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Design of a Permeator Against Vacuum for Tritium extraction from eutectic lithium-lead in a DCLL DEMO

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One of the most important issues in future fusion power plants is the extraction of tritium generated in the breeders in order to achieve self-sufficiency. When the breeder is a liquid metal one of the most promising techniques is the Permeation Against Vacuum, whose principle is based on tritium diffusion through a permeable membrane in contact with the liquid metal carrier and its further extraction by a vacuum pump. A conceptual design of permeator has been developed, taking into account the features of a DEMO reactor with a Dual Coolant Lithium Lead (DCLL) breeder blanket. The study is based on the analysis of different membranes and geometries aiming at the overall efficiency (extraction capability) of the device, as well as its compatibility with the breeder material. The permeator is based on a rectangular section multi-channel distribution where the liquid metal channels and vacuum channels are alternated in order to maximize the contact area and therefore to promote tritium transport from the bulk to the walls. The resulting permeator design has an excellent estimated extraction efficiency, of 80%, in a relatively compact device.

Keywords: fusion, DCLL, tritium, permeation against vacuum, membrane

1. Introduction

The European Union is taking a range of pre-conceptual research and development activities towards the realization of a demonstration power plant called DEMO. The fuel to be used in this near term reactor is deuterium and tritium, since its reaction presents a high fusion cross section. Knowing that there is no natural source for tritium production, self-sufficiency becomes one of the most important functions in a fusion power plant.

Four breeder blanket (BB) concepts are being developed in order to accomplish this requirement, among other functions [1]. Tritium production in the BB is made by irradiating a lithium compound with the neutrons generated in the fusion reactions, by incorporating a material suitable to multiply neutrons, like the lithium-lead (PbLi) alloy.

One of the most promising concepts of BB is the Dual Coolant Lithium Lead (DCLL), which uses liquid PbLi as primary coolant, tritium breeder and neutron multiplier. Helium is used for cooling the wall exposed to the plasma and the supporting structures. A closed PbLi loop connects the BB to a heat exchanger for power extraction. The most important components in the loop are the pump, a purification system and a Tritium Extraction System (TES). The last is used for extracting tritium from the liquid metal, thanks to the use of a Tritium Extraction Unit (TEU), to be finally pumped to the tritium plant [2].

There are different TEU under study; among them Permeation Against Vacuum (PAV) has been identified as a promising technique for tritium extraction from flowing PbLi in DEMO. PAV is expected to provide the necessary extraction rate with high efficiency, minimizing the tritium mobile inventory concentration in the PbLi loop and the amount of tritium leaked to other systems.

In this work, a study to develop a PAV involving the material selection and the performance to obtain the highest possible recovery has been carried out. It has been fulfilled by aiming at the requirements imposed by DEMO design when a DCLL BB is considered. The paper is organized as follows: Section 2 describes the most important aspects of the PAV technique; in Section 3 an analytical expression for the efficiency of a given permeator is presented; a particular case of tritium extraction for a DEMO DCLL is studied in Section 4; a proposal of PAV for DCLL as well as a discussion on the results is presented in Section 5. Finally, conclusions are drawn in Section 6.

2. Permeation Against Vacuum

2.1 Description of the technique

The concept is based on tritium diffusion through a permeable membrane thanks to a pressure gradient established between the two membrane surfaces. As is shown in fig. 5 from [3], one of them is in contact with the PbLi, where the tritium has a low solubility. Because of the reduced activity of lithium in this alloy, there is no risk of hydride formation [4]. The other surface is connected to a vacuum system. When tritium reaches the vacuum side, is extracted by a vacuum pump and sent directly to the tritium plant of the fusion reactor.

The processes taking place in the permeator are those related with the tritium transport in the liquid and with the permeation through the membrane. Thus, the permeation is influenced at least by four factors: diffusivity, solubility, surface absorption and desorption, and hydrogen-crystallographic defects interaction. The experiments are usually attempted to be made in

conditions of negligible influence of defects but having no impact on permeability [5]. In this case the permeation has two limit regimes: the Diffusion Limited Regime (DLR) and the Surface Limited Regime (SLR). In the present study, a permeation limited by diffusion is assumed because when the surface processes are fast enough it is possible to take advantage of the pressure gradient generated by vacuum in one side of the membrane.

