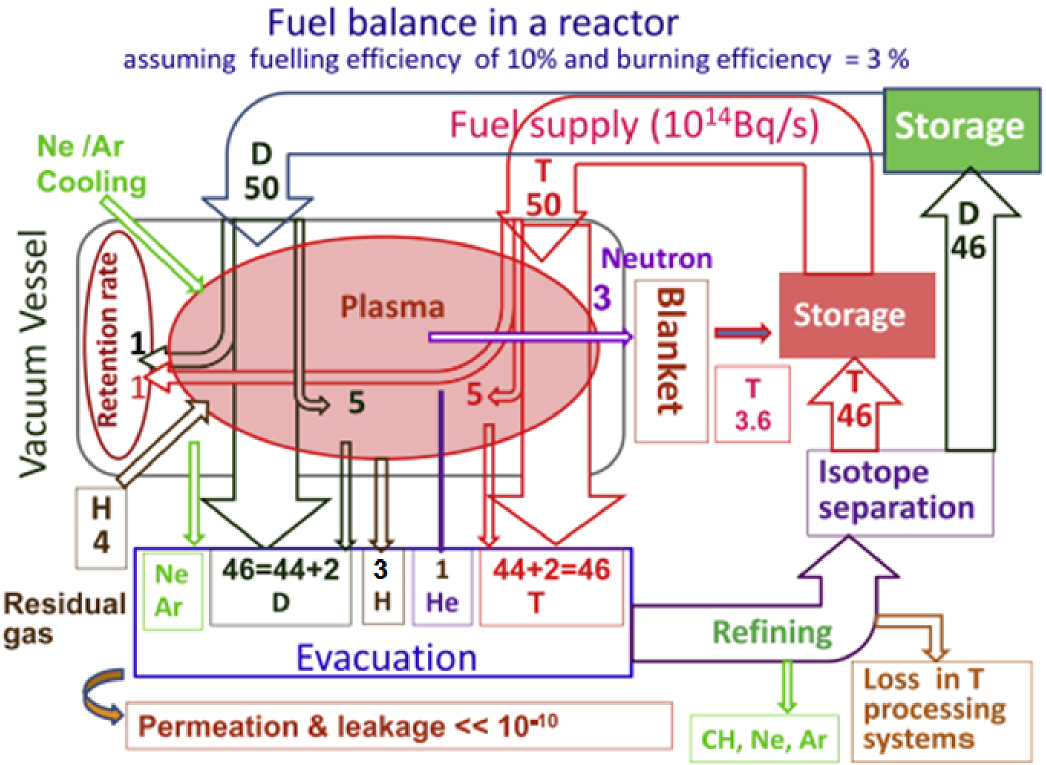
Lack of tritium could be overcome at any time (it can be generated in any fission reactor) but technical, political and economic issues associated could be significant – Alternatives: startup with DD (?)



Total tritium stockpile in Canada, Romania, and Republic of Korea, with ITER D–T operations commencing in 2040 (CFETR consumption is assumed to take place in 2045)

M. Kovari et al., Nucl. Fusion 58 (2018) 026010

Tritium processing



T. Tanabe / Journal of Nuclear Materials 438 (2013) S19–S26

Tritium processing

Tritium processing

Main functions:

- tritium recovery from breeders (liquid or solid)
- tritium recovery from coolant
- other air/water detritiation treatments

Main processes:

- tritium extraction from LiPb (liquid breeder)
- tritium separation from He (purging of solid breeders, coolant purification)
- tritium extraction from tritiated water (coolant purification, water from oxidation of tritiated gases, other detritiation treatments like ADS)

Tritium processing

Processes for tritium extraction from LiPb

Gas Liquid contactor (**GLC**) is a vertical column where a liquid and a gas phase are put in contact by flowing counter-currently:
- spray columns
- bubble columns
- packed columns

Permeator Against Vacuum (PAV)
Dense metal membrane devices

Membrane Gas Liquid Contactor (MGLC)
Porous metal membranes (a combination of a GLC and a PAV)

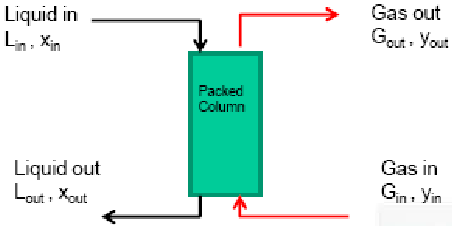
Tritium processing

Packed column

Among the different possible technologies proposed for TES, a gas liquid contactor (GLC) is the reference solution for the tritium extraction unit. However, also the option of the permeator is considered.

The packed columns are vertical columns filled with packing or other device providing a large interfacial surface between liquid and gas phase in both counter-current and co-current flow.

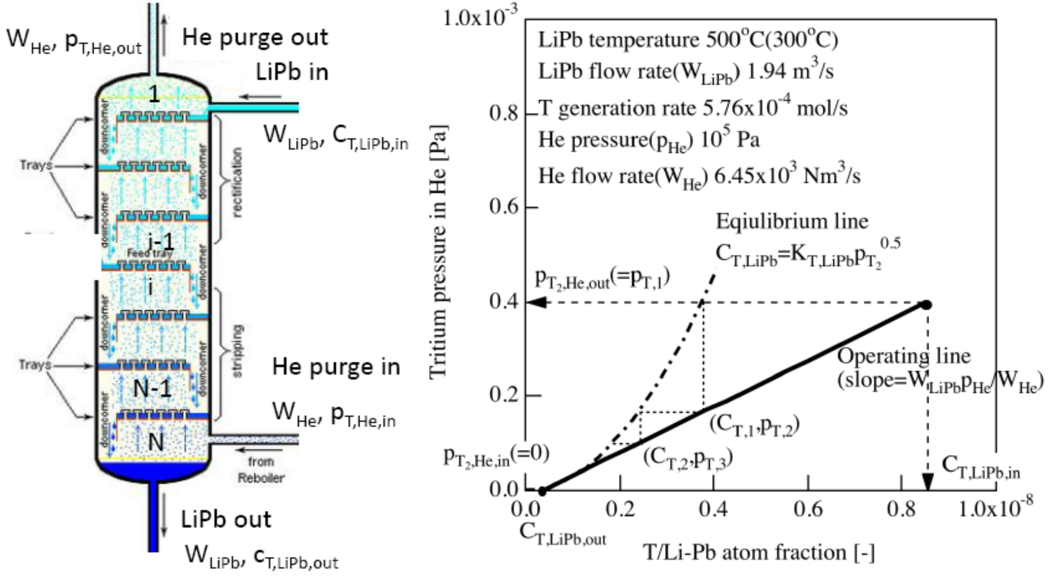
Packed columns for HCLL TEU contain the metal filler Mellapak 750Y. TEU has variable operational temperature up to 450° C.



