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The current studies on the development of the EU DEMO breeding blanket include among the options the use of liquid Lithium-Lead (17Li-83Pb) as tritium breeder (and multiplier), with different coolants. As the tritium is steadily produced in the blanket during the reactor operation, suitably efficient strategies for the Tritium Extraction System (TES) from the breeder must be developed, allowing a closed fuel cycle in situ and avoiding tritium accumulation in the machine. The Permeator Against Vacuum (PAV) appears to be one of the most promising solutions to achieve this goal. In this paper, the performance of a PAV system is studied by means of different models describing the transport of tritium in the liquid PbLi and in the metallic membrane separating it from the vacuum. The comparison of the results for different membrane materials and size of the device, for a given target efficiency, allows to optimize the PAV design, also taking into account corrosion issues. The approximations and limitations of the adopted models are also addressed.

Keywords: DEMO breeding blanket, Lithium-Lead, Tritium Extraction System (TES), Permeator Against Vacuum (PAV)

I. Introduction

The history of the DEMO breeding blanket designs in the EU is now over 20 years long.[1,2] Several of the options currently under study, namely the Dual-Coolant Lithium-Lead (DCLL), the Helium-Cooled Lithium-Lead (HCLL) and the Water-Cooled Lithium-Lead (WCLL), use liquid Lithium-Lead (17Li-83Pb), PbLi in the following, as tritium breeder (and multiplier).

As the tritium is steadily produced in the blanket during the reactor operation, and as it is needed to close the fuel cycle in situ, while at the same time it cannot be left to accumulate an inacceptable inventory in the machine, suitably efficient strategies for the Tritium Extraction System (TES) from the breeder must be developed.

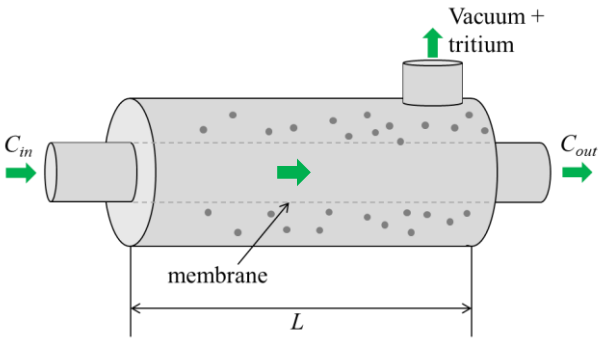
Among the TES options currently under consideration, the Permeator Against Vacuum (PAV), appears to be one of the most promising solutions for the DCLL, HCLL and WCLL Breeding Blanket (BB).[3]

The PAV operating principle is simple: PbLi with a certain concentration of tritium flows in a channel delimited for a given length by a membrane permeable to tritium. Vacuum is maintained on the other side of the membrane, so that the difference between the tritium partial pressure on the two sides of the membrane drives the flux of tritium from the PbLi side to the extraction, leaving a lower tritium concentration at the outlet of the channel. As opposed to the undesired permeation of tritium through the walls in the rest of the circuit, which may lead to safety issues and reduced tritium breeding ratio, here the relatively low solubility and the resulting high partial pressure of tritium in PbLi, together with a suitably permeable and corrosion resistant material for the membrane, are at the basis of the PAV potential for operation.

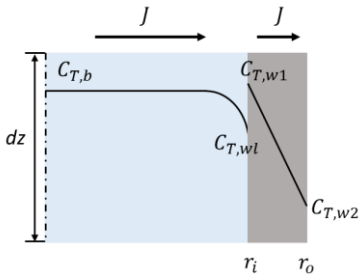
In this paper, two models for the prediction of the PAV efficiency are applied to the HCLL and WCLL. The first model, widely adopted in these kind of analyses,[4,5,6] considers only bulk phenomena in the membrane, while the second accounts for both diffusion and surface effects at the interface membrane-vacuum, as they may lead to an intermediate regime.[7] The analysis, application and systematic comparison of both models, objective of the current work, is motivated by the necessity to understand whether the presence of surface effects may influence the PAV design parameters (i.e. channel length for a prescribed efficiency). Both models are also used to assess by a sensitivity study how the main TES design parameters affect the PAV system size for a given target efficiency requested. The influence of tritium inlet concentration and PbLi temperature in the HCLL/WCLL range of operation is considered, as well as the role of the physical constants entering the model (e.g. Sievert constant and diffusivity).

II. Model description

In Figure 1 a sketch of the PAV geometry is given, as well as a plot of the radial section of the pipe considered by the models adopted in the following, with the identification of the physical quantities of interest.