2.2 Membrane

A qualitative study on different membranes has been performed. The proposed materials must fulfill the following requirements:

- High permeability (Φ) to tritium
- Withstand high temperatures
- Corrosion resistance Mechanical resistance
- Malleability

For instance, the use of Aluminum, Nickel, Titanium, Zirconium and Copper should be avoided due to their low tritium diffusivity and bad compatibility with PbLi [4, 6].

Table 1 summarizes the list of materials selected for the study and their respective coefficient of permeability (Φ) to tritium at 550 ºC (materials marked with * have been studied at a temperature of 350 ºC and their permeability values extrapolated to 550 ºC for the case of this study). There is a lack of data for tritium and an extrapolation from hydrogen values has been performed. Thus, the classical mass dependence has been used, that is for diffusivity the pre-exponential factor is proportional to $1/\sqrt{m}$, m = mass of diffusing atom; for the solubility there is no classical mass dependence [7].

Table 1: Material coefficient of permeability at 550 ºC (Φ, mol /m/s/$Pa^{0.5}$)

material	Φ	material	Φ
Niobium[8]	1.41E-7	α-Fe [11]	1.48E-10
Vanadium[8]	8.72E-8	Pd/SiO_2/V [12]	2.55E-9
Tantalum[8]	6.41E-8	Pt/SiO_2/V [12]	3.14E-12
$V_{90}Co_{10}$ [8]	6.93E-8	$Nb_{30}Ti_{35}Co_{35}$ [13]*	1.52E-8
VCr_4Ti_4 [8]	7.51E-9	$Nb_{40}Hf_{30}Co_{30}$ [14]*	2.86E-8
Palladium[9]	1.29E-8	SS316 [15]	1.09E-11
Platinum[9]	2.27E-12	SS304 [15]	1.35E-11
Pd/25%Ag[10]	1.28E-8		

An evaluation of the possibilities to use each material shown in table 1 for the PAV manufacturing is depicted in section 5.

2.3 Geometry

The key point of this study is to design a structure whose configuration maximizes the contact area between the membrane and the flowing PbLi to promote tritium transport from the bulk to the wall. The design must ensure a uniform flow distribution across the permeator and minimize the pressure drop.

Different PAV designs can be found in the literature, mostly based in cylindrical configurations [3, 16 and 17], aiming at different shapes. Some of them are still under theoretical research [16, 17] and their implementation into a power plant must be improved because they are based on a high number of elements, length and PbLi velocity.

In the present paper a different configuration has been proposed in order to

- enhance the actual extraction capability
- facilitate the manufacturing process
- improve the vacuum performance
- minimize the size with regard to the available space

A squared multi-channel component where the PbLi flowing channels and vacuum channels are alternated has been evaluated, as will be shown in section 4.

3. Estimation of the PAV efficiency

One of the critical points in the development of a PAV is the efficiency, η, which can be defined as:

$$\eta = \frac{C_i - C_T}{C_i} = 1 - \frac{C_T}{C_i} \qquad (1)$$

Eq. 1 shows the definition of the efficiency as the ratio between the tritium not permeated (difference of concentration at the inlet (C_i) and the outlet (C_T)) and the tritium concentration at the inlet.

The corresponding analysis in terms of the tritium transport and permeator geometry has been made according to the relations described in [17-20]. Thus, an expression of the efficiency for a squared channel in a PAV has been derived, as shown in eq. 2.

$$\eta = 1 - \exp\left[-\frac{2 \cdot K_T \cdot L}{v \cdot h} \cdot \frac{\frac{\Phi}{z \cdot K_L \cdot K_T}}{1 + \frac{\Phi}{z \cdot K_L \cdot K_T}} \right] \qquad (2)$$

K_L is the tritium solubility in the liquid metal, Φ the tritium permeability through the membrane, K_T the mass transport coefficient and v the PbLi velocity in the channels. Concerning the geometrical parameters L is the PAV length, h the distance between channels and z the wall thickness.