A.Ciampichetti, Tritium Technologies of ITER and DEMO breeding blankets, Current research topics in Nuclear Fusion Engineering, Politecnico di Torino 18-01-2010

Tritium processing

T extraction using counter-current LiPb-He flow



T recovery ratio of 0.95

Since the equilibrium line is concave, LiPb-He counter-current operation is not efficient.
T concentration in LiPb and He is very low, the He flow rate is very large.

S. Fukada et al., Modeling of tritium release and permeation from LiPb blanket and design of tritium and heat recovery system for fusion reactors, MFE/IFE system modeling Workshop, Jan 31- Feb 1, 2011, Idaho Falls

Tritium processing

- **Bubble columns:** the experiments at “Melodie loop” (France) demonstrated low tritium extraction efficiency (due to small liquid-gas contact area)

- Packed columns: it was demonstrated that H= 800 mm at 673 K (high gas-liquid contact area 750 m^2 m^{-3}) attain high efficiency (close to 30%)

Table 10: Results of the tests on packed column [59].

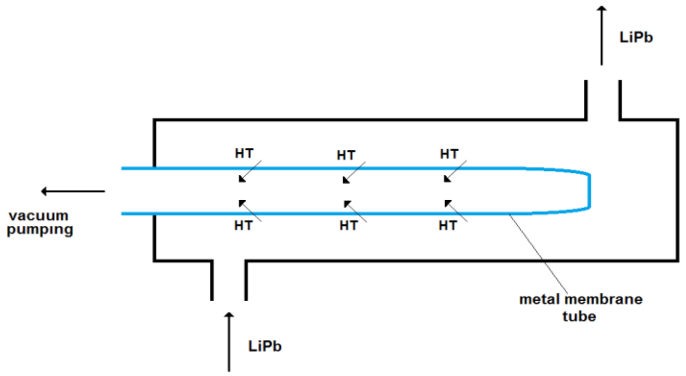
Test n.	LM flow-rate ($L \cdot h^{-1}$)	Ar flow-rate ($NL \cdot h^{-1}$)	P_{H2} (Pa)	η (%)
1	70-90	6	1200-1350	20-22
2	30-50	6	1000-1100	29-31
3	30-50	30	975-1000	29-31
4	30-50	6	450-475	23-25
5	30-50	6	220-230	23-25

N. Alpy et al., “Hydrogen extraction from Pb-17Li: results with 800 mm high packed column” Fus. Eng. Des. 49-50 (2000) 775-780

Tritium processing

Permeator against vacuum (PAV)

- - candidate materials (high T diffusivity, low solubility): α-Fe, ferritic-martensitic steel, V and V-alloys, Nb, Ta
- - expected efficiency of α-Fe membranes: 90% at 973 K and LiPb velocity of 0.1 m s^{-1}

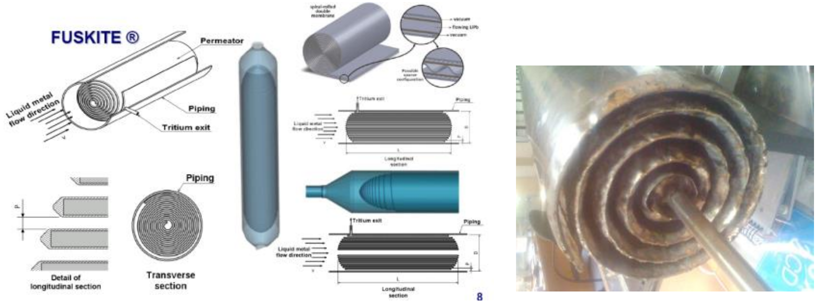


Tritium processing

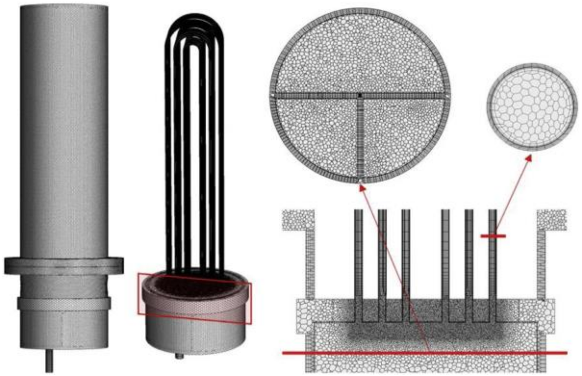
PAV issues

- complicate structure
- low stability (erosion-corrosion, oxidation, etc.)
- Low permeability or embrittlement

α-Fe has low hydrogen permeability (7,99 10^{-11} mol m^{-2} s^{-1} $Pa^{-0.5}$ at 400 °C), refractory metals (i.e., Nb and V) exhibit hydrogen permeability higher than Pd-alloys (e.g., 3,59 10^{-6} and 3,40 10^{-7} mol m^{-2} s^{-1} $Pa^{-0.5}$ at 400 °C for the Nb and V, respectively), but are characterized by high hydrogen solubility then involving low durability (embrittlement) and safety concerns (high tritium inventory).