(a)



(b)

Fig. 1. Schematics of the Permeator Against Vacuum (PAV) system[8] (a) and radial section of the channel, with identification of the unknowns of the problem (b).

II.A Simplified analytical model

A first modelling option, adopted in Refs. 9-11 and described in detail in Ref. 10, focuses on the transport of tritium in the PbLi bulk and in the membrane, neglecting both the presence of helium bubbles mixed with PbLi and all surface effects on both the PbLi and vacuum side, assuming that permeation through the membrane is influenced only by bulk phenomena (diffusion). As a consequence of the latter assumption, Sievert's law $C = K\sqrt{p}$ is applicable and the concentration $C_{T,w2}$ on the vacuum side (p = 0) vanishes. At the interface between PbLi and the membrane the continuity of tritium

partial pressure is usually assumed in the literature, obtaining the following relation through the Sievert constants K_s for the membrane and K_l for PbLi:

$$\frac{C_{T,w1}}{K_s} = \frac{C_{T,wl}}{K_l}. \quad (1)$$

The mass transfer coefficient of tritium in PbLi, appearing in the definition of the flux J (Ref. 10), is taken from Ref. 12. The efficiency is evaluated as $\eta = 1 - C_{out}/C_{in}$. The tritium inlet concentration C_{in} is assumed constant. Calculations are carried out assuming a requested efficiency of 80% (Refs. 4 and 13) and obtaining the corresponding needed length of the system L, for given geometrical (channel hydraulic diameter and membrane thickness) and physical (temperature and pressure) parameters. Due to the linearity of the model, the efficiency is independent on the inlet concentration C_{in}.

II.B Model accounting for surface effects

While the previous model assumes that permeation through the membrane is influenced only by bulk phenomena (diffusion), a model accounting also for surface phenomena is now introduced, as suggested in Ref. 7. In this model, recombination and desorption processes occurring at the membrane-vacuum interface could be as rate limiting as diffusion, and tritium flux J at this interface can be modeled with the relation:[14,15]

$$J = 2k_R \cdot C_{T,w2}^2 \quad (2)$$

where the recombination constant k_R is introduced. The boundary condition at the vacuum side is again $p = 0$, but this does not imply $C_{T,w2} = 0$ as in the previous model, since Sievert's law is not applied here.

Regarding surface effects occurring at the PbLi-membrane interface, to the best of our knowledge there is no model in the literature describing the kinetics of atomic hydrogen in non-equilibrium conditions (i.e. Sievert's law not applicable). Only models for gas-solid interfaces, where non-dissolved atomic hydrogen interacts with a metal, are available.[7,14] Therefore, we assume for the sake of simplicity equilibrium conditions on this surface, thus allowing to use (1), also considering that surface effects occurring at the vacuum side should be dominant with respect to those occurring at the PbLi side. In fact, at the PbLi-membrane interface, no dissociation energy must be provided (tritium is in atomic form in PbLi) and two atoms of tritium are not required to be adjacent in order to be desorbed.

Eq. (2) is coupled with the equations of the previous model[10] in the bulk of PbLi and in the membrane. The result is a set of equations (tritium balance in PbLi, flux in PbLi boundary layer, diffusive current in the membrane, eq. (2), eq. (1) applied at the PbLi-membrane interface) that cannot be solved analytically, and, more importantly, the inlet concentration C_{in} now affects the efficiency of the PAV. Since the HCLL and WCLL designs are characterized by different values of C_{in}, this parameter is considered in the parametric analysis presented in the next sections.