The properties of PbLi recommended for tritium transport calculations in tritium breeding blankets are given in [21] from where tritium solubility calculated by Reiter has been selected. Mass transport coefficient depends on tritium diffusivity in the liquid, the distance between membranes (since it is the representative dimension of the flowing system) and the Sherwood number. This last parameter is correlated with fluid conditions through the function defined by Linton & Sherwood [22] which is the most suitable for this kind of fluid [23] under the conditions showed.

The dependence of the PAV efficiency with different parameters will be evaluated over a wide range in the section 4.

4. Design results of a PAV for DCLL DEMO

The input parameters used for this study (total mass flow rate of 30000 kg/s and **550 ºC** temperature) have been taken from the DCLL concept proposed in [2]. The considered DEMO has 16 sectors, each one having a PbLi loop and, therefore, one TES per sector. The mass flow rate will be spread in the 16 PbLi loops leading to 1875 kg/s per TES, which means a total number of 16 PAV.

The maximum allowable velocity in the PAV channels is set at 1 m/s in order to minimize the pressure drop in the loop and the corrosion rate in the permeator. In addition, low PbLi velocities imply higher efficiency values, as shown in eq. 2.

The bulk velocity in each channel (eq. 3) is fixed by the PbLi mass flow rate, $\dot{m}$, the cross section area, $a \cdot h$ (where a is the PAV width), the total number of channels, N, and the liquid metal density, ρ_{PbLi}.

$$\dot{m} = v \cdot h \cdot a \cdot N \cdot \rho_{PbLi} \qquad (3)$$

The role of the permeator length, height of the flowing channels or PbLi velocity in the efficiency is intuitive from eq. 2. If L is quite large the efficiency will approach 1. The same occurs if the PbLi is at almost stagnant conditions or the channels are very narrow. However the combination of these parameters at realistic values, together with the thickness of the membrane, should be studied in detail.

Therefore, a detailed analysis of the different geometrical parameters which take part in eq. 2 has been performed. The main objective is to optimize the PAV efficiency with reasonable dimensions of the device, maintaining the velocity under 1 m/s, as mentioned before.

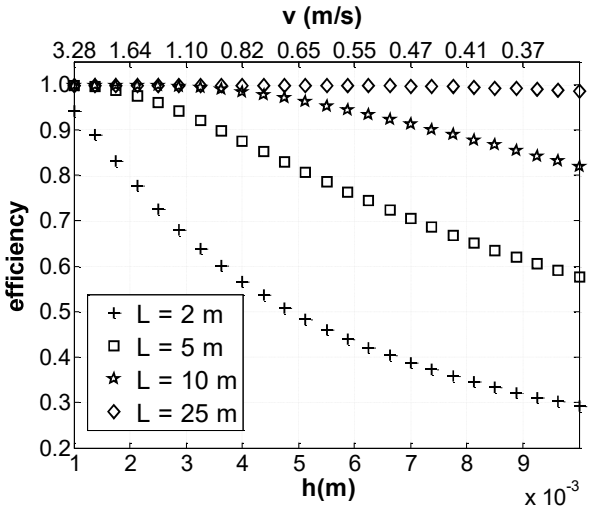

Fig.1. Relation between efficiency, distance between membranes and velocity. (a = 0.75 m, z = 1 x 10^{-3} m, N = 80, Φ = 1.28 x 10^{-8} mol/m/s/$Pa^{0.5}$)

4.1 Length

Figure 1 shows the effect of the permeator length on the PAV efficiency for a distance between membranes ranging from 0.002 to 0.01 m. Therefore according to eq. 3 the velocity of the liquid metal varies between 0.32 and 3.27 m/s. It can be deduced that, when the length is decreased, a lower h is needed to achieve a fixed value of efficiency. This is because the tritium has less surface to permeate, being compensated by reducing the distance of diffusion in the bulk. The permeation is limited by mass transport so the efficiency is improved by reducing the distance between membranes due to its relation with the mass transport coefficient [22].

Thus, a value of 5 m length will be chosen in order to have a distance between plates of about 5 x 10^{-3} m which gives a velocity below 1 m/s with an efficiency of 80% (fig. 1).

