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Tritium Extraction from Liquid Blankets of Fusion Reactors via Membrane Gas–Liquid Contactors

Silvano Tosti *  and Luca Farina 

ENEA, Nuclear Department, ENEA Frascati Research Center, Via E. Fermi 45, 00044 Frascati, Italy;
luca.farina@enea.it

* Correspondence: silvano.tosti@enea.it

Abstract: The exploitation of fusion energy in tokamak reactors relies on efficient and reliable tritium management. The tritium needed to sustain the deuterium–tritium fusion reaction is produced in the Li-based blanket surrounding the plasma chamber, and, therefore, the effective extraction and purification of the tritium bred in the Li-blankets is needed to guarantee the tritium self-sufficiency of future fusion plants. This work introduces a new technology for the extraction of tritium from the Pb–Li eutectic alloy used in liquid blankets. Process units based on the concept of Membrane Gas–Liquid Contactor (MGLC) have been studied for the extraction of tritium from the Pb–Li in the Water Cooled Lithium Lead blankets of the DEMO reactor. MGLC units have been preliminarily designed and then compared in terms of the permeation areas and sizes with the tritium extraction technologies presently under study, namely the Permeator Against Vacuum (PAV) and the Gas–Liquid Contactors (GLCs). The results of this study show that the DEMO WCLL tritium extraction systems using MGLC require smaller permeation areas and quicker permeation kinetics than those based on PAV (Permeator Against Vacuum) devices. Accordingly, the MGLC extraction unit exhibits volumes smaller than those of both PAV and GLC.

Keywords: tritium; Pb–Li eutectic alloy; fusion reactors breeding blankets; membrane gas–liquid contactors



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1. Introduction

Recent energy transition policies are promoting the penetration of clean energy into the energy systems: in 2023, more than 560 gigawatts (GW) of renewable energy capacity have been added [1]. Nevertheless, most of the increase in global energy demand has been supplied by fossil fuels responsible for a new CO₂ emissions peak, so future energy policies will need to consider any source capable of contributing to decarbonization to meet the mitigation of greenhouse gas emissions and to limit increases in the global mean surface temperature [2]. Fusion energy generation has, in the long term, the potential to contribute to the achievement of the sustainable development goals (SDG), and, in view of that, several new governmental, public–private, and private initiatives have recently been launched to accelerate the development and demonstration of fusion energy [2–5]. Among the significant fusion programs, the EUROfusion Consortium developed the DEMO project aimed at demonstrating the technological and economic viability of magnetically confined fusion reactors by producing about 500 MW of net electricity and achieving tritium self-sufficiency.

The fusion reaction between deuterium and tritium is the most studied since it can attain the highest reaction rate at a lower temperature than other fuels. While deuterium is

extracted from natural sources, tritium is produced in a Li blanket by the reaction between neutrons and Li isotopes through the reactions:



with n a 14 MeV neutron and n' a thermal (slow) neutron.

The blanket can consist of Li ceramics (solid breeders) or liquid Li materials. The addition of a neutron multiplier (Pb) with a breeder material (Li) is realized in the eutectic Pb–Li alloy that, accordingly, exhibits the high tritium breeding ratio needed to guarantee the tritium self-sufficiency of the reactor. Pb–Li has high thermal conductivity, useful for efficiently extracting heat from blankets, while its low tritium inventory meets the safety requirements [6]. For this reason, the eutectic Pb–Li alloy (lithium 15.6 at. %) is the best candidate material as a liquid breeder of fusion reactor blankets. Presently, three design options are taken into consideration for the DEMO reactor: the Helium Cooled Lithium Lead (HCLL), the Water-Cooled Lithium Lead (WCLL), and the Dual Coolant Lithium Lead (DCLL) [7–11]. A liquid Pb–Li breeder is also considered as one option for the European Test Blanket Module (EU TBM) of the ITER project [12].

Once produced by the reactions (1) and (2) into a liquid breeder, the tritium is absorbed into the Pb–Li according to Sieverts' law. This expression relates the hydrogen partial pressure in the gas phase with the concentration of hydrogen absorbed in the liquid:

$$c_H = k_S P_H^{0.5} \quad (3)$$

where c_H is the hydrogen concentration in the liquid metal (at. fr. m^{-3}), k_S is the Sieverts' constant (at. fr. $\text{m}^{-3} \text{Pa}^{-0.5}$), and P is the hydrogen partial pressure in the gas phase (Pa) in equilibrium with the liquid Pb–Li. So far, the measured values of k_S for hydrogen into Pb–Li at the temperatures of interest for fusion applications are spread in the range 10^{-8} – 10^{-6} at. fr. $\text{Pa}^{-0.5}$, then introducing significant uncertainties in the design of liquid blankets [13]. The hydrogen isotope solubility into Pb–Li is a key parameter for assessing both the efficiency of the tritium extraction systems and the tritium inventory. In fact, its value determines the sizing of the tritium extraction units and impacts on the tritium inventory and the safety analysis of the breeding blankets.

In this work, a new technology based on Membrane Gas–Liquid Contactors (MGLCs) made of stainless steel porous membrane tubes is proposed to recover tritium from the liquid Pb–Li alloy. Such membranes are commonly used for gas and air filtering in industries like pharmaceuticals, chemical processing, and petrochemicals. These membranes can be produced by sintering metal powders so that, in principle, the different metals and alloys and several geometries other than tubes can be adopted to realize reliable and cost-effective process units for the breeding blankets and tritium extraction systems.