III. Results

III.A Comparison of different membrane materials

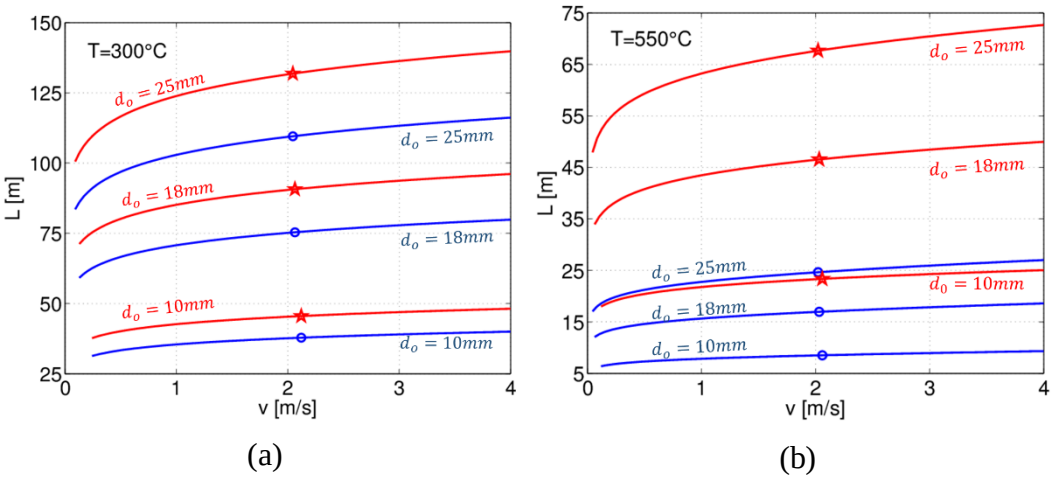
The first set of results presented focuses on the comparison of the choice of the membrane material, performed adopting the simplified analytical model, in order to identify as a first step the membrane with the best performance. The two main options, based on previous analysis,[11] are Fe and Nb. Nb has a higher tritium permeability with

respect to Fe, thus allowing to improve the extraction performance, but it tends strongly to oxidize at high temperature. For this reason, a Pd coating should be envisaged, although it is not considered in this preliminary design stage. However, the Pd coating should not affect our results significantly, because of its high permeability[16] and small thickness (in the range of μm).

The data adopted for the comparison are summarized in Table I. Two temperature values are chosen, assuming as minimum the design outlet T of PbLi in the BB, and then considering an upper limit, still compatible with the material mechanical properties, to increase efficiency. The channel hydraulic diameter and PbLi flow speed are varied in a reasonable range: the minimum speed ensures Re = 10^4 and therefore a turbulent regime, while the upper limit is still compatible with corrosion constraints. A parametric analysis on the diffusivity D_l is also performed, assuming the minimum and maximum value available in the literature, since the range of variation of this parameter covers several orders of magnitude. The same approach is also adopted for the choice of the Sievert's constant, bearing in mind that the K_l value from Ref. 20 is considered to be the most reliable and conservative.

TABLE I: Input data adopted in the analytical model calculations for the comparison of Fe and Nb as membrane materials.

η [-]	0.8	
Membrane thickness [mm]	0.5	
T_{PbLi} [°C]	300	550
ρ_{PbLi} [kg/m^3]	$9.8 \cdot 10^3$ (Ref. 17)	$9.5 \cdot 10^3$ (Ref. 17)
μ_{PbLi} [Pa s]	$2.2 \cdot 10^{-3}$ (Ref. 17)	$1.0 \cdot 10^{-3}$ (Ref. 17)
D_l [m^2/s]	$8.6 \cdot 10^{-10}$ (Ref. 18)	$4.8 \cdot 10^{-9}$ (Ref. 18)
	$6.5 \cdot 10^{-10}$ (Ref. 19)	$9.9 \cdot 10^{-10}$ (Ref. 19)
K_l [mol/m^3/Pa$^{0.5}$]	$2.8 \cdot 10^{-2}$ (Ref. 20)	$6.3 \cdot 10^{-2}$ (Ref. 20)
	$9.9 \cdot 10^{-4}$ (Ref. 21)	$1.1 \cdot 10^{-3}$ (Ref. 21)
P_{Nb} [mol/m/s/Pa$^{0.5}$]	$6.2 \cdot 10^{-6}$ (Ref. 16)	$6.5 \cdot 10^{-7}$ (Ref. 16)
P_{Fe} [mol/m/s/Pa$^{0.5}$]	$1.6 \cdot 10^{-11}$ (Ref. 16)	$1.5 \cdot 10^{-10}$ (Ref. 16)



(a) (b)

Fig. 2. PAV length as a function of PbLi flow speed adopting Nb as membrane material for different hydraulic diameters d_o. K_l from Ref. 20. Circles: D_l from Ref. 18; stars: D_l from Ref. 19. (a): operation temperature T = 300 °C; (b): T = 550 °C .

The use of a Fe membrane turns out not to be practical (not shown), since very long (from hundreds to thousands of meters, depending on temperature and speed) channels are needed to achieved the required efficiency. [11]

In the case of Nb, see Fig. 2, increasing the diameter affects the results because the ratio between the cross section and wetted perimeter increases. The needed channel length shows a non-linear behavior with respect to speed, typical when transport across PbLi is rate limiting. The PAV shows better performances, i.e. a shorter channel is sufficient, at higher temperatures, due to the increased tritium diffusivity in PbLi.