4.2 Membrane thickness

From eq. 2, when the membrane thickness is increased the permeation through the membrane becomes more difficult and, consequently, the efficiency decreases. Fig. 2 shows the dependence of the efficiency with the thickness, for different channel heights and number of channels. As can be seen at lower values of channel height the variation of z is more accentuated. The permeation is being more affected by the membrane thickness than by the distance between membranes, in other words, the permeation is becoming limited by the diffusion through the membrane. On the other hand, one can reduce the flow velocity for a fixed mass flow rate by increasing the cross section, i.e. by increasing the value of h and N. Thus the variation is less accentuated because the tritium permeation is limited by mass transport in the liquid metal.

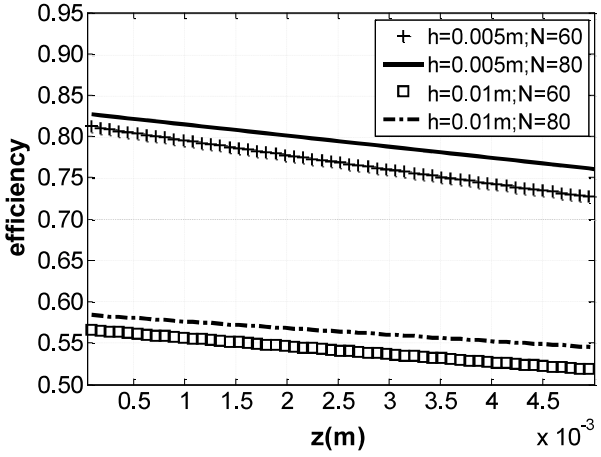

Fig.2. Relation between wall thickness and efficiency. (a = 0.75 m, L = 5 m, Φ = 1.28 x 10^{-8} mol/m/s/$Pa^{0.5}$)

Then a value of 10^{-3} m thickness is selected for the following calculations because it gives an acceptable efficiency and lower values could lead to strength problems. Considering this value and a total length of 5 m, fig. 2 shows that 80% efficiency is achieved for a distance between membranes of 5 x 10^{-3} m.

4.3 Channels width

Other factor affecting the PAV efficiency and its manufacturing is the channel width. The permeator effectiveness is improved when the value of width is increased because of the expansion in the channel cross section, which makes the fluid runs slower, and in the contact area. Furthermore when the number of channels and the distance between membranes are higher the velocity is also decreased. However, in this case the efficiency is not enhanced because the value of *h* has a great influence through the tritium diffusion from the bulk to the wall.

The total width is set at 0.8 m because lower values need larger number of ducts to maintain the velocity below 1 m/s.

4.4 Number of flowing ducts

From eq. 3 the relation between the number of flowing channels and the efficiency is easily followed. When varying the number of flowing channels for a fixed mass flow rate the velocity is also modified, therefore – it changes the permeator efficiency. When the number of channels is high so that the mass flow is spread in a high number of ducts, the lower velocity leads to a higher extraction rate. However, the influence of this magnitude in the efficiency is not relevant compared to other parameters since a reduction of 50 % of channels (and therefore an increase of 100 % in velocity) only represents a 5% loss of efficiency. Thus, a number of 60 flowing ducts (121 considering also the vacuum ducts) is selected in order to design a permeator with a PbLi velocity below 1 m/s, leading to a squared structure.

4.5 Sensitivity analysis

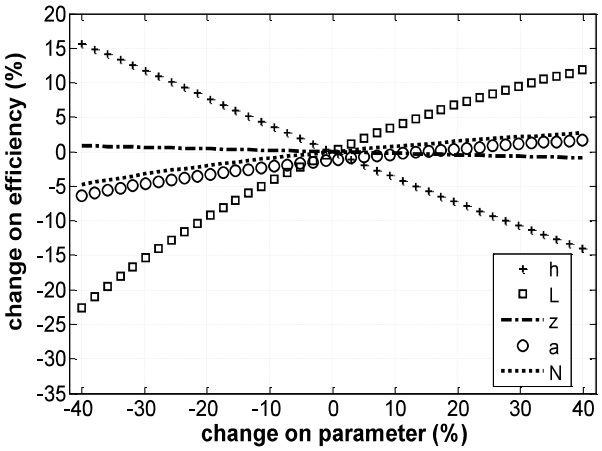

Fig.3. Dependence of efficiency with the parameters studied. (h = 5 x 10^{-3} m, L = 5 m, z = 1 x 10^{-3} m, a = 0.80 m, N = 60, Φ = 1.28 x 10^{-8} mol/m/s/$Pa^{0.5}$)