In particular, MGLC extraction units are preliminarily designed and compared with the presently studied technologies for the extraction of tritium from the liquid blankets, namely the Gas–Liquid Contactors (GLCs) and the Permeator Against Vacuum (PAV).

2. Tritium Extraction from Liquid Blankets

The reference process studied for the tritium extraction from the liquid metal uses the Gas–Liquid Contactors (GLCs), vertical columns where an inert gas (He, Ar) rises up counter-currently to the Pb–Li stream fed at the top (300–500 °C). Gas–Liquid Contactors can consist of packed bed columns, bubble columns, or spray columns [6,14]. Inside these process units, the transport of tritium takes place, driven by its partial pressure gradient

between the liquid and gas phases, corresponding respectively to the Pb–Li and the inert gas streams. Due to their high packing area ($750 \text{ m}^2 \text{ m}^{-3}$), at 400°C , the packed columns have achieved 30% tritium extraction efficiency [15], while preliminary results of recent experiments have evaluated the extraction efficiency values over 40% at 450°C [16].

The Permeator Against Vacuum (PAV)

As an alternative to the Gas–Liquid Contactors, Permeator Against Vacuum (PAV) devices have been proposed for extracting tritium from liquid Pb–Li. Figure 1 represents schematically the PAV concept: Pb–Li flows in the shell side of a permeator unit consisting of a dense metal tube. The tritium solubilized into the liquid metal diffuses selectively through the membrane and is collected by vacuum pumping inside the membrane lumen.

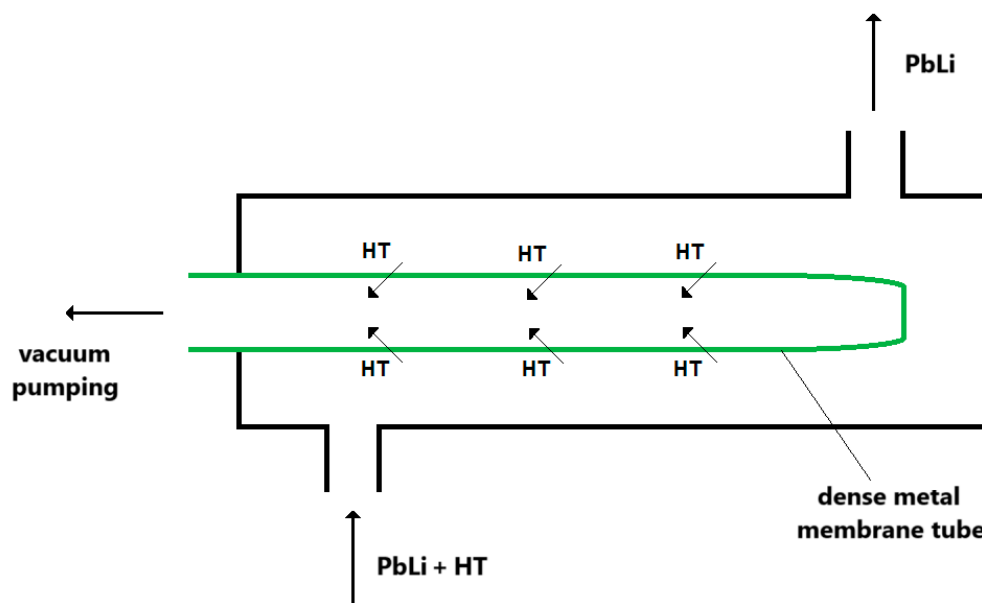


Figure 1. Scheme of the Permeator Against Vacuum (PAV): hydrogen isotopes permeate selectively through the dense metal tube from the Pb–Li into the membrane lumen, where they are extracted by vacuum pumping.

In a PAV permeator, the hydrogen moves from the Pb–Li to the gas phase downstream of the metal membrane through four main steps: (i) the tritium diffusion through the boundary layer of the liquid metal, (ii) the diffusion through the metal wall, (iii) the recombination of T atoms into molecules over the downstream metal membrane surface, and (iv) the tritium diffusion through the boundary layer of the gas phase. The main contribution to the mass transfer resistance is given by the diffusion through the metal wall (step ii) and the recombination of the T atoms over the metal surface (step iii). The last contribution becomes the controlling one when the membrane surfaces are oxidized, as it will be discussed for the Nb-based PAV in Section 5.1.1.

In particular, the diffusion through the metal wall (step ii in the above description) is ruled by Fick's law:

$$J = \frac{D}{th} (c_{H,up} - c_{H,down}) \quad (4)$$

where J (mol s^{-1}) is the hydrogen flow rate at the gas–liquid interface inside the pores of the membrane D ($\text{m}^2 \text{ s}^{-1}$) is the hydrogen diffusion coefficient through the metal lattice, th (m) is the thickness of the metal membrane, and $c_{H,up}$ and $c_{H,down}$ (mol m^{-3}) are the hydrogen concentration upstream and downstream of the metal membrane, respectively.

The use of PAV devices for the extraction of hydrogen solubilized into liquid metals leads to benefiting the features of the membrane processes: continuous and simple opera-

tional modes, modularity, and high extraction efficiency. Specific configuration geometries consisting of spiral-rolled double membranes and cylindrical and concentric multi-channel PAV have been studied for improving extraction efficiency [17,18]. The adoption of these design configurations characterized by many joints could, however, reduce the reliability of the PAV units because of the occurrence of leaks due to the erosion–corrosion of Pb–Li [19].