III.B Comparison of modelling options for the Nb membrane

Having identified Nb as the best option for the membrane material, we now compare the results obtained with the model not accounting for surface phenomena to those obtained with the model accounting for surface effects. To evaluate the

permeating tritium flux in this case, the value of k_R in (2) is necessary. k_R can be expressed as:[13,21]

$$k_R = \sqrt{\frac{2}{3}} \frac{1.3 \cdot 10^{24}}{N_A \cdot K_{S0}^2 \cdot \sqrt{T}} \exp\left(\frac{2(E_S - E_C)}{RT}\right) \quad (3)$$

where K_{S0} = 0.127 mol m^{-3} Pa$^{-0.5}$ (Ref. 21) is the pre-exponential coefficient of Sievert constant, E_S = –34 kJ mol^{-1} (Ref. 22) is the heat of solution of tritium in PbLi, E_C = 40 kJ mol^{-1} is the activation energy for dissociative adsorption (considering the more conservative value from Ref. 22) and N_A is the Avogadro number. The term $\sqrt{2/3}$ is introduced to take into account that the formula for k_R was originally defined for deuterium.

Since the model depends on the inlet concentration of tritium, we consider two different ranges: 20-30 Pa for HCLL and 50-80 Pa for WCLL. Starting from the inlet pressure of tritium in PbLi, the corresponding concentration C_{in} is obtained applying Sievert's law.

To understand how C_{in} and k_R affect the system performance, a parametric analysis is carried out, see Fig. 3. The increase of the inlet concentration in the range 20-80 Pa leads to a non-negligible reduction of the PAV length (up to 15%), especially if the lower operating temperature is assumed. This is due to the imposed relation (2) between the tritium flux and the concentration, which is affected by its inlet value. In fact, if C_{in} increases, consequently also $C_{T,w2}$ increases. Since the tritium flux J is proportional to $C_{T,w2}^2$, its increase will results in a reduced length of the PAV system needed to achieve the prescribed 80% efficiency. Instead, results obtained with the first model do not depend on C_{in}, as already commented in Section II.A.

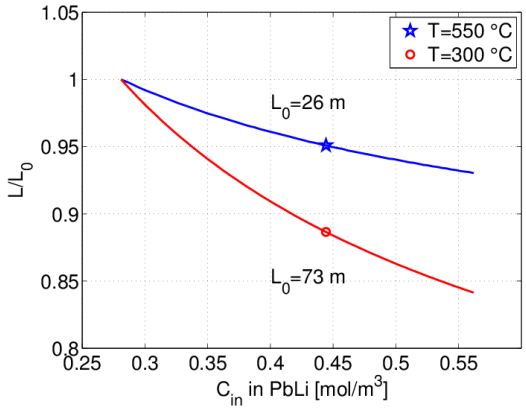
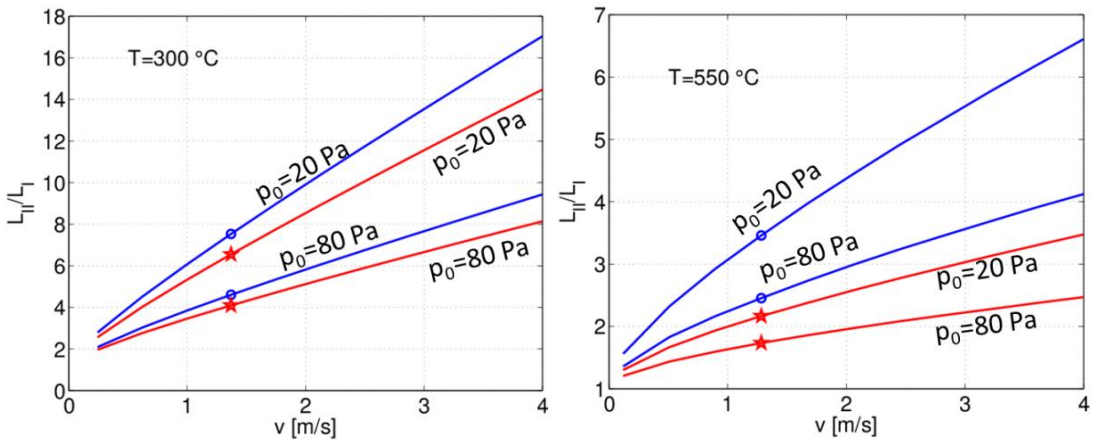


Fig. 3. Normalized PAV length as function of inlet concentration. Data adopted: $d_0=10$ mm; $v=0.5$ m/s; K_l from Ref. 20 and D_l from Ref. 19, conservatively.