The importance of all these parameters on the theoretical efficiency is depicted in fig. 3. They have been varied keeping the rest constant and with the values given in the figure. It is clear that the most critical parameters are the height and the length. As expected, an increase in the distance between membranes implies a decrease in efficiency, even though when the velocity is being reduced. For instance, an *h* increment of 40% leads to a η reduction of 15%. On the other hand, an increase of 40% in *L* shows an improvement of 12% in the efficiency. However, an impact of a 10% variation on *L* or *h* on the overall dimensions of the PAV is different for each parameter. Taking into account that one of the premises was to have a compact device, the length will be maintained as low as possible and, hence, the driver parameter is the distance between membranes.

5. Discussion on the proposed design

The parametrical analysis of the PAV efficiency driven by eq. 2 leads to a design whose geometrical characteristics, shown in table 2 and fig. 4, have been fixed aiming at the need of maximize the contact area between the liquid metal and the membrane. It is important to notice that these values have been optimized for the PbLi conditions in a DCLL DEMO reactor.

Table 2: Geometric characteristics

Parameter		Value
Width	a	0.800 m
Channel height	h	0.005 m
Length	L	5.000 m
Membrane thickness	z	0.001 m
Number of channels	N	60 (121)
Velocity	v	0.818 m/s
Total height	H	0.727 m
Total membrane area in contact with PbLi		480 m^2
Structural material volume		0.434 m^3
Volume of PbLi		1.050 m^3

Feasibility of potential materials to be used as PAV membrane (table 1) has been studied. For a fixed PAV geometry (table 2) most of them have high efficiencies (fig. 5), showing a clearly exponential relation following eq.3. A saturation limit is observed at 1.25 x 10^{-8} mol/m/s/$Pa^{0.5}$ from which an increase on permeability does not improve the efficiency. This is the midpoint between the two processes that limit the permeation: mass transport in the PbLi and membrane processes. Thus, at high values of permeability the tritium transport in the liquid is the governing step because membrane processes are faster. However, not all of them will withstand the conditions to which they shall be subjected.

Compounds with platinum are not viable since its permeability is quite low, therefore presenting efficiencies lower than 0.01. The same situation is applied to stainless steel (η = 0.03), which additionally presents high corrosion rate under PbLi flows [24], and to iron with an efficiency of 25%.

Some of the composite materials showing high efficiency (for instance $Pd/SiO_2/V$, $Nb_{30}Ti_{35}Co_{35}$, $Nb_{40}Hf_{30}Co_{30}$; $\eta \approx 0.80$) could have many issues in the fabrication process e.g. an exact control of the Nb-based compounds microstructure is mandatory in order to avoid hydrogen impermeable alloys [13], and their compatibility with PbLi is unknown.

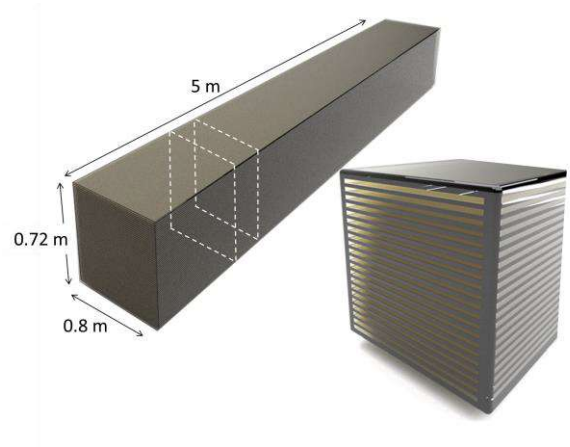

Fig.4. Left) Overall PAV design showing the length (L), width (a) and total height (H). Right) Detailed view of PbLi flowing channels (not real dimensions).

The most used compound for hydrogen purification systems is palladium (η = 0.80) [25]. This compound and its silver alloy (η = 0.80) seem to be adequate to be used as the permeator membrane but they have not been tested in lithium lead yet. On the other hand, Tantalum, Niobium and Vanadium (η = 0.81) have great permeability characteristics and good compatibility with the eutectic lithium lead alloy in static conditions [26]. For this reason these materials are selected as the most suitable membranes for the PAV manufacturing. However, since the database of corrosion resistance to this alloy is almost limited to steels and to some metals in static conditions the compatibility with PbLi is a weak point that needs to be improved in the future.