The most studied materials for the PAV membranes are the α -Fe and the refractory metals (V, Nb). The first one is cheap but is characterized by modest hydrogen permeability ($7.99 \times 10^{-11} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-0.5}$ at 400°C) [20]. The Nb and V have large hydrogen permeabilities (3.59×10^{-6} and $3.40 \times 10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-0.5}$ at 400°C , respectively), but such membrane materials could suffer embrittlement due to the high hydrogen solubility of the refractory metals [20].

V-based PAVs have exhibited tritium extraction efficiencies close to 39% [21], while PAV units made of Nb and designed for the WCLL of DEMO are expected to have extraction efficiency values of about 20% at 330°C and up to 50% at higher temperatures [22].

3. MGLC for Tritium Extraction from Pb–Li

With the aim to reduce the hydrogen mass transfer resistance, the use of porous membranes has been studied to replace the metal membranes used by PAV. The resulting device made of a porous membrane acts as a special Gas–Liquid Contactor where the interface between the liquid Pb–Li and the gas phase is realized inside the membrane pores [23–25]. It is named Membrane Gas–Liquid Contactor (MGLC) and can be seen as a combination of the GLC and PAV concepts, as shown in Figure 2. In an MGLC, the liquid Pb–Li partially penetrates the pores of the membrane, where it gets in touch with a gas phase consisting of hydrogen isotopes extracted by inert gas or vacuum pumping. The geometry (mainly the pore size) and the materials of the porous membranes have to be designed in order to ensure that, under the operating conditions of the extraction systems, the liquid metal doesn't pass into the gas phase.

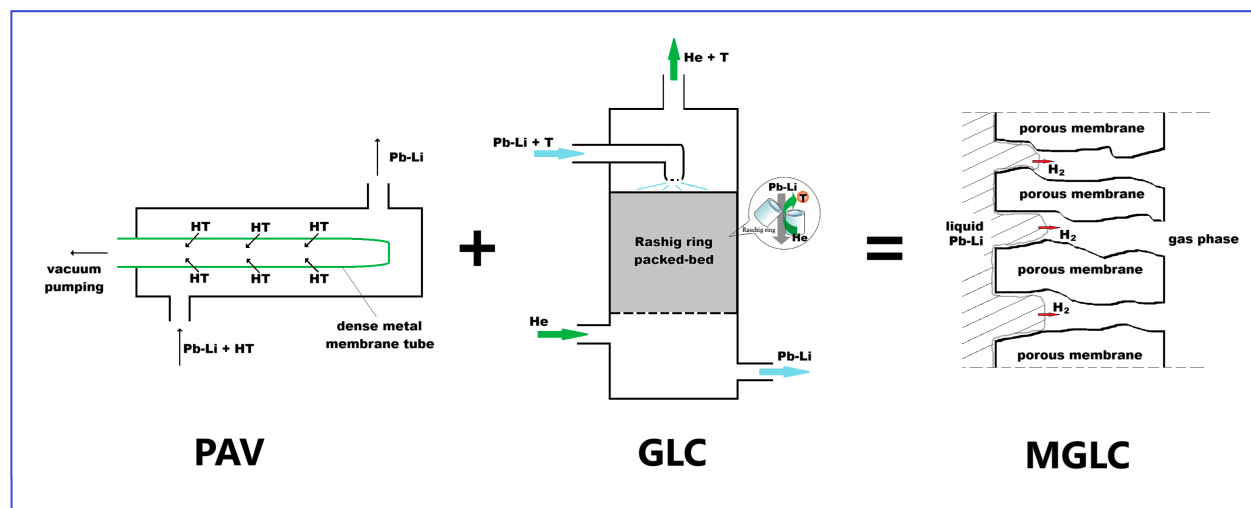


Figure 2. The Membrane Gas–Liquid Contactor concept: this device works as a special Gas–Liquid Contactor where the interface between the liquid Pb–Li and the gas phase is realized inside the membrane pores.

First tests used MGLC made of stainless steel porous tubes; however, functional materials studied to improve the resistance to the erosion–corrosion of the Pb–Li could be proposed. For instance, special metal alloys or ceramics (e.g., SiC) could be used to produce porous membrane tubes via powder sintering.

3.1. Hydrogen Isotopes Transport in MGLC

Thanks to the use of porous membranes, the MGLC has hydrogen mass transfer resistances lower than those of the PAV units that, mainly, are controlled by the hydrogen diffusion through the lattice of the metal dense wall separating the liquid metal and gas phases. In the MGLC concept, the mass transfer resistance through the metal wall is missing, while the resistance due to the hydrogen diffusion through the gas phase inside the membrane pores is present, a term that is negligible, as it will be discussed in the next section.

The model of hydrogen transport through the MGLC is shown in Figure 3. Under the assumption to neglect the hydrogen diffusion from the bulk liquid metal to the Pb–Li boundary layer, two main steps are identified: (i) the transport from the Pb–Li boundary layer to the gas phase inside the membrane pores and (ii) the diffusion through the gas inside the pores.