I. Martínez et al., A demonstrator of a PAV for tritium recovery from LLE at HCLL TBM loop operational ranges, IEA International Workshop on Liquid Metal Breeder Blankets, 23-24 September, 2010 (Madrid), Spain

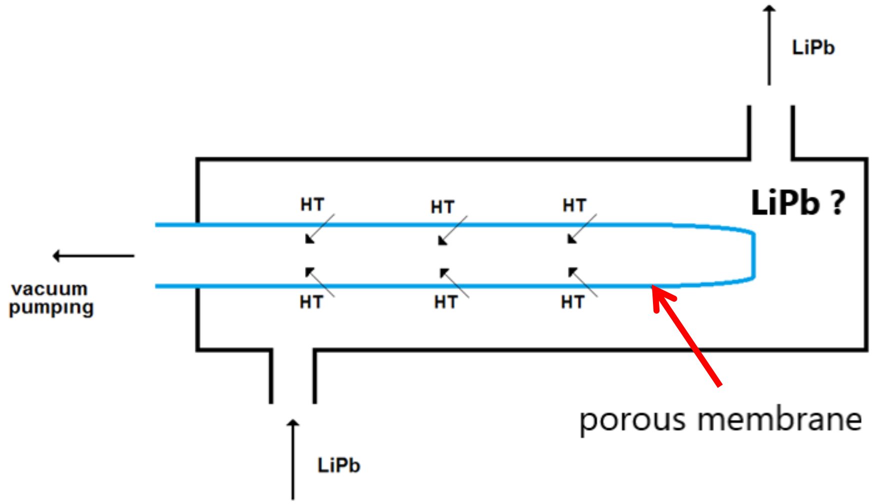


F. Papa et al., Engineering design of a Permeator Against Vacuum mock-up with niobium membrane, Fusion Engineering and Design 166, May 2021, 112313

Tritium processing

Membrane Gas Liquid Contactor (MGLC)

What happens if we use a porous membrane in the PAV?



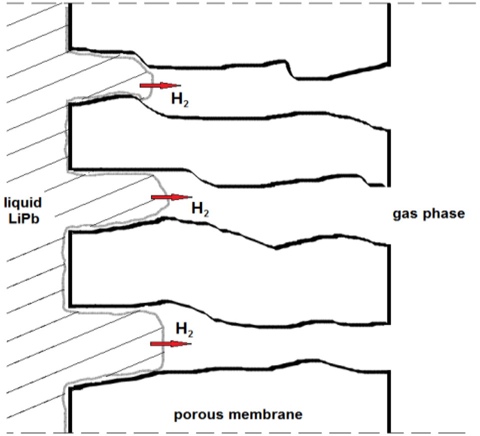
Tritium processing

A porous membrane is not wetted by liquid LiPb when its pores size (D) is given by the Washburn equation:

$$P_L - P_G = -\frac{4\,\gamma \cos\theta}{D}$$

Pore size of about 3-7 µm

is assessed for the extraction of tritium from liquid LiPb at 300-500 °C and 200-400 kPa

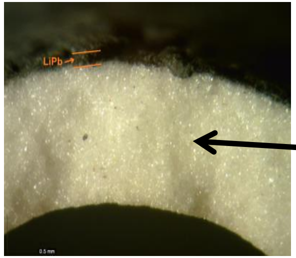


Hydrogen isotopes mass transfer takes place through the interface between the liquid LiPb and the gas phase inside the membrane pores

Advantages of MGLC vs. PAV

- better resistance to the erosion-corrosion of the LiPb flow at high temperature (e.g., use of ceramic membrane)

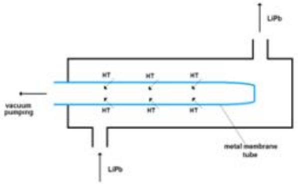
- reduced mass transfer resistance (absence of the metal wall)



Alumina tubular membrane, pore size 10 µm immersed in liquid LiPb at 450 °C

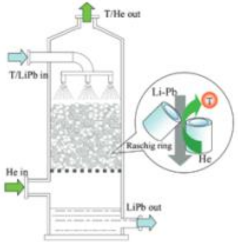
Tritium processing

PAV

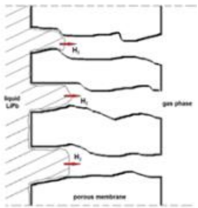


+

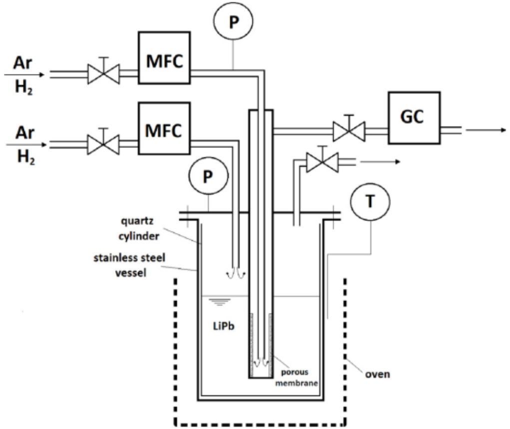
GLC

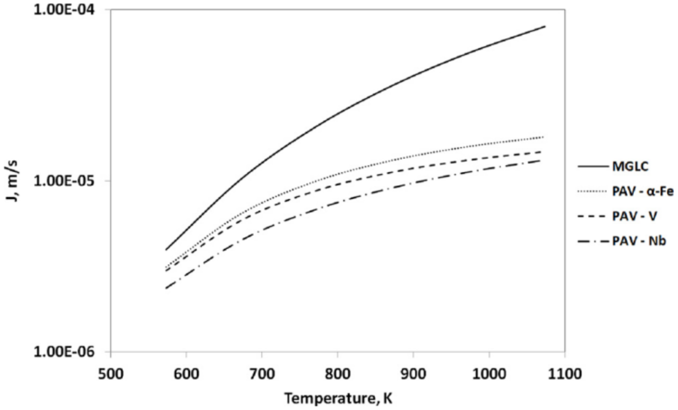


MGLC



"Processo a supporto poroso per l'estrazione di idrogeno ed isotopi da metalli liquidi, e relativo apparato"
Italian Patent Application 102018000003185 (2018)





Comparison of calculated overall mass transfer coefficient between PAV and MGLC systems.