(a) (b)

Fig. 4. Comparison of the Nb membrane PAV system length as a function of PbLi flow speed foreseen by the two models (I = simplified, II = including surface effects), considering two different values of tritium inlet pressure, two temperatures of operation and d_o = 10 mm. K_l from Ref. 20, conservatively. Circles: D_l from Ref. 18; stars: D_l from Ref. 19. (a): operation temperature T = 300 °C; (b): T = 550 °C

In Fig. 4 we compare the needed PAV system length foreseen by the two models illustrated above. Fig. 4 demonstrates that surface phenomena play a non-negligible role in tritium transport, affecting the resulting PAV length, which is especially true when

the resistance associated to the diffusion in PbLi is lower, i.e. at higher flow speed and assuming a higher value of D_l (resulting in an increased mass transfer coefficient), while the resistance associated to recombination is independent of the flow speed, and becomes therefore more dominant. In conclusion, all results show that transport in the membrane is a limiting factor and needs to be considered in the system design phase.

The influence of the value of the Sievert constant is also analyzed. The value of K_l affects the solution of the problem since it is used in the evaluation of the inlet condition C_{in} and it also appears in Eq. (1). The assumption of a smaller K_l implies a reduced inlet concentration, resulting in a longer PAV length needed (see Fig. 3). However, the use of a smaller Sievert constant[20] in Eq. (1) at the PbLi-membrane interface results in a more relevant reduction of the PAV length with respect to the previous effect. Figure 5, comparing two limiting values of K_l proposed in the literature, shows that the uncertainty on K_l strongly affects the PAV length, especially for increasing flow speed.

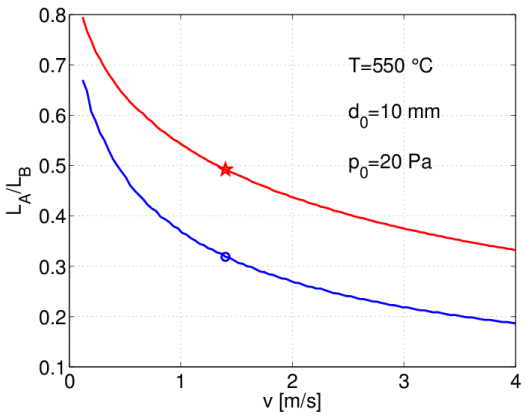


Fig. 5. Comparison of the PAV system length with Nb membrane for two different values of Sievert constant. L_A stands for the length computed using K_l from Ref. 21, while L_B with K_l from Ref. 20. Circles: D_l value from Ref. 18; stars: D_l value from Ref. 19.

IV. Conclusions and perspective

In this work an analysis of the PAV system is presented, adopting two different models for tritium transport. The first model neglects surface phenomena at material interfaces, assuming diffusion as the limiting phenomenon, while the second takes into account both bulk diffusion and surface phenomena. Both models require a set of parameters characterizing PbLi and the membrane material.

A sensitivity study was performed with respect to the main design parameters (inner PAV channel diameter, PbLi speed and inlet temperature), and input parameters (tritium solubility and diffusivity in PbLi, PAV inlet concentration), showing their impact on the PAV length for a prescribed efficiency of 80%.

The results show that the most suitable material for the membrane is Nb, and the comparison of the two modelling options shows that surface phenomena are a limiting factor, changing the length of the PAV channels up to a factor 3 in the most conservative scenario, and therefore cannot be neglected, .

Moreover, both the Sievert constant and the diffusivity, which are known only with high uncertainty, strongly affect the system design. Assuming the most conservative values for these two constants in the simplified model, PAV length becomes few tens of meters, a value that is much different from the few meters obtained by previous studies[4,5,6]. The reason for this difference is clearly associated to the choice of the value of these parameters.

Both models lead to a value of the channel length of several tens of meters, in order to reach the design extraction efficiency of 80%. Therefore, to limit the geometrical size of the PAV system, the possibility to arrange the pipes in concentric spirals is currently under consideration. In this perspective, the potential effect of the

different geometry on tritium permeation will be assessed, and other engineering aspects influencing the design, such as the pressure losses along the channels, will be evaluated. An experimental campaign on a mock-up test of the PAV system is foreseen to validate the computational results.

Acknowledgments

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