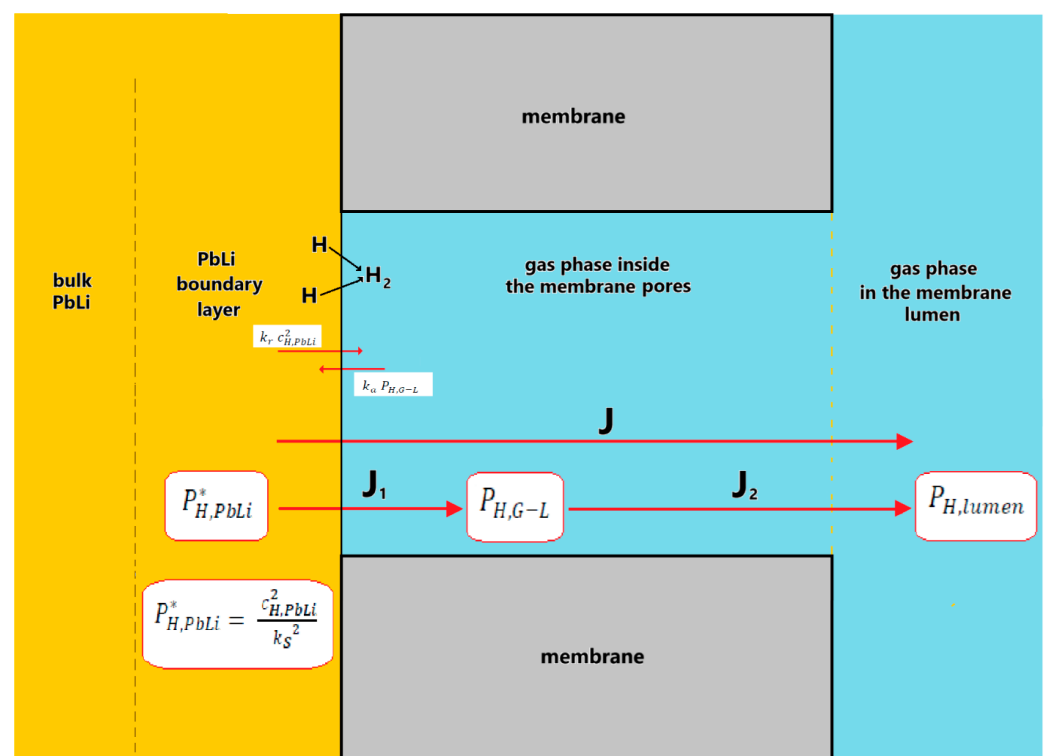


Figure 3. Scheme of the hydrogen mass transport from Pb–Li layer to membrane lumen: the hydrogen partial pressure gradient from Pb–Li boundary layer (yellow) to the gas phase inside the membrane pores (light blue) is the driving force of the permeation process.

The hydrogen flow rate from the Pb–Li boundary layer to the gas phase inside the membrane pores occurring at the gas–liquid interface (J_1 , mol s^{−1}) is given by the balance of two opposite flow rates:

- (i) The recombination of hydrogen atoms into molecules from the liquid metal into the gas phase expressed by $k_r c_{H,PbLi}^2$;
 - (ii) The adsorption of hydrogen from gas into the liquid phase expressed by $k_a P_{H,G-L}$.
- Accordingly, the resulting expression of J_1 is

$$J_1 = k_r c_{H,PbLi}^2 - k_a P_{H,G-L} \quad (5)$$

with the recombination coefficient expressed by k_r (m⁴ mol^{−1} s^{−1}), the concentration of hydrogen into Pb–Li by $c_{H,LiPb}$ (mol m^{−3}), the hydrogen adsorption coefficient by k_a (mol

$\text{m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$), and the hydrogen partial pressure in the gas phase in contact with the liquid metal by $P_{H,G-L}$ (Pa).

The recombination coefficient can be taken from the literature [26], while the hydrogen adsorption coefficient k_a is assessed by posing $J = 0$ in Formula (5), a condition realized when steady-state conditions are achieved. Then combining Equations (3) and (5), it results in

$$k_a = k_r k_S^2 \quad (6)$$

According to Sieverts' law (3), the partial pressure of hydrogen into Pb–Li $P_{H,LiPb}^*$ (Pa) is expressed by

$$P_{H,PbLi}^* = \frac{c_{H,PbLi}^2}{k_S^2} \quad (7)$$

and Formula (4) becomes

$$J_1 = k_a (P_{H,PbLi}^* - P_{H,G-L}) \quad (8)$$

It is noteworthy that the adsorption coefficient k_a represents the inverse of the mass transfer resistance of the hydrogen transport from the Pb–Li boundary layer to the gas phase inside the membrane pores.

The transport due to hydrogen diffusion through the gas inside the pores is given by

$$J_2 = \frac{Pe}{th} (P_{H,G-L} - P_{H,lumen}) \quad (9)$$

with Pe ($\text{mol m}^{-1} \text{s}^{-1} \text{Pa}^{-1}$) the permeability of hydrogen diffusing through the membrane pores, th (m) the thickness of the porous membrane, and $P_{H,lumen}$ (Pa) the hydrogen partial pressure in the membrane lumen. The term th/Pe stands for the contribution to the mass transfer resistance due to the hydrogen diffusion through the gas inside the pores.

At steady state, the hydrogen flowing from the Pb–Li boundary layer to the gas phase inside the membrane pores equals the hydrogen diffusing through the gas inside the pores, and, combining Formulas (9) and (10), it is

$$J = h (P_{H,PbLi}^* - P_{H,lumen}) \quad (10)$$

where h ($\text{mol m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$) is the overall mass transfer coefficient related to the transport from the Pb–Li boundary layer to the gas phase in the membrane lumen. It is given by

$$h = \frac{1}{\frac{1}{k_a} + \frac{th}{Pe}} = \frac{1}{\frac{1}{k_r k_S^2} + \frac{th}{Pe}} \quad (11)$$

The expression (11) puts evidence that the overall mass transfer resistance is given by the parallel of two terms: (i) the mass resistance due to the recombination of the hydrogen atoms over the gas–liquid interface and their desorption from the metal, and (ii) the mass resistance in the permeation of the hydrogen through the gas phase inside the membrane pores. The relationship between the two terms ($\frac{1}{k_a}$ and $\frac{th}{Pe}$) is useful to identify which mechanism is controlling this phenomenon, as it will be discussed afterwards.