S. Tosti et al., Membrane gas-liquid contactor for tritium extraction from Pb-Li alloys, Fusion Eng. Des. 158 (2020), 111737.

Tritium processing

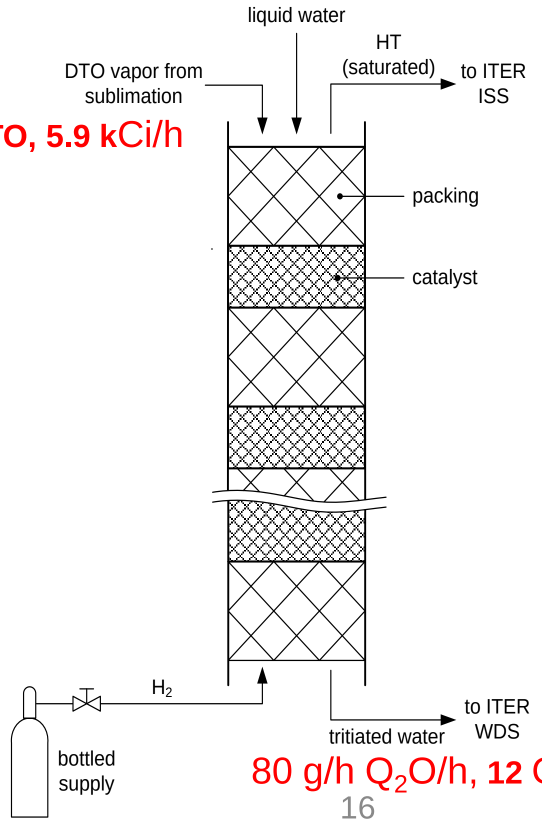
Liquid Phase Catalytic Exchange (LPCE) for ITER WD

Outline conceptual design
4,2 g/h (100 g/day) DTO vapor flow rate
72 g/h H_2O liquid water feed flow rate (mixing factor 20)
Water for mixing could be tritium contaminated
Moisture in HT to be condensed and returned
48 g/h H_2 flow rate (molar ratio 6)
Trade off to mixing factor, column length, outlet concentration
H_2 to be added could also be slightly contaminated
80 g/h (4.2 mol/h) tritiated water flow rate at 150 Ci/kg
to Water Detritiation System (capacity > 20 kg/h @ 10 Ci/kg)
Column height about 4 m, column diameter about 3 cm
Upper section of the column to be easily replaceable
Catalyst lifetime could be limited due to
high tritium concentration (no problem for VPCE)

$$DTO + H_2 = H_2O + DT$$

72 g/h H2O

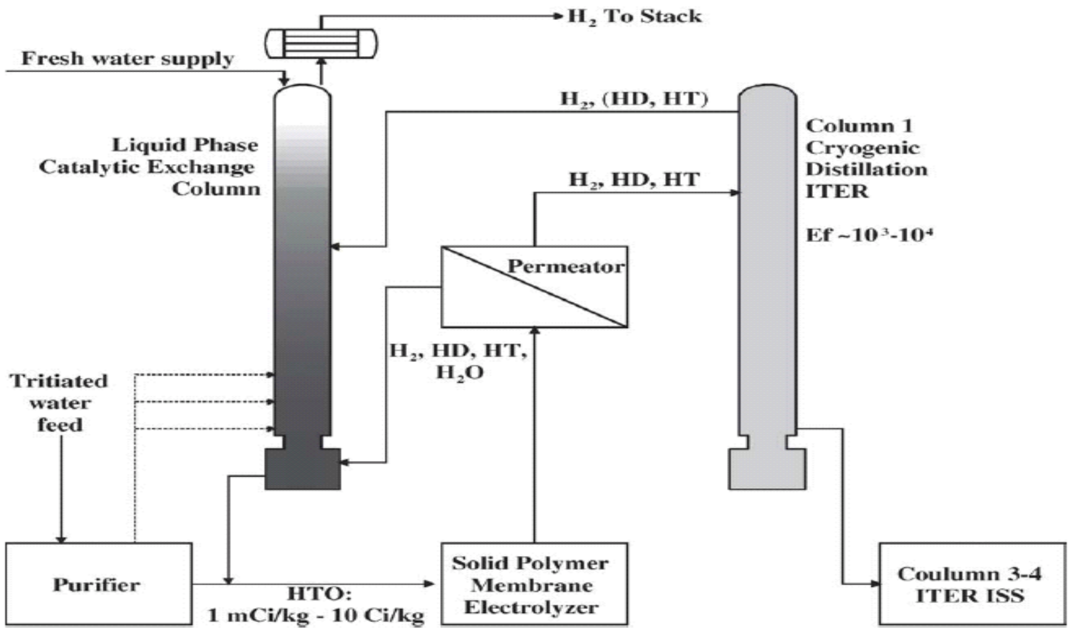
4.2 g/h DTO, 5.9 kCi/h



80 g/h Q_2O/h, 12 Ci/h

Tritium processing

Combined Electrolysis Catalytic Exchange (CECE)

