

Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile

# Tritium migration pathway and role of the coolant purification system

Technical meeting on Plasma Physics and Technology Aspects of the Tritium Fuel Cycle for Fusion Energy

13 October 2022

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# **Basics on permeation**

### *<u>Hydrogen permeation through dense</u>* <u>*metal*</u> is a serial process which includes several steps:

- Diffusion of Q<sub>2</sub> molecules
- Physical adsorption of molecules on high pressure side
- Dissociation into metal surface
- Atomic diffusion by concentration gradient
- Recombination on metal surface
- Molecular desorption in gas phase

Q = H, D, T

- Diffusion of Q<sub>2</sub> molecules

#### Materials

- Surface condition (i.e., oxide layer)
- Bulk defects (i.e., vacancies, interstitials, impurities, etc.)



- Diffusion + convection (advection)
- Liquid metal (LiPb) + magnetic field → Magneto Hydrodynamic (MHD) effect



#### Others

- Chemical equilibrium btw. Q<sub>2</sub> and other components
- Co-permeation (or back permeation)
  - $\rightarrow$  non-linearity in the diffusion gradient

## **Basics on permeation**

Depending on the complexity and the extension of the problem, the permeation models can reach different levels of details



- Accurate description of the permeation phenomemon (physics and geometry)
  - Limited extension

Output: tritium concentration profile to support the blanket design



## System Level



 Less accurate description of the permeation problem

□ Very large extension

Output: tritium inventory and release (permeation and leaks) to support the design of several systems

## **Basics on permeation**



Permeation is driven by the slowest phenomenon

<u>Surface-limited</u>: the permeation rate depends on the dissociation and recombination constants ( $\sigma$ k) and is proportional to  $p^1$ 

<u>Diffusion-limited</u>: the permeation rate depends on solubility (S) and diffusivity (D) inside the bulk and is proportional to  $p^{0.5}$ 

- "Database" (scientific literature) of hydrogen transport parameters for breeder blanket and steam generator functional materials (RAFM = Reduced Activation Ferritic Martensitic Steel, like Eurofer or Incoloy)
- ✓ Good agreement among the hydrogen transport parameters reported in literature
- Missing data in "real" conditions (i.e., surface status, very low hydrogen pressure, etc.)

P <sub>H</sub> R <sub>J</sub>			
Unified	permeation models		
(σk,D,S	) proportional to $p^n$		
Table 1: Overview of	f the most relevant & recent papers on hydrogen permeation		
First Author (Year)	Interest - Main outcome		
Calderoni (2011) [3]	Measurement of permeability on <u>Incoloy</u> for VHTR applications with H <sub>2</sub> diluted in He (tritium tests are also oncoince)		
Esteban (2009) [4]	Measurement of permeation, diffusion, solubility and trapping in <u>Glicop AI25</u> (ODS RAFM Cu rich alloy) and comparison with other Cu and Cu rich alloys, showing higher trapping due to ODS and alumina nanoparticules, but lower		
Esteban (2007) [5]	permeasures compared to other KAFM Measurement of permeation, diffusion, solubility and trapping in <u>Eurofer</u> and comprehensive comparison with literature		
Takeda (2004) [1]	Measurements for <u>Hasteloy</u> with H and D, comprehensive comparision with literature, for application to HTR		
Esteban (2002) [6]	Measurement of permeation, diffusion, solubility and surface rate constant for <u>Incoloy 800</u> with bare or oxidised surfaces, showing effective barrier with oxide layer		
Esteban (2002) [7]	Measurement of permeation, diffusion, solubility and trapping in <u>Optifer</u> and study of isotope effect with H and D		
Esteban (2000) [8]	Measurement of permeability, solubility and diffusivity for <u>Optifer</u> with deuterium, and isotope extrapolation given full data for H, D, and T, comparison with other RAFM materials		
Esteban (2000) [9]	Discussion of permeation regimes (surface-limited / diffusion-limited / intermediate), measurement of surface rate constant for Optifer with deuterium		
Perujo (1997) [10]	Discussion of different measurement approaches (gas evolution or permeation), measurement of permeability, solubility and diffusivity for MANET III with H and D, discussion on oxidation of surfaces, measurement of surface constants with slightly oxidiated surfaces		
Perujo (1996) [11]	Measurement of surface rate constant for <u>Inconel</u> and <u>316L</u> with tritium at low pressure (below 1 Pa), reference to other studies showing surface-limited permeation regime		
Serra (1995) [12]	Theory / model of surface-limited regime, and measurement of surface rate constant for <u>Manet</u> , experiments show curface limited behaviour up to 104 Pa		
Röhrig (1975) [2]	Review of high temperature alloys permeability values, and measurement over large pressure range, also studying water vapour effect		