4. Design of the Membrane Gas–Liquid Contactor

The preliminary design of MGLC units for the extraction of tritium from the liquid Pb–Li relies on the calculation of the proper membrane pore size, allowing the direct contact between the liquid and gas phases and avoiding the Pb–Li entering the membrane lumen. Then, the overall hydrogen mass transfer coefficient has been assessed under the operating conditions of WCLL blankets.

4.1. Calculation of the MGLC Pore Size

Imbibition into porous materials can be modeled by the Washburn equation that, in principle, is applied to describe the behavior of a liquid in a capillary tube. Inside the pores of the MGLC, at equilibrium conditions, the difference between the external pressure acting on the liquid, P_L (Pa), and the pressure of the gas phase present in the membrane lumen, P_G (Pa), is balanced by the forces due to the liquid's surface tension γ (N m^{-1}):

$$P_L - P_G = -\frac{4\gamma \cos \theta}{d} \quad (12)$$

where d is the pore diameter (m), and θ is the contact angle between the penetrating liquid and the membrane wall. Practically, the contact angle of liquid metals on solids is around 140° .

The Pb–Li surface tension γ vs. temperature T (K) is given from [27]

$$\gamma = 0.52 - 0.11 \times 10^{-3}T \quad (13)$$

with γ in N m^{-1} and T , temperature, in K.

The pore size of the MGLC for the extraction of tritium from the liquid Pb–Li has been assessed via the above formulas in the temperature range of 300–500 °C and for different values of pressure drops (ΔP) between upstream and downstream sides of the porous membrane, as reported in Table 1.

Table 1. MGLC pore diameter vs. ΔP for the temperatures of 300, 400, and 500 °C.

ΔP , kPa	Pore Diameter, m		
	at 300 °C	at 400 °C	at 500 °C
50	2.80×10^{-5}	2.73×10^{-5}	2.67×10^{-5}
100	1.40×10^{-5}	1.37×10^{-5}	1.33×10^{-5}
200	7.00×10^{-6}	6.83×10^{-6}	6.66×10^{-6}
300	4.67×10^{-6}	4.55×10^{-6}	4.44×10^{-6}
400	3.50×10^{-6}	3.41×10^{-6}	3.33×10^{-6}
500	2.80×10^{-6}	2.73×10^{-6}	2.67×10^{-6}
1000	1.40×10^{-6}	1.37×10^{-6}	1.33×10^{-6}

It is verified that membranes of pore sizes $\leq 3 \mu\text{m}$, such as those adopted in the following analysis, can withstand ΔP up to 400 kPa.

4.2. Assessment of the MGLC Overall Mass Transfer Coefficient

The MGLC overall mass transfer coefficient evaluated via Formulas (5) to (10) of the transport model discussed in Section 3.1 has been compared with the results of an experimental work in which a porous membrane has been tested for extracting hydrogen from Pb–Li [25]. The Membrane Gas–Liquid Contactor consisted of a porous 316L stainless steel tube of average pore size around $2.8 \mu\text{m}$ with dimensions of 42 mm in length and outer/internal diameters of 12.9/8.6 mm, respectively, corresponding to a membrane area of $1.418 \times 10^{-3} \text{ m}^2$. As discussed in Section 4.1, such a porous membrane can withstand upstream–downstream membrane pressure drops up to 400 kPa.

In the experiments carried out at 370 °C, the following value of the overall mass transfer coefficient has been assessed:

$$h = 7.249 \times 10^{-8} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1} \quad (14)$$

The main parameters of the transport model discussed in the previous section depend on the Sieverts' constant k_S . Therefore, the transport analysis has to consider the large spread of the literature data around the Sieverts' constant and, in turn, the uncertainties reflected in the evaluation of the model parameters and the overall mass transfer coefficient.

The dependance of the Sieverts' constant on the temperature is given by

$$k_S = k_S^0 e^{-\frac{E_S}{R \cdot T}} \quad (15)$$

where k_S^0 (at. fr. $\text{m}^{-3} \text{Pa}^{-0.5}$) is the pre-exponential factor, E_S is the activation energy (J mol^{-1}), R the gas constant ($\text{J mol}^{-1} \text{K}^{-1}$), and T (K) is the temperature.

The Sieverts' constant values k_S^0 and E_S reported in Table 2 have been used for this assessment and are representative of the large spread of data available in literature.

Table 2. Pre-exponential factor and the activation energy of Sieverts' constant.

k_S^0 ($\text{mol m}^{-3} \text{Pa}^{-0.5}$)	E_S (J mol^{-1})	Ref.
7.29×10^{-4}	1350	Reiter [28]
2.68×10^{-2}	6100	Shumacher et al. [29]
2.73×10^{-1}	12,844	Aiello et al. [30]

For the recombination coefficient (k_r , $\text{m}^4 \text{mol}^{-1} \text{s}^{-1}$), the following expression has been adopted [31]:

$$k_r = 1.14 \cdot 10^{-1} \cdot \exp\left(\frac{-29717}{R \cdot T}\right) \quad (16)$$

Through the expression (5), the values of the Sieverts' constant and the recombination coefficient have been used to calculate the adsorption coefficient k_a in the temperature range of 300 to 600 °C, as reported in Table 3. The Sieverts' constant from Reiter is around 3 to 4 orders of magnitude smaller than the k_S values from Shumaker et al. [29] and Aiello et al. [30], are close to each other and range from 10^{-8} to $10^{-6} \text{mol m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$. At 330 °C, the design temperature of the tritium extraction system of the DEMO Water Cooled Lithium Lead (WCLL), the adsorption coefficient k_a goes from 10^{-10} to $10^{-7} \text{mol m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$.