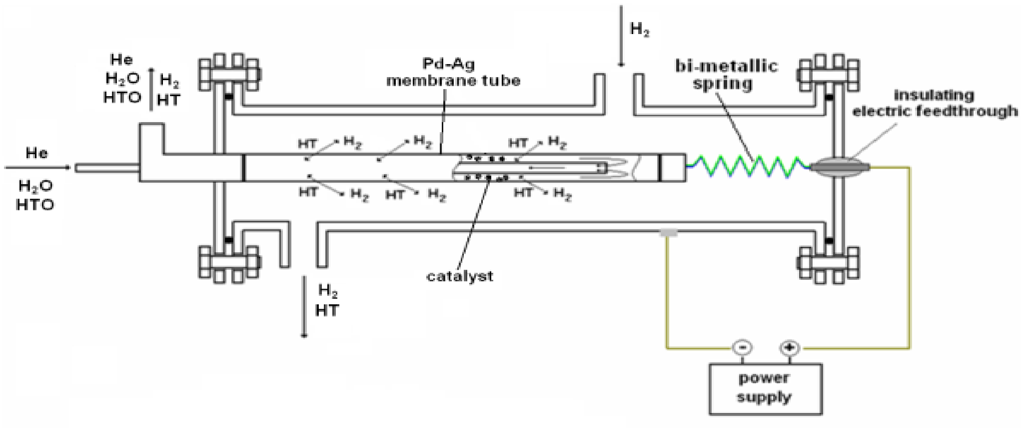


M. Glugla, ITER fuel cycle R&D: Consequences for the design,Fusion Engineering and Design 81 (2006) 733–744

Tritium processing

Pd-based membrane reactor

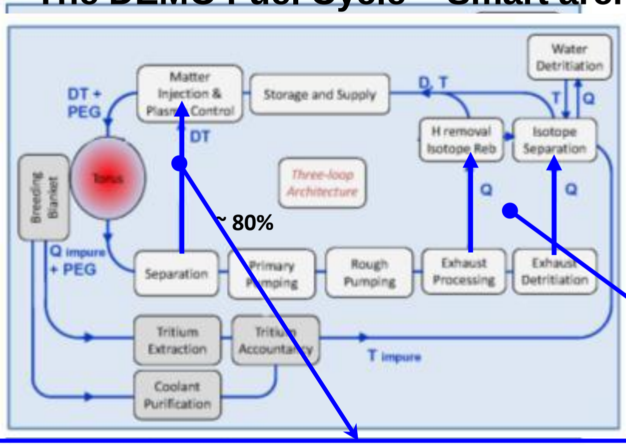
"Reattore a membrana per il trattamento di gas contenti trizio"
Italian Patent n. RM2010A000330 (16.06.2010)

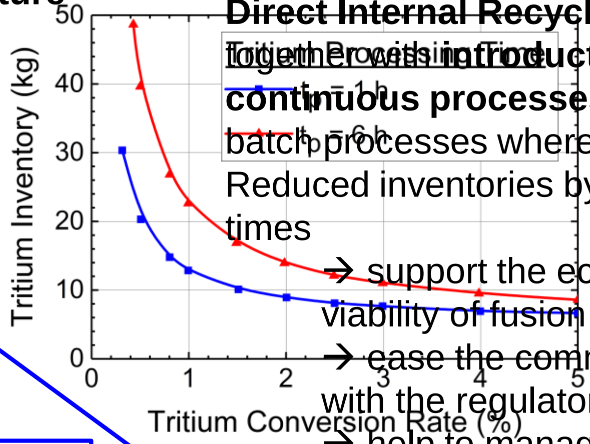


S. Tosti et al., Design of Pd-based membrane reactor for gas detritiation, Fusion Engineering and Design (2010) 10.1016/j.fusengdes.2010.11.021

Tritium processing

The DEMO Fuel Cycle – 1st Try
The DEMO Fuel Cycle – Smart architecture





Direct Internal Recycling (DIR)

Together with **introduction of continuous processes** to replace batch processes wherever possible. Reduced inventories by lower cycle times

