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Dimensioning of ideal membrane cascade systems for the separation of binary gas mixtures for nuclear fusion applications

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Abstract

In the DEMO reactor highly efficient separation systems (> 80%) are required for the recovery of tritiated species and therefore ensure a continuous re-fuelling of the plasma. Examples of such systems are the tritium extraction system for the separation of Q₂ from He (as in the case of the European Helium Cooled Pebble Bed blanket) and the separation of DT molecules from plasma enhancement gases (e.g., Ar, Xe, ...) at the exhaust of the vacuum vessel (Q = H, D, T). For these systems, porous inorganic membranes have been proposed. However, due to the similar sizes and/or masses between the molecules, the separation of the porous membranes can be rather limited. Therefore, to comply with the performance requirements desired in a fusion reactor, multi-stage membrane systems are required. In this paper, a numerical algorithm is presented to estimate the minimum number of stages required for a relevant range of selectivities and performance requirements (i.e., enrichment factor *EF* and recovery fraction *RF*). The results show that the number of stages greatly depends on the membrane's selectivity α , *EF* and *RF*. For instance for $\alpha = 2 \rightarrow 10$ the number of stages decreases from 16 to 6 for *EF* = 20 and *RF* = 90%. In addition, the injection flow was found to have a significant impact in the overall membrane's surface area and power consumption. At the last part of this paper the feasibility and viability of a membrane cascade system are discussed regarding to its size, power consumption and impact on the tritium inventory.

Keywords: gas separation, inorganic membranes, membrane cascade, fuel processing, nuclear fusion

1. Introduction

Mature separation technologies relying on cryogenic distillation or adsorption columns are planned to be used at ITER. For instance, cryogenic columns operated below 30 K for the separation of hydrogen isotopologues (i.e., H₂, HD, D₂, HT, DT, T₂) are foreseen to be used in the isotope separation system of the ITER fuel cycle [1, 2]. Moreover, adsorption columns relying on ZrCo getters to trap the hydrogen isotopologues and zeolite beds to remove water at 298 K are proposed to be used in

the tritium extraction system of the HCPB test blanket module [3]. Although these technologies have a high readiness level, they also present some drawbacks. On the one side, the use of cryogenic temperatures lead to high operation costs, especially because large flows (i.e., purge gas) will be required for fusion reactors. On the other side, adsorption columns need to be regenerated and redundant systems are required to be operated alternatively which raises safety concerns due to the high-inventory of tritium [4]. As a matter of fact, trapping technologies are inherently non-continuous separation systems. Beyond the concerns regarding to tritium inventory, it is also an important drawback for the DEMO machine which has to demon-

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strate the tritium self-sufficiency (e.g., reactor-relevant operation times) [5]. Tritiated species that remain trapped are not available to fuel the plasma before the regeneration of the traps. Therefore, membrane technologies have been considered for the past decade as a viable and cost-effective alternative for the separation of gases in different applications [6]. Pd/Ag membranes (and membrane reactors) were also successfully developed for fusion applications since more than 20 years to continuously separate gas species in the fuel cycle of the fusion reactors [7–9]. More recently, inorganic porous membranes were also proposed for two additional applications in the European DEMO fuel cycle. First, a combination of porous inorganic membranes (e.g., zeolite-ceramic) and palladium-based membrane reactors have been considered for the tritium extraction system of the solid HCPB blanket [4]. Furthermore, porous ceramic membranes (e.g., α -Al₂O₃) have been recently proposed for the recovery of unburnt DT molecules mixed with plasma enhancement gases (e.g., Ar) coming from the torus exhaust [10]. The use of membranes technologies may ensure lower tritium inventories, improve the management of tritiated species in the fuel cycle, and guarantee continuous process operation [11]. In addition, its modularity and potential lower costs for operation are also a great benefit [12]. However, in these applications the separation efficiencies which can be achieved are usually limited since the molecules have similar diameters (e.g., the kinetic diameters of He and H₂ are, respectively, 0.26 nm and 0.289 nm). Therefore, a membrane cascade must be considered instead to achieve the separation performance required.