Table 3. Assessment of the adsorption coefficient, k_a .

T , °C	Adsorption Coefficient k_a , $\text{mol m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$		
	Reiter [28]	Shumaker et al. [29]	Aiello et al. [30]
300	6.76435×10^{-11}	1.24669×10^{-8}	5.72631×10^{-8}
400	1.85834×10^{-10}	4.60620×10^{-8}	3.22229×10^{-7}
500	3.93088×10^{-10}	1.21368×10^{-7}	1.15976×10^{-6}
600	7.00370×10^{-10}	2.56145×10^{-7}	3.11293×10^{-6}

The hydrogen transport in the gas phase inside the pores of the membrane is defined by the values of the Knudsen number (Kn) expressed by

$$Kn = \lambda / r \quad (17)$$

with λ (m) the free mean path and r (m) the pores' size.

Two transport regimes are identified for the hydrogen transport in the gas phase:

- The viscous (Poiseuille) regime occurring when the mean free path is smaller than the pore size ($Kn < 0.01$).
- The molecular (Knudsen) regime occurring when the mean free path is larger than the pore size ($Kn > 1$).

In the temperature range of 300–400 °C, Kn is of the order of 1×10^{-1} , and, therefore, the hydrogen diffusion through the MGLC pores follows a mixed regime of Poiseuille–Knudsen. In agreement with previous evaluations [32], in the considered temperature range, the calculated values for the gas permeance $\left(\frac{Pe}{th}\right)$ are around $2 \times 10^{-5} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$.

Therefore, the MGLC here considered exhibits the mass transfer resistance in the gas phase inside the pores of the membrane th/Pe ($\approx 10^5 \text{ mol}^{-1} \text{ m}^2 \text{ s Pa}$) much smaller than the resistance due to the recombination of hydrogen atoms and their desorption from Pb–Li $1/k_a$, i.e., ($\approx$ from 10^7 to $10^{10} \text{ mol}^{-1} \text{ m}^2 \text{ s Pa}$). This means that the hydrogen transfer at the Gas–Liquid interface due to the hydrogen atoms recombination and then the hydrogen desorption from the liquid metal is controlling the transport mechanism. Since the mass transfer resistance through the gas phase in the membrane pores is negligible and according to (11), it results $h \approx k_a$. The dependance of the overall mass transfer coefficient (h , $\text{mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$) on the temperature can further be obtained by extrapolating the experimental results:

$$h = 7.40 \times 10^{-5} e^{\frac{-41328.12}{RT}} \quad (18)$$

In Figure 4, the overall hydrogen mass transfer coefficient obtained from the experiments is compared with the values calculated from the literature. This analysis confirms that, in good approximation, the values of the Sieverts' constant determined by Shumacher et al. [29] and Aiello et al. [30] leads to assessing overall mass transfer coefficients close to that determined by the experiments.

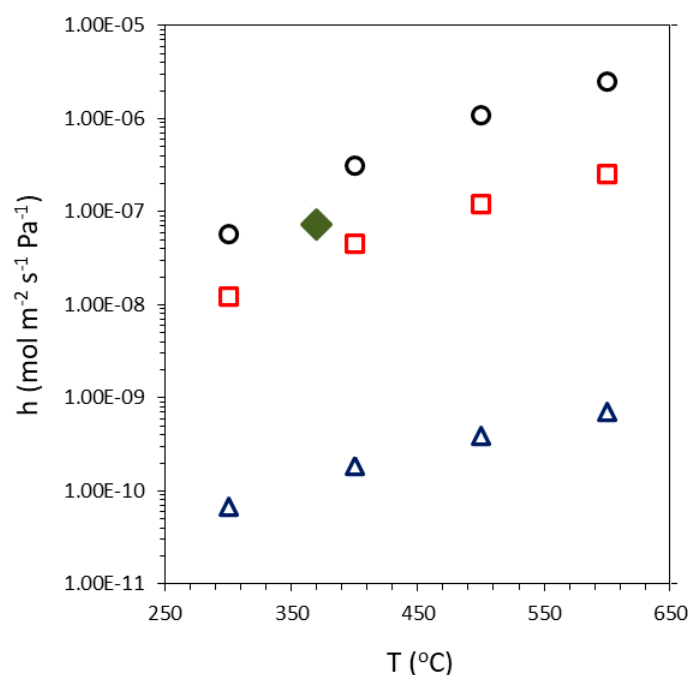


Figure 4. Comparison of the overall hydrogen mass coefficient obtained from the experiment with the values calculated from the literature: Aiello et al. [30] (White circle black), Schumacher et al. (White square red) [29], Reiter (White triangle blue) [28] and Tosti et al. (Black diamond green) [25].

5. Comparison of the Tritium Extraction Units Based on MGLC, PAV, and GLC (Packed Columns)

The Tritium Extraction and Removal (TER) system of the water-cooled lithium–lead (WCLL) blanket for the European DEMO Tokamak Reactor has the main function of extracting the tritium produced in the breeding modules from Pb–Li [33].

This work designs the preliminary DEMO WCLL tritium extraction system made of MGLC membranes. Further, the MGLC membranes will be compared with the tri-

tium extraction units based on the most promising technologies, namely PAV and GLC, presently developed.

The main design parameters of the WCLL tritium extraction and removal system are reported in Table 4. Specifically, the Pb–Li circuit consists of four loops for the outboard (OB) and two for the inboard (IB) blanket sectors.