→ support the economic viability of fusion

→ ease the communication with the regulator

→ help to manage safety requirements

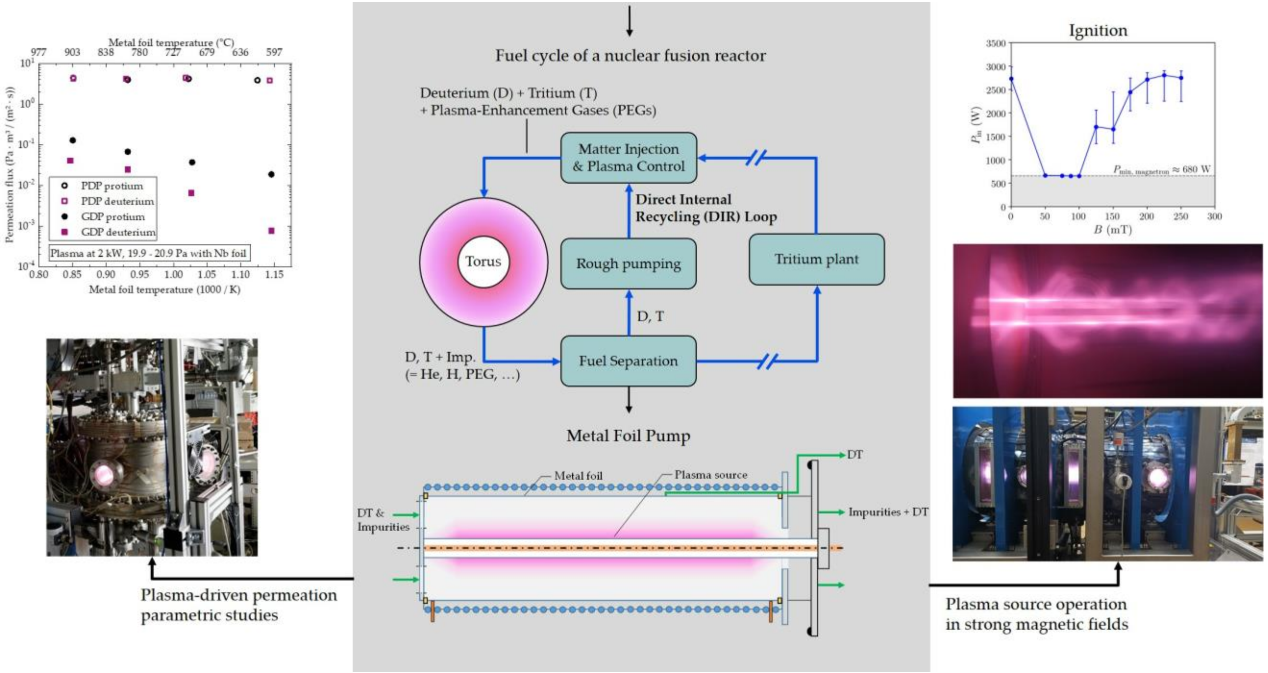
Direct Internal Recycling (DIR) and less start-up inventory potentially reduces the TBR requirements
DIR potentially reduces the footprint of the tritium plant
→ DIR requires to develop first-of-its-kind technologies: Metal Foil Pump (MFP) at HERMESplus (KIT)

- Showstopper: Inventories are excessive for the achievable numbers of the tritium conversion rate
- Approach: **Smart** architecture to reduce inventory.

Isotope Re-balancing based on use of materials with opposite isotope effect (experiments started, KIT)

Tritium processing

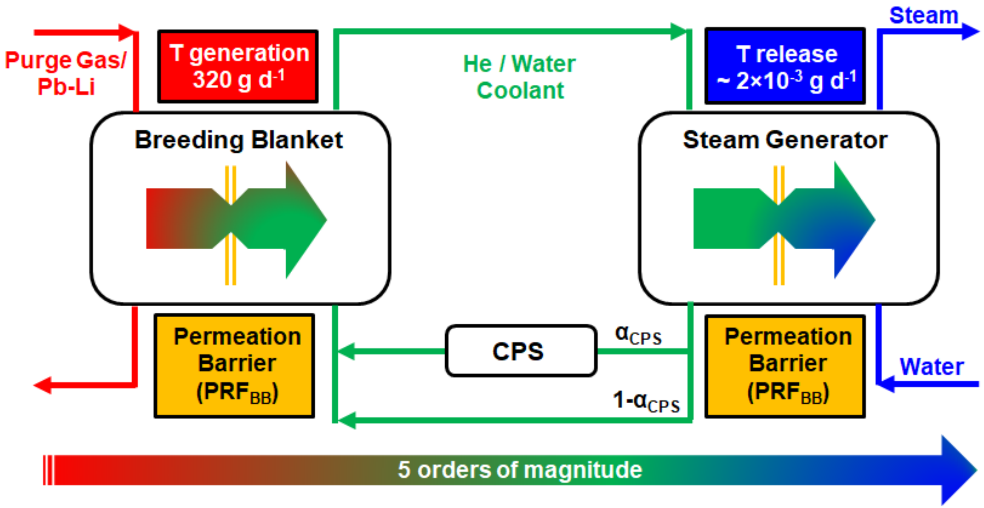
Metal Foil Pump



Kathage, Y.; Vazquez Cortes, A.; Merli, S.; Day, C.; Giegerich, T.; Hanke, S.; Igitkhanov, J.; Schulz, A.; Walker, M. Experimental Progress in the Development of a Metal Foil Pump for DEMO. *Plasma* **2023**, *6*, 714-734

Tritium control

How effective should tritium control be?
T generation = 320 g/d ⬄ T release = 2 x 10^{-3} g/d
5 orders of magnitude!!



(from: A. Santucci – ENEA)

Tritium control

Breeding Blanket converts neutron energy into heat and collect efficiently it by coolant

Colburn theory describes a perfect **analogy in the transport phenomena** of **heat, momentum, and mass transfer**. The basic mechanisms and mathematics of heat, mass, and momentum transport are essentially the same (same formulas, parameters, etc.).

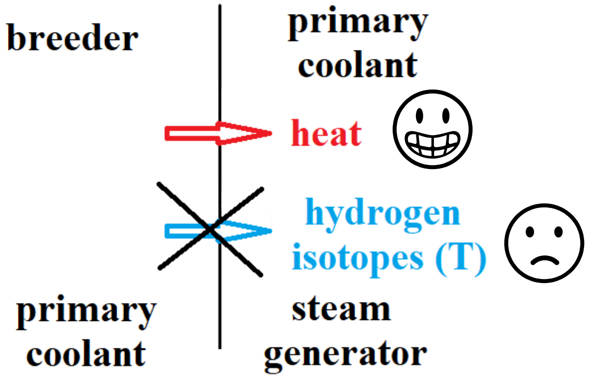
Main design issue for BB design: maximize heat transfer ⬄ minimize tritium permeation

From breeder to primary coolant and from primary coolant to steam generator: two transport phenomena take place in parallel:

- **heat transfer (to be maximized)**
- **tritium mass transfer (to be minimized)**

I.e. => the main design parameters affects the two transport phenomena in the same way:

- **temperature, pressure**
- **thickness**
- **geometry (surface areas, …)**



Tritium control

Antipermeation strategies

- Use of coatings
- Control of (gas/water) coolant chemistry
- Combination of coatings and coolant chemistry

Key aspects under study:

- Development of coatings (several activities on-going, at KIT and ENEA Bra/IIT): performances (permeation reduction, effect on heat transfer) and stability
- Proper design of Coolant Purification System (CPS)

Open issues:

- Verify manufacturability and costs of coatings
- (before ITER TBM) > Need of test beds for exp. of coatings under reactor-relevant conditions (combined effect of irradiation, purge gas chemistry, etc.)
- Cross-cutting aspects: modify coolant chemistry could produce corrosion and activation products (safety concerns)

Tritium control

Antipermeation strategies – Al-based coatings

- **Al2O3 coating developed by Pulsed Laser Deposition (PLD) (IIT-ENEA)**

In the *PLD* a high-power pulsed laser beam is focused inside a vacuum chamber to strike a target of the material (EUROFER). Al is vaporized from the target and as result, a high homogeneous layer of alumina is deposited.

- **Al2O3 coating developed by Atomic Layer Deposition (IIT-ENEA)**

- Chemical Vapor Deposition (CVD)
- Growth at the atomic scale levels
- Control through self-limited reactions

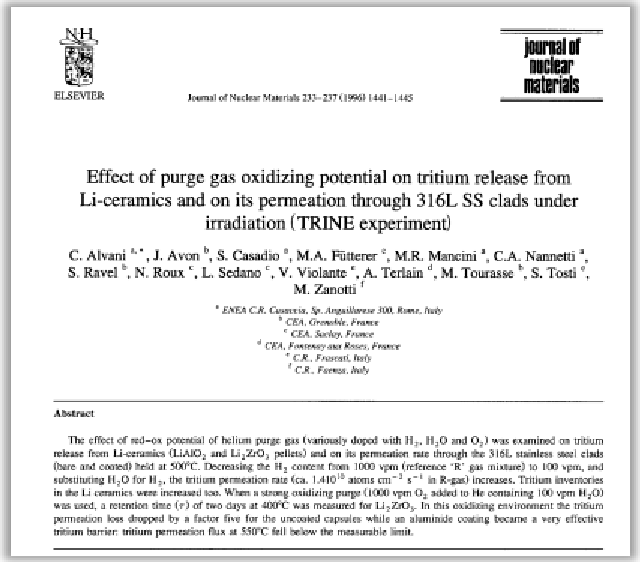
- **Al-based corrosion barriers - Electrochemical ECX process (KIT)**

The ECX process is based on the electrodeposition of aluminum from an ionic liquid for the fabrication of Fe-Al barriers. Al is diffused gradually into the Eurofer.

Tritium control

Antipermeation strategies - TRINE experiment

Combination of <u>coatings</u> and <u>purge gas chemistry</u> (He addition with H_2, H_2O and O_2)



ELSEVIER

Journal of Nuclear Materials 233–237 (1996) 1441–1445

journal of nuclear materials

Effect of purge gas oxidizing potential on tritium release from Li-ceramics and on its permeation through 316L SS clads under irradiation (TRINE experiment)

C. Alvani [a,*], J. Avon [b], S. Casadio [a], M.A. Fütterer [c], M.R. Mancini [a], C.A. Nannetti [a], S. Ravel [b], N. Roux [c], L. Sedano [c], V. Violante [e], A. Terlain [d], M. Tourasse [b], S. Tosti [e], M. Zanotti [f]

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Abstract

The effect of red–ox potential of helium purge gas (variously doped with H_2, H_2O and O_2) was examined on tritium release from Li-ceramics ($LiAlO_2$ and Li_2ZrO_3 pellets) and on its permeation rate through the 316L stainless steel clads (bare and coated) held at 500°C. Decreasing the H_2 content from 1000 vpm (reference 'R' gas mixture) to 100 vpm, and substituting H_2O for H_2, the tritium permeation rate (ca. $1.4 10^{10}$ atoms cm^{-2} s^{-1} in R-gas) increases. Tritium inventories in the Li ceramics were increased too. When a strong oxidizing purge (1000 vpm O_2 added to He containing 100 vpm H_2O) was used, a retention time (τ) of two days at 400°C was measured for Li_2ZrO_3. In this oxidizing environment the tritium permeation loss dropped by a factor five for the uncoated capsules while an aluminide coating became a very effective tritium barrier: tritium permeation flux at 550°C fell below the measurable limit.

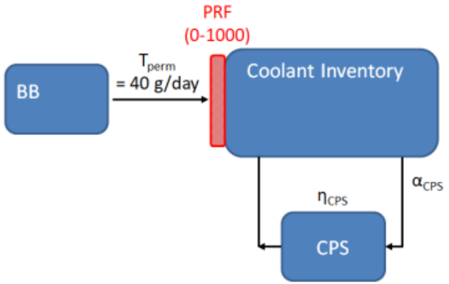
<u>Li-ceramics exposed in thermal reactor</u> (SILOE, Grenoble) for studying the <u>effect of gas purge chemistry</u> on both:
- <u>tritium release</u> from breeder
- <u>tritium permeation</u> through stainless steel (both uncoated and with aluminide coatings)