## Outline

- □ The problem of Tritium migration in DEMO
- □ EDFA tasks (2012-2014)
  - □ FUS-TPC code
- □ EUROfusion (FP8) activities (2014-2018)
  - □ EcosimPro
  - Development of mitigation strategy to the Tritium permeation
    - □ Anti-permeation barriers
    - □ Coolant Purification System (CPS)
- □ KDII#2 activity (2018-2020)
  - Main results of the sensitivity analysis
- Conclusions



# The problem of Tritium migration in DEMO

Two key requirements for DEMO are:

- The demonstration of the production of net electricity
- The tritium self-sufficiency
- need to guarantee an optimum heat transfer in both Breeder Blanket (BB) and Steam Generator (SG) regions
- ✓ achieve a Tritium generation rate of about 320 g d<sup>-1(\*)</sup>
- ✓ efficient Tritium Extraction and Recovery (TER) system, very limited Tritium losses

(\*) Value referred to a DEMO configuration having a fusion power of 2 GW operating continuously at full power day with a tritium breeding ratio of 1.05



Large metallic surface areas characterized by thin wall operated at high temperature + Elevated Tritium concentration



Perfect conditions to enhance the Tritium permeation!

A. Santucci et al., "The issue of Tritium in DEMO coolant and mitigation strategies", <u>https://doi.org/10.1016/j.fusengdes.2020.111759</u>

## EFDA task (2012-2014)

- 1. Large literature review on:
  - State of the art and previous simulation works
  - Material data including antipermeation barriers
  - Tritium extraction/removal processes in Processes in He, water, PbLi

EFDA-Power Plant Physics & Technology

	Report for TA WP12-DAS-02			
Task Area: DAS - Design Assessment Studies				
TA Title:	A Title: In-vessel Components Design and Integration			
IDM Reference:	EFDA_D_2L8QPK, v.0.1			
Author(s):	D. Demange, R. Wagner (KIT-ITEP-TLK), F. Franza (KIT-INR)			
	S. Tosti, A. Santucci (ENEA-Frascati), A. Ciampichetti (ENEA-Brasimone)			
Tritiur	n migration issue	ENEN SKIT		
Tritiur	n permeation is high	n mitigations		
La	rge surface areas Textra	ction from blanket		
Th	in wall thickness T extra	ction from coolant		
Hi	<ul> <li>High temperatures</li> <li>Anti-permeation barriers</li> </ul>			
Production ~ 360 g/d He = 0.1% He = 500°C He = 0.1% He = 500°C He = 500°C				



## EFDA task (2012-2014)

 Development of a numerical tool using simplified model to simulate 3 blankets (HCPB, HCLL, WCLL) + parametric analysis

## Sodium-Cooled Fast Reactors Tritium Permeation Code adapted for fusion (**FUS-TPC** under Matlab)



*F. Franza* et al. "A model for tritium transport in fusion reactor components: The FUS-TPC code", <u>https://doi.org/10.1016/j.fusengdes.2012.01.002</u>

- <u>F. Franza</u> et al., "Tritium transport analysis in HCPB DEMO blanket with the FUS-TPC code", <u>https://doi.org/10.1016/j.fusengdes.2013.05.045</u>
- A. Santucci et al., "Impact of tritium solubility in liquid Pb-17Li on tritium migration in HCLL and WCLL blankets", https://doi.org/10.1109/SOFE.2013.6635438
- A. Santucci et al., "Model improvements for tritium transport in DEMO fuel cycle", https://doi.org/10.1016/j.fusengdes.2015.05.050

D. Demange et al., "Tritium management and anti-permeation strategies for three different breeding blanket options foreseen for the European Power Plant Physics and Technology Demonstration reactor study", <u>https://doi.org/10.1016/j.fusengdes.2014.04.028</u>

## EFDA task (2012-2014)

### Main outcomes of the sensitivity analysis

Table 56: Compilation of different scenarios for WCLL



#### On simulation works

Different design input data, assumptions, simplifications, etc.