Table 4. Main design parameters of the tritium extraction and removal system of the WCLL blanket [33].

Temperature, °C	330
Tritium partial pressure, Pa	55
Tritium conc. in Pb–Li, mol m ^{−3}	1.41×10^{-2}
Total mass flow rate, kg/s	1127 (OB) + 499 (IB)

5.1. PAV

The PAV devices presently under study are made of refractory metals (Nb and V) and adopt tube- or flat-shaped membrane configuration. PAV with flat membranes requires higher permeation surfaces than tubular PAV, although the flat membranes can be allocated in units of volumes smaller than those of tubular membranes. In addition, the reliability of many welded tubes should be compared with that of welded or flanged flat components, and consequently, the choice of PAV configuration also relies on the manufacturing and joining techniques available for the different geometries.

For the DEMO WCLL, two PAV configurations are studied:

- Permeable U-shaped Nb tubes inside a vacuum vessel;
- V plates welded to stainless steel structures.

5.1.1. Nb PAV

The Nb PAV consists of permeable niobium tubes inside a cylindrical vacuum vessel. Nb tubes are in the configuration of U-shaped parallel channels immersed in Pb–Li while the vacuum is pumped inside the Nb pipes to remove the gas extracted [34].

The presence of oxidation over the Nb surface can reduce the overall hydrogen permeation through the metal membrane; in this case, the transport mechanisms occurring at the metal surfaces are the controlling ones (surface-limited regime). When clean metal surfaces can be guaranteed, the PAV mass transport resistance is mostly due to the hydrogen diffusion through the metal lattice, and the permeation fluxes are higher (diffusion-limited regime). Based on the results of experimental campaigns, the design of the Nb PAV has been carried out for two permeation regimes: diffusion- and surface-limited [22,35]. The resulting size of the PAV unit designed for one OB loop of the DEMO WCLL is reported in Table 5 [33].

The overall PAV extraction system (four loops in OB and two loops in IB) requires about 21,500 and 5600 m² of permeation surface in the surface- and diffusion-limited regimes, respectively.

Table 5. Nb PAV unit designed for one loop of the OB [33].

	Surface-Limited Regime	Diffusion-Limited Regime
vessel diameter, m	6	4
height of tank, m	8	6
number of tubes	1600	855
tube diameter, mm	9.2	9.2
tube length, m	57	27.75
total surface area, m ²	3723	968

5.1.2. V PAV

The V PAV is made of metal permeable membranes in a flat configuration. Firstly, 1 m long flat membranes were welded to a steel structure by TIG and electron beam welding, and the formation of leaks and loss of integrity of the components were experienced [36]. Then, a new and more reliable prototype has been built based on a screwed attachment of vanadium by using graphite gaskets [37]. Table 6 shows the sizing of a modular V PAV unit capable of treating a flow rate of 55 kg/s is reported.

Table 6. V PAV modular unit [33].

width, m	1.90
channel height, m	5×10^{-3}
length, m	15
membrane thickness, m	1×10^{-3}
total surface area, m ²	2400

Scaling-up these results to the entire PAV extraction system shows that about 30 modular V PAV units are needed for processing the flow rate of four Pb–Li loops in OB and two loops in IB, i.e., 1626 kg/s. The overall permeation surface of the 30 V PAV units is 70,900 m².

5.2. GLC

Gas–Liquid Contractors using vertical packed-bed columns for applications in the chemical and process industry rely on mature technology. GLCs has also proposed the extraction of tritium from liquid Pb–Li, and presently, this is the reference technology in the European fusion program. Based on the experimental evaluation of the GLC performances in extracting hydrogen isotopes from liquid Pb–Li [35], GLC columns have been designed with reference to the extraction system of the DEMO WCLL, as reported in Table 7 [33].

Table 7. GLC columns for one loop of the OB and the IB, respectively [33].

	OB	IB
height of the vessel, m	9–11	4–5.5
external diameter of the vessel, m	2–3	2–3

5.3. MGLC

The MGLC proposed for tritium extraction from the WCLL blanket is made of tubular porous membranes, as described in Section 4. The MGLC has the same design configuration adopted by the Nb PAV (U-shaped permeator tubes inside a cylindrical vessel). In such a way, most of the thermo-fluid dynamic conditions and design features assumed for the Nb PAV (fluid velocity, pressure drops, etc.) remain also fixed for the MGLC, and the comparison between these two technologies is strictly coherent.

Under the assumption of using stainless steel porous membranes of pore size 2–3 µm, the overall hydrogen mass transfer coefficient of the MGLC determined at 330 °C by the expression (17) is $1.95 \times 10^{-8} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$. Table 8 reports the design characteristics of the MGLC unit needed for processing the Pb–Li of one OB loop; in particular, in this evaluation, the vessel sizes of the extraction unit have been scaled from those of the Nb PAV one. The overall permeation surface area of the entire extraction system, processing the Pb–Li of four loops in OB and two loops in IB, is 2376 m².

Table 8. MGLC unit designed for one loop of the OB.

vessel diameter, m	2
height of tank, m	8
number of tubes	178
tube diameter, mm	9.2
tube length, m	57

5.4. Comparison of the Results

Compared with the other PAV-based extraction systems, the MGLC exhibits significantly lower permeation areas: by about a factor of 2 from 9 with respect to the Nb PAV, respectively working under surface- and diffusion-limited conditions, and by a factor larger than 30 with respect to the V PAV (see Table 9).

Table 9. Comparison of the overall permeation area of the entire extraction systems (four loops in OB and two loops in IB) based on the PAV and MGLC.