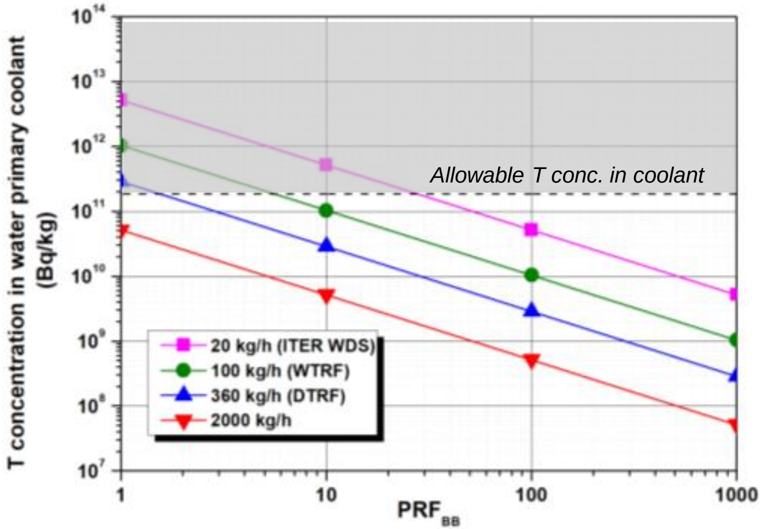
Results:
- no significant effect on T permeation of H_2 and water addition into He purge
- <u>effectiveness of O_2 addition</u> to He purge to reduce permeation (by <u>3 orders of magnitude</u>) due to "self-healing" of cracks in aluminide layers in presence of oxidizing atmosphere

Tritium control

Coolant Purification System Analysis

A fraction of the coolant (α_{CPS}) is routed inside the CPS





Allowable Tritium concentration in water coolant based on CANDU experience between 5-10 Ci kg^{-1}

1.85×10^{11} Bq kg^{-1} (or 5 Ci kg^{-1})

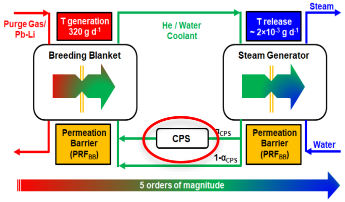
PRF_{BB}	CPS requirement	T conc. in coolant, Bq kg^{-1}	T inventory in coolant, g
1	100 × ITER WDS	5.15×10^{10}	~ 30
10	Wolsong	1.03×10^{11}	~ 60
100	ITER WDS	5.15×10^{10}	~ 30

(from: A. Santucci – ENEA)

Tritium control

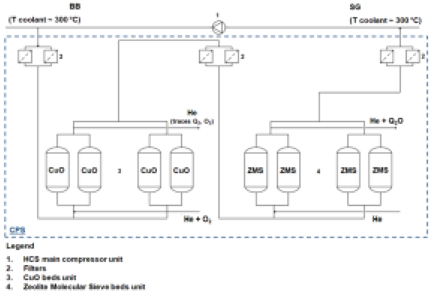
Coolant Purification System Analysis

The Coolant Purification System (**CPS**) unit treats a certain by-pass (α_{CPS}) of the primary coolant with the aim to extract the permeated Tritium



He-CPS

1. Scale-up of the ITER reference process (CuO and MS beds)



2. New process based on novel getter material (ZAO alloy)

Water-CPS

"standard" Water Detritiation Processes

Facility	Front-end process	Flowrate, kg/h	Operation
JET WDS	Direct electrolysis	3.7 – 5.6	Off-line
ITER WDS	Combined Electrolysis and Catalytic Exchange	20	Off-line
CTRF (Cernavoda)	Liquid Phase Catalytic Exchange	40	Off-line
WTRF (Wolsong)	Liquid Phase Catalytic Exchange	100	Off-line
DTRF (Darlington)	Vapor Phase Catalytic Exchange	360	Off-line

- On-line: a Certain by-pass is continuously routed inside the CPS

- Off-line: the entire water primary coolant is discharged after one year of DEMO operation and processed in a dedicated off-line facility

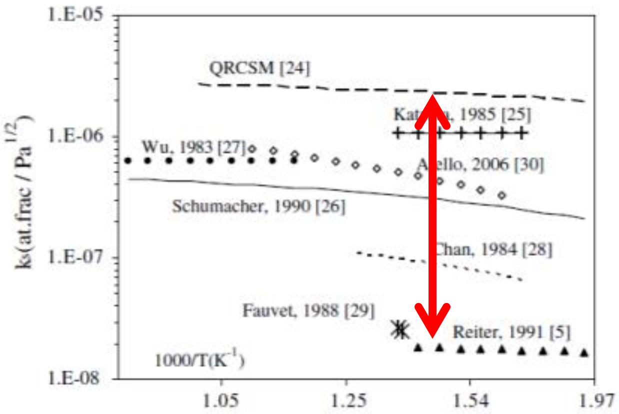
(from: A. Santucci – ENEA)

Tritium control

Tritium extraction from BB: Large uncertainty in the measurements of Sieverts' constant

This an open issue for both: i) the assessment of tritium inventory assessment in BB, and ii) the design LiPb extraction systems

More than 2 orders of magnitude for measured values cannot be explained by the different measurement techniques adopted (adsorption, desorption, etc.) and different operating conditions (T, P, vacuum, etc.)



E. Mas de les Valls et al., Journal of Nuclear Materials 376 (2008) 353–357

Possibly, high reactivity of Li involves "uncontrolled" formation of Li-compounds (e.g. oxides and hydroxides in presence of air/humidity in traces) and other impurities that change significantly the composition and behaviour of the hydrogenated Li-alloy

- Need of analysing chemically LiPb before and after experiments
- Need of experiments with liquid LiPb under operating conditions as close as possible to real systems (e.g. large test-beds with LiPb loops)
- New testing campaigns for measuring the Sieverts' constant at CIEMAT, KIT and ENEA Brasimone

ENEA

Conclusions

A specific characteristics of the fusion fuel cycle: all process units are inside the power plant reducing environmental impact and increasing the safety

Main open issues:

- Limited tritium availability for DEMO start-up: a “tritium window” would be open until around 2050

- Tritium processing: need of new approaches for inventory minimization (smart T processing architecture, DIR), control of permeation into coolant, extraction from LiPb, uncertainties on Sieverts’ constant of LiPb

Technologies under development: metal foil pump, anti-permeation coatings, coolant chemistry, PAV, MGLC

Thank you for your attention

Any questions ?

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