Due to the high price, it would be convenient to test some other compounds not tested on PbLi yet but showing a high efficiency, for instance the vanadium alloys ($V_{90}Co_{10}$, VCr_4Ti_4; η ≈ 0.80).

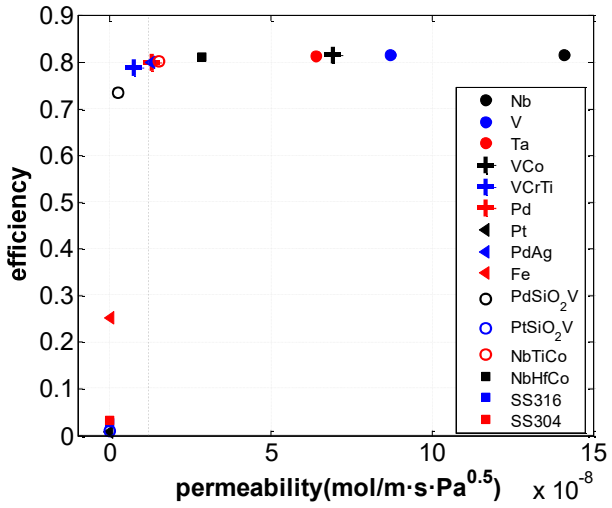

Fig.5. Relation between permeability and efficiency for a fixed PAV geometry. (h = 5 x 10^{-3} m, L = 5 m, z = 1 x 10^{-3} m, a = 0.80 m, N = 60)

In spite of these encouraging perspectives, some issues must be taken into account. Firstly, calculations have been made assuming a diffusive-limited regime. The disfavored case where the process is limited by surface should be also studied in order to avoid the waste of the pressure gradient generated by the vacuum. Secondly, various aspects must be highlighted regarding the membrane fabrication: the raw material of the compounds suitable to be used as membrane (Ta, Nb, V) can be an issue if the membrane production is not allowed due to the geometrical requirements; the purity of the material is also important since it can lead to a different permeation rate or to a change in the mechanical properties. The membrane purity can also affect the compatibility of the material with the PbLi. In addition, an evaluation of the manufacturing process aiming at the material availability and to the mechanical performance (e.g. against thermal loads) should be made.

Finally, the partial pressure of tritium in the whole mass flow rate for DCLL-BB is really low (about 110 mPa for a TBR = 1.13, 30000 kg/s and 550 ºC), which does not assure a DLR. It is assumed that, after some recirculation in the PbLi loop, the tritium partial pressure will be high enough to reach DLR operational conditions. Since the flux through the membrane depends on the pressure gradient and in order to extract most of the tritium, tritium concentration in the vacuum side is assumed to be negligible.

6. Conclusions

Transport processes in a rectangular channel occurring during tritium extraction from PbLi have been studied to obtain an expression for the efficiency of a permeator against vacuum. A diffusive limited regime has been considered. This relation, which depends on geometrical parameters and magnitudes involved in the permeation process, is the basis for the design and optimization of a PAV.

The efficiency dependence with the different geometrical parameters of the permeator has been evaluated over a wide range. A high influence of the distance between membranes and velocity has been observed. However, the limitations on mass flow rate and in order to reduce as much as possible the PAV dimensions it has been necessary to keep the velocity just below 1 m/s. Thus, the channel height must be in the order of a few millimeters to accomplish a high efficiency. In this case, 5 x 10^{-3} m is enough for a well performance of an 80% with a square-shaped PAV with 5 m length, 0.80 m width and 0.72 m high.

Different membrane materials have been studied taking into account requirements on tritium permeability, PbLi compatibility, working temperature and malleability. There are various materials offering similar efficiencies for this design (Palladium-Silver, Vanadium, Tantalum…). However, the high price and the unknown compatibility with PbLi reduce the possibilities.

The viability of this technique must be tested in a experimental facility to confirm its application as tritium processing solution in fusion reactors.

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