### On materials

- Large uncertainty in the tritium solubility value (i.e., Sieverts' constant) in LiPb (2 o.o.m.) → large impact on T permeation and inventory
- Need of effective anti-permeation (anti-corrosion) barriers

#### On processes

- Efficient tritium extraction system (eta TER ≥ 0.8). Very demanding especially for T extraction from LiPb
- □ Helium CPS, Technology for T recovery from He
   → He cooled fission can be relevant
- Water CPS, T processing from water at large scale is energy consuming

## Outline

## □ The problem of Tritium migration in DEMO

- □ EDFA tasks (2012-2014)
  - □ FUS-TPC code

## □ EUROfusion (FP8) activities (2014-2018)

- □ EcosimPro
- Development of mitigation strategy to the Tritium permeation
  - □ Anti-permeation barriers
  - □ Coolant Purification System (CPS)
- □ KDII#2 activity (2018-2020)
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## EUROfusion (FP8) activities (2014-2018) - EcosimPro

The tritium transport analysis at system level has been implemented in Model implemented by CIEMAT using the object-oriented modelling software EcosimPro

- $\checkmark\,$  Integration of the secondary coolant (IHX and/or SG)
- $\checkmark\,$  Tritium permeation through pipes into rooms
- ✓ Introduction of  $H_2$ , HT,  $T_2$  co-permeation
- ✓ Effect of doping agent (i.e., H₂ and H₂O) in purge gas and in primary coolants
- ✓ Updated design

Uncertainty of the tritium transport property in the molten salt (HITEC salt) used in the IHX







About the strategy to mitigate tritium permeation, two main activities are carried out

Anti-permeation barriers(i.e., dedicated coatings or natural oxide layer)



✓ Coolant Purification System (CPS)

 $\alpha_{CPS} \rightarrow coolant \ fractiony \ inside \ CPS$ 

 $\eta_{CPS} \rightarrow CPS \ efficiency$ 



Taken from D. ladicicco et al., Efficient hydrogen and deuterium permeation reduction in  $Al_2O_3$  coatings with enhanced radiation tolerance and corrosion resistance, Nucl. Fusion 58 (2018) 126007





### **Development of anti-permeation barriers**

Mainly  $AI_2O_3$  coatings deposited with different techniques (PLD: Pulsed Laser Deposition and ALD: Atomic Laser Deposition)



### **Development of anti-permeation barriers**

## H<sub>2</sub> permeation tests at ENEA-Brasimone



 $\rm H_2$  permeation flux in EUROFER 97 bare sample goes from 6.5×10<sup>-9</sup> mol m<sup>-2</sup> s<sup>-1</sup> at 350°C up to 4×10<sup>-6</sup> mol m<sup>-2</sup> s<sup>-1</sup> at 550 °C



Deposition capability in complex shapes

Effectiveness under neutron irradiation

### **Coolant purification system**

A fraction of the primary coolant  $(\alpha_{CPS})$  is routed inside a dedicated system, the CPS, for removing tritium with a certain efficiency  $(\eta_{CPS})$ 



#### **Coolant flow rate inside the HCPB-CPS**



Tritium mass balance in coolant  $\alpha_{CPS}F = \frac{F_{T,p}/PRF_{BB}}{\eta_{CPS}\left(c_0 - \frac{F_{T,p}/PRF_{BB}}{F}\right)}$ 

- α<sub>CPS</sub>: coolant fraction to be processed inside the CPS
- $\eta_{CPS}$ : CPS efficiency
- F<sub>T,p</sub>: tritium permeation rate from blanket into coolant
- PRF<sub>BB</sub>: the Permeation Reduction Factor
- c<sub>0</sub>: the allowable tritium concentration in coolant

#### Coolant flow rate inside the WCLL-CPS

Input	Value		
η <sub>cps</sub>	0.9		
F <sub>T,p</sub> , g/d	42.93		
PRF <sub>BB</sub>	100		
C <sub>0</sub>	3 × 10 <sup>-6 (*)</sup> (*) corresponding to a T concentration inside water coolant of 1.85 × 10 <sup>11</sup> Bq kg <sup>-1</sup> (5 Ci kg <sup>-1</sup> )		
	$\alpha_{CPS} F \sim 45 \text{ kg h}^{-1}$ 15		