The number of stages integrating the membrane cascade is highly dependent on the so-called separation factor (of each stage). In addition, the required surface area for each stage is dependent on the permeances of the gases and on the feeding flow. Furthermore, since separation is driven by the pressure difference across the membrane, compressors are required between stages which impact the total power required to operate the cascade. Last but not least, all these quantities are also influenced by the overall separation requirements. The estimation of the number of stages, their surface area and the power

required to operate them are crucial to discuss the feasibility and the interest of membrane cascades at reactor scale. At the best of our knowledge in the literature there is no study which presents a comprehensive analysis of the impact of these parameters (e.g., selectivity) on the cascade dimensioning for fusion applications. Thus, a model and a sensitivity analysis to support the dimensioning of membrane cascades was developed in the perspective of gas separation for fusion application, and the results are discussed with regards to feasibility and cost-effectiveness.

2. Mathematical description of an ideal membrane cascade

2.1. Definition of an ideal cascade

The classical membrane cascade arrangement consists of several separation stages placed in series, as depicted in figure 1. In this configuration, a feeding flow $F_{f,inj}$ is routed to the so-called injection stage. Due to the pressure difference across the membrane, the feed flow of each stage is divided into permeate and retentate flows, that are used to feed the next and previous stages, respectively. Overall, the flow $F_{f,i}$ feeding stage i is the sum of the retentate flow $F_{r,i+1}$ from stage $i + 1$ and the permeate flow $F_{p,i-1}$ from stage $i - 1$, as presented in equation (1).

$$F_{f,i} = F_{f,inj}\delta_{i,inj} + F_{r,i+1} + F_{p,i-1}, \quad (1)$$

where $\delta_{i,inj} = 1$ for $i = inj$ and $\delta_{i,inj} = 0$ otherwise. f, p and r stand for feed, permeate and retentate, respectively.

The stages placed after the injection stage belong to the enriching section (N stages), whereas the stages placed before form the stripping section (M stages). The enriching section is where the enrichment of the species exhibiting the highest permeance takes place, and the stripping section is where it is depleted. Thus, the concentrations of the gas species in the flows $F_{r,i+1}$ and $F_{p,i-1}$ (which both feed stage i) will be necessarily different, and thus dilution of previously concentrated streams will occur. As a consequence, energy spent in separating the

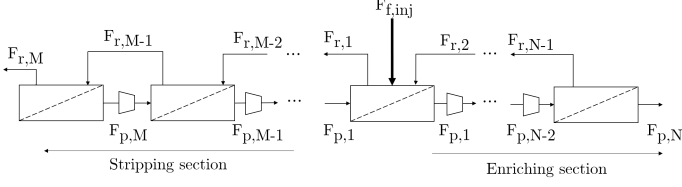


Figure 1: Schematic diagram of a membrane cascade in series arrangement. The enriching section is where the species with the highest permeance is enriched, and the stripping section is where this species is depleted.

$$S_{\text{stg}} = \frac{y_p}{\frac{x_r}{1-x_r}} \equiv \frac{Y_p}{X_r} \quad (3)$$

$$h = \frac{y_p}{\frac{x_f}{1-x_f}} \equiv \frac{Y_p}{X_f} \quad (4)$$

$$t = \frac{x_f}{\frac{x_r}{1-x_r}} \equiv \frac{X_f}{X_r} \quad (5)$$

$$S_{\text{stg}} = h \times t = h^2 = t^2 \quad (6)$$

Using (6) and the definitions of h and t , two relations, given by equations (7) and (8), can be derived for y_p and x_r depending solely on x_f and S_{stg} .

$$y_p = \frac{\sqrt{S_{\text{stg}}} X_f}{1 + \sqrt{S_{\text{stg}}} X_f} \quad (7)$$

$$x_r = \frac{\frac{x_f}{\sqrt{S_{\text{stg}}}}}{1 + \frac{x_f}{\sqrt{S_{\text{stg}}}}} \quad (8)$$

2.3. Ideal selectivity and pressure ratio

The separation factor, introduced in the previous section, quantifies the separation efficiency of one gas with respect to the other. Furthermore, there is another quantity which gives a first estimation for the separation performance of a permeable membrane, and it is defined by the ratio of the permeances Π_i of two gaseous species a and b (equation (9)). This quantity is defined such that it is larger than 1. Thus, according to (9), species a is enriched in the enriching section of the cascade, and depleted in the stripping section.

$$\alpha \equiv \frac{\Pi_a}{\Pi_b} \quad (9)$$

The permeation of gas species across a membrane is driven by the partial pressure difference between the feed and permeate sides of the membrane. Therefore, the enrichment attainable in a single separation unit is not only limited by the permselectivity α but also limited by the feed-to-permeate pressure-ratio γ defined in equation (10). It can be shown that the permeate concentration y_p obtained by feeding a membrane with a feed