	Nb PAV	V PAV	MGLC
permeation area, m ²	21,500 and 5600 ¹	70,900	2376

¹ surface- and diffusion-limited regime, respectively.

The comparison of the sizes and volumes of the tritium extraction units designed for one OB Pb–Li loop based on the PAV, GLC, and MGLC are reported in Table 10. This analysis relies on the previous studies on MGLC testing and the DEMO WCLL tritium extraction systems design [25,33].

Table 10. Comparison among the extraction units designed for one OB Pb–Li loop based on the PAV, GLC, and MGLC.

	Nb PAV	V PAV	GLC	MGLC
permeation area, m ²	3723 and 968 ¹	6840	-	412
unit's height, m	8 and 6 ¹	-	9–11	8
unit's volume, m ³	226 and 75 ¹	34	71	25
technological issues to be verified/solved	Oxidation status of membrane surfaces and optimization of Nb-tubes geometry	Integrity of stainless steel V-weldings	Need of tritium extraction system from helium	Erosion–corrosion and closure of membrane pores by impurities

¹ surface- and diffusion-limited regime, respectively.

The MGLC unit exhibits the smallest volume, significantly smaller than that of the Nb PAV, 2.8 times smaller than that of the GLC, and 1.4 times smaller than that of the V PAV. Both permeation areas and volumes of the extraction units are strictly related to tritium safety and costs: definitively, on these design aspects, the MGLC exhibits better performance than the alternative technologies.

6. Conclusions

Process units based on the concept of Membrane Gas–Liquid Contactor (MGLC) have been studied for the extraction of tritium from the Pb–Li in DEMO WCLL blankets. The MGLC technology has been then compared with the extraction systems presently under study. The MGLC units, consisting of porous stainless steel membranes, exhibit smaller hydrogen mass transfer resistance with respect to PAV (Permeator Against Vacuum) devices.

The preliminary design of MGLC tritium extraction units has been carried out by using the results of a previous experimental campaign. The proposed MGLC design

adopts the configuration of U-shaped permeator tubes inside a cylindrical vessel, the same configuration previously studied for the Nb PAV. Based on this configuration, the permeation area and sizes of a tritium extraction unit of one OB loop of the WCLL blanket have been assessed. The results have been compared with the design features of the analogous units adopting alternative technologies, namely the Nb PAV, the V PAV, and the packed-bed columns (GLC). The MGLC unit has a permeation area smaller by a factor of 2 and 9 than that of the Nb PAV (U-shaped tube configuration), as assessed for the surface- and diffusion-limited regimes, respectively. Moreover, the MGLC unit has a permeation area 30 times smaller than the V PAV (plate configuration). By considering the overall sizes, the MGLC extraction unit exhibits the smallest volume, significantly smaller than that of the Nb PAV, 2.8 times smaller than that of the GLC, and 1.4 times smaller than that of the V PAV.

These preliminary results show that the DEMO WCLL tritium extraction systems using MGLCs are expected to require smaller permeation areas and quicker permeation kinetics than those based on PAV (Permeator Against Vacuum) devices.

Further design analyses are expected to evaluate in detail the aspects related to reliability, safety, and costs of the new technology.

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List of Abbreviations

c_H	hydrogen isotope concentration in the Pb–Li (at. fr. m^{-3})
$c_{H,up}$	hydrogen concentration upstream the metal membrane ($mol\ m^{-3}$)
$c_{H,down}$	hydrogen concentration downstream the metal membrane ($mol\ m^{-3}$)
$c_{H,LiPb}$	hydrogen concentration in Pb–Li ($mol\ m^{-3}$)
d	pore diameter (m)
D	diffusion coefficient of hydrogen through the metal lattice ($m^2\ s^{-1}$)
DCLL	Dual Coolant Lithium Lead
E_S	Sieverts' constant activation energy ($J\ mol^{-1}$)
GLC	Gas–Liquid Contactor
h	overall mass transfer coefficient ($mol\ m^{-2}\ s^{-1}\ Pa^{-1}$)
HCLL	Helium-Cooled Lithium Lead
ITER	International Thermonuclear Reactor
J	hydrogen flow rate ($mol\ s^{-1}$)
k_a	adsorption coefficient ($mol\ m^{-2}\ s^{-1}\ Pa^{-1}$)

k_r	recombination coefficient ($\text{m}^4 \text{mol}^{-1} \text{s}^{-1}$)
k_S	Sieverts' constant (at. fr. $\text{m}^{-3} \text{Pa}^{-0.5}$)
k_S^0	Sieverts' constant pre-exponential factor (at. fr. $\text{m}^{-3} \text{Pa}^{-0.5}$)
Kn	Knudsen number
MGLC	Membrane Gas–Liquid Contactor
P	hydrogen isotopes partial pressure in the gas phase (Pa)
PAV	Permeator Against Vacuum
Pe	hydrogen permeability through the membrane pores ($\text{mol m}^{-1} \text{s}^{-1} \text{Pa}^{-1}$)
P_G	pressure of the gas phase (Pa)
$P_{H,G-L}$	hydrogen partial pressure at the Gas–Liquid interface (Pa)
$P_{H,lumen}$	hydrogen partial pressure in the membrane lumen (Pa)
P_L	pressure of the liquid phase (Pa)
TBM	Test Blanket Module
th	membrane thickness (m)
WCLL	Water-Cooled Lithium Lead
<i>Greek letters</i>	
Γ	surface tension of the liquid metal (N m^{-1})
θ	contact angle of the penetrating liquid over the porous material ($^\circ$)

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