Helium CPS: pre-conceptual design

#### 1<sup>st</sup> Proposal: scale-up of the ITER process



- Large, but feasible, size of the beds (CuO + ZMS)
- ✓ Impact of the H<sub>2</sub> content in Hecoolant on the time before regeneration → p<sub>H2</sub> ≤ 300 Pa

Legend

- 1. HCS main compressor unit
- 2. Filters
- CuO beds unit
- 4. Zeolite Molecular Sieve beds unit



Helium CPS: pre-conceptual design

#### 2<sup>nd</sup> Proposal: based on Non-Evaporable Getters (NEGs)

	Cioucorto	Temperature T (K)					
	Sieverts' parameters		Embrittlement limit a	573		773	
Getter alloy	Α	В	(torr l/g)	K(T) Torr/(torr l/g)²	q <sub>0</sub> (T) (torr l/g)	K(T) Torr/(torr l/g)²	q <sub>0</sub> (T) (torr l/g)
ST707	4.8	6116	20	1.338E-06	14.98	7.726E-04	0.62
ST101	4.82	7280	20	1.303E-08	151.74	2.524E-05	3.45
ZAO	5.76	7290	~100	1.090E-07	52.46	2.134E-04	1.19



Legend

- HCS main compressor unit
- 2. Filters
- Non regenerable getters
   NEGs beds
- 5. Heaters
- 6. External pump

 Among the NEGs, the ZAO alloy is the most suitable for He-CPS application

#### Water-CPS: pre-conceptual design



#### Water-CPS: pre-conceptual design

#### Water Distillation



- Packed column with Sulzer CY Gauze Packing
- Operative pressure: 10 kPa
- Anti-permeation barrier with PRF 100
- Tritium content in feeding flow rate: 5 Ci kg<sup>-1</sup>
- Tritium content in bottom stream, sent to WDS: 100 Ci kg<sup>-1</sup>



## KDII#2 activity (2019-2020)

#### Scope of the Key Design Integration Issue (KDII) #2:

Addresses some specific issues of the design of breeding blanket and ancillary systems related to the use of helium or water as a coolants for the blanket and impact on the overall plant design

#### Methodoly for the Tritium migration issue:

- Collect input from different plant designers (Breeding Blanket, Tritium Extraction and Recovery, Coolant Purification system, Anti-permeation barriers, Primary Heat Transfer System, Power Conversion System, etc.)
- Identify a reference scenario and suitable ranges for the sensitivity analysis
- □ Perform tritium simulation analysis at system level (EcosimPro) with the most updated plant architecture
- □ Analise the simulation results with respect to some "Operating Limit"



EFDA\_D\_2P2X46

#### Key Design Integration Issue #2 Summary Report 2020

Date: 07/10/2022 IDM number: EFDA\_D\_2P2X46

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A. Spagnuolo et al. "Key Design Integration Issue #2 Summary Report 2020", IDM number: EFDA\_D\_2P2X46

A. Spagnuolo et al., "Integrated design of breeding blanket and ancillary systems related to the use of helium or water as a coolant and impact on the overall plant design", <u>https://doi.org/10.1016/j.fusengdes.2021.112933</u>



# KDII#2 activity (2019-2020)





Fig. 2. Schematic drawing of the tritium and heat removal paths of the Water-cooled DEMO architecture.

Legend BB: Breeding Blanket CPS: Coolant Purification System IHTS: Intermediate Heat Transfer System PCS: Power Conversion System PHTS: Primary Heat Transfer System TER: Tritium Extraction System VVPSS: Vacuum Vessel Pressure Suppression System



Table 6. Sensitivity matrix for WCLL DEMO 2017 base

	Parametric scan			Scan
Case ID	Parameter (unit)	Min/ Intermediate	Max	411
1	CPS by pass flow rate [kg/h]	20	360	
2	TER efficiency [%]	80	95	
3	T permeation rate through the FW [mg/d]	0	20	
4	PRF in BB [-]	1	1000	
5	H <sub>2</sub> partial pressure [ppm]	8	100	
6	PRF in PbLi loop[-]	100	1000	
7	Leak rate at the SG/IHX [gal/d] [51]	3	5	



Figure 9. WCLL Breeding Blanket Schematic of Tritium Flow.