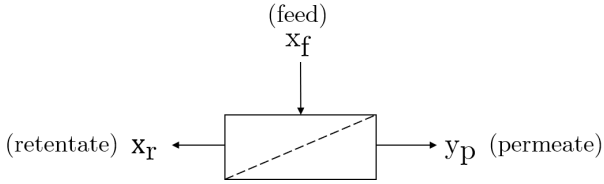


Figure 2: Single stage diagram, specifying the feed, permeate and retentate sides of the membrane with the corresponding concentrations: x_f , y_p , x_r .

gaseous species is wasted due to this dilution. Therefore, to ensure the minimum separation energy, the concentrations in the streams coming from $i + 1$ (i.e., $x_{r,i+1}$) and $i - 1$ (i.e., $y_{p,i-1}$) must be equal to the feed concentration $x_{f,i}$, as expressed by equation (2). This equation expresses the condition for an ideal cascade [13].

$$x_{f,i} = x_{r,i+1} = y_{p,i-1}, \quad (2)$$

2.2. Stage separation factor

The separation factor is a dimensionless parameter which quantifies the separation efficiency of a membrane for two different gas species. Three different separation factors, presented in equations (3) – (5), are commonly used in the literature. S_{stg} , which relates the permeate (y_p) and retentate (x_r) concentrations, is the stage separation factor; h , relating the permeate and feed (x_f) concentrations, is the head separation factor; t , defined by the feed and retentate concentrations, is the tail separation factor (refer to figure 2). For a separator with one inlet, it can be shown that $h = t$ leading to the relation presented in equation (6) holds [13].

131 concentration x_f depends on both α and γ according to equation
 132 (11) [14]. This relation is actually obtained for an idealized case
 133 of well-mixed mixtures, where the concentrations of the species
 134 are constant along the membrane module. In other words, the
 135 mixing rate of the species is considered to be much higher than
 136 their diffusion rates [15].

$$\gamma \equiv \frac{P_f}{P_p} \quad (10)$$

$$y_p = \frac{\gamma}{2} \left[x_f + \frac{1}{\gamma} + \frac{1}{\alpha - 1} - \sqrt{\left(x_f + \frac{1}{\gamma} + \frac{1}{\alpha - 1} \right)^2 - 4 \frac{\alpha x_f}{(\alpha - 1)\gamma}} \right], \quad (11)$$

137 In figure 3 the permeate concentration, given by equation
 138 (11), is plotted against the pressure-ratio γ for different selec-
 139 tivities α . Two regimes can be identified: (i) selectivity-limited
 140 when $\alpha \ll \gamma$ and (ii) pressure-limited when $\gamma \ll \alpha$. For low
 141 selectivities (e.g., $\alpha \leq 3$), the permeate concentration increases
 142 for rather small values of γ and eventually reaches a plateau
 143 where a further increase of γ (i.e., increase of the driving force)
 144 does not lead to an increase of the enrichment. At these condi-
 145 tions, the increase in the permeate concentration is selectivity-
 146 limited. When the selectivity α is increased, the range where
 147 y_p is dependent on γ is wider. At these conditions, the concen-
 148 tration in the permeate side is limited by the pressures across
 149 the stage. A yellow region is displayed between $\gamma = 2$ and
 150 $\gamma = 20$ in the same figure to indicate the practical range of
 151 values which can be applied, considering the pumping systems
 152 availability and economical viability [14, 16]. In this region,
 153 both situations (i.e., pressure limited and selectivity limited) oc-
 154 cur, showing that both the impact of γ and α on the membrane
 155 performances have to be considered.

156 Figure 4 depicts the plot of y_p as a function of α for $\gamma =$
 157 20, considered as the maximum available of pressure-ratio cur-
 158 rently achievable. Despite the high selectivities (i.e., $\alpha > 100$),
 159 the maximum achievable permeate concentration, for an initial
 160 concentration of 2 mol%, is limited to 4 mol%. In this case,
 161 the use of membranes with selectivities above 100 would be