# KDII#2 activity (2019-2020)

#### **HCPB**

## Table 5. HCPB DEMO 2017 baseline weferen

Parameter (unit)	HCPB Case-u	Case		
CPS by pass flow rate [kg/s]	3			
CPS efficiency [%]	90			
Partial pressure in He coolant [Pa]	0			
He purge gas flow rate [kg/s]	0.497			
TER efficiency [%]	80			
T permeation rate [mg/d]	3.5			
He coolant leakage frequency [% inventory/yr]	4			
PRF in IHX [-]	100			
Molar fraction of steam in purge gas [%]	0			
Leak rate at the IHX[kg/h] [51]	D 0			
Table 7. Sensitivity matrix for HCPB DEMO 2017 baseline protection Sca				
Parametric scan		redn		

	Parametric scan					
Case ID	Parameter (unit)	Min/ Intermediate	Max			
1	CPS by pass flow rate [kg/s]	2	4			
2	CPS efficiency [%]	80	95			
3	Partial pressure in He coolant [Pa]	1	300			
4	He purge gas flow rate [kg/s]	0.25	0.497			
5	TER efficiency [%]	80	98			
6	T permeation rate [mg/d]	0	60			
7	He coolant leakage frequency [% inventory/yr]	0	36.5			
8	PRF in IHX [-]	1	1000			
9	Molar fraction of steam in purge gas [%]	0	4			
10	Leak rate at the IHX[kg/h] [51]	0.024	0.34			





Fluid flow

# Updated plant architecture Legend

**BB: Breeding Blanket CPS: Coolant Purification System IHTS: Intermediate Heat Transfer** System **PCS: Power Conversion System** PHTS: Primary Heat Transfer System **TER: Tritium Extraction System VVPSS: Vacuum Vessel Pressure** Suppression System



Figure 10. HCPB Breeding Blanket Schematic of Tritium Flow.

# KDII#2 activity (2019-2020)

Operating limit description	Limit value*	Notes
Release in Tokamak building	1 [g/y]	This limit is posed on the environmental releases to the Stack. However, in this work, it is conservatively used to compare the results in term of permeation rates from the PHTS and TER piping into the Tokamak building.
Threshold to activate the Detritiation System (DS)	10 <sup>8</sup> [Bq/m³]	The limit for serving a room by the DS system depends on the concentration of tritium per volume inside the room. In ITER the threshold to switch from Heating Ventilation and Air Conditioning (HVAC) to DS is $1x10^8$ Bq/m <sup>3</sup> . (ITER_D_35YGFT v1.2). In DEMO, the same threshold for DS intervention is assumed.
Threshold for nuclear classification of the secondary/intermediate systems according to Equipment Sous Pression Nucléaire (ESPN) Directive	370 [GBq]	<ul> <li>Equipment is considered nuclear equipment if:</li> <li>It is submitted to the Pressure Equipment Directive (PED);</li> <li>Provide confinement of radioactive nuclides for at least one of its normal operating conditions. This means that if secondary/intermediate system is not ESPN, the activity of any secondary/intermediate system component (vessels, not piping) should be kept under 370 MBq (10 mCi) during normal operation. The activity of tritium, <sup>16</sup>N and <sup>17</sup>N are divided by a factor 1/1000 according to ESPN rules, allowing consequently an increase of the total contamination of secondary/intermediate systems to 370 GBq (10 Ci).</li> </ul>

\* For a complete explanation of the values assumed for the different operating limit, please refer to: A. Spagnuolo et al. "Key Design Integration Issue #2 Summary Report 2020", IDM number: EFDA\_D\_2P2X46



## Conclusions

- □ Tritium migration is an important issue in DEMO
- Main contributors to the problem have been identified and activities to mitigate this issue are in placed
- □ Relevant experiments are on going in the FP9 to:
  - Reduce the uncertainty in the Sieverts constant in LiPb
  - Measure the Tritium permeation under "more DEMO relevant conditions" (i.e., presence of steam, co-permeation, oxidized surface, etc.)
- □ Tritium properties in molten salt (HITEC) still require proper investigation



# Thank you!



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