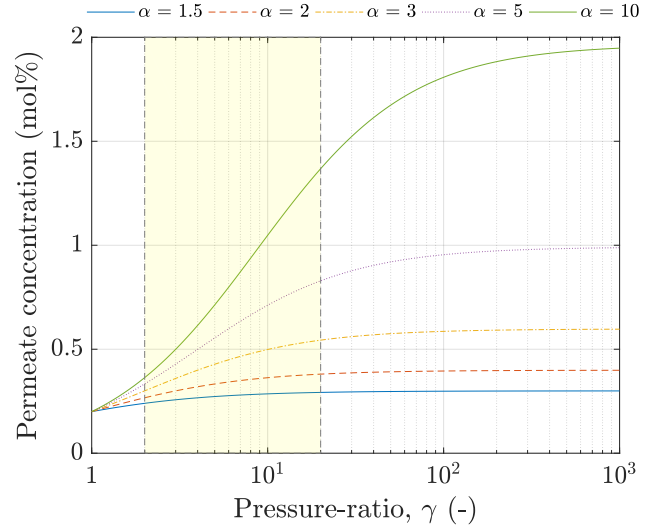


Figure 3: Permeate concentration as a function of the pressure-ratio for different selectivity values, obtained for $x_f = 0.2$ mol%. In yellow, the region for the practical pressure-ratio values is presented. $\alpha = 1.5$: solid blue line; $\alpha = 2$: dashed orange line; $\alpha = 3$: dot-dashed dark yellow line; $\alpha = 5$: dotted purple line; $\alpha = 10$: solid green line.

of marginal advantage, since its performance would be significantly limited by γ .

In addition, it can be shown (refer to Appendix A for derivation) that the separation factor S_{stg} (equation 6) depends on both α and γ via equation (12) [17]. From this equation it can be easily seen that when $\gamma \rightarrow \infty$, then $S_{\text{stg}} \rightarrow \alpha$, and the highest performance is attained. On the contrary, when $\gamma \rightarrow 1$, $S_{\text{stg}} \rightarrow 1$, and thus no separation takes place. Therefore, in practice, the range of the stage separation factor is between 1 and α .

$$S_{\text{stg}} = \frac{\alpha - y_p(\alpha - 1)\gamma^{-1}}{1 + (1 - y_p)(\alpha - 1)\gamma^{-1}} \quad (12)$$

2.4. Enrichment factor and recovery fraction

The selectivity is a well-established parameter which is used to compare and discuss separation performance of membranes. Nevertheless, for industrial applications, especially those relying on membrane cascades, two additional parameters are used to express their performance: the enrichment factor (EF) and the recovery fraction (RF). The enrichment factor, given by equation (13), quantifies the increase in concentration of the desired product at the last stage of the cascade (N in figure 1) in

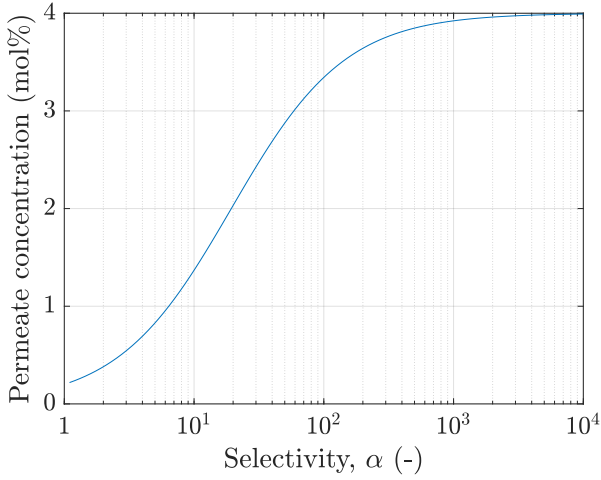


Figure 4: Permeate concentration as a function of the selectivity for $\gamma = 20$ and $x_f = 0.2$ mol%.

respect to the concentration of the initial feeding flow at the injection stage. The recovery fraction RF , given by equation (14), is the relative amount of product extracted at the last stage N in respect to the injection stage. These two quantities are used as requirements for the design and dimensioning of a membrane cascade (e.g., to determine the required number of stages), as discussed below.

$$EF = \frac{y_{p,N}}{x_{f,inj}} \quad (13)$$

$$RF(\%) = 100 \times \frac{y_{p,N} F_{p,N}}{x_{f,inj} F_{f,inj}} \quad (14)$$

2.5. Determination of the number of stages, concentrations and flows

The number of stages of a membrane cascade can be determined from the performance requirements EF and RF , the initial feed concentrations $x_{f,inj}$ and feed flow $F_{f,inj}$. The required flows and concentrations at the permeate side of stage N (i.e., the last of the enriching section), $F_{p,N}$ and $y_{p,N}$, and at the retentate side of M (i.e., the last stage of stripping section), $F_{r,M}$ and $x_{r,M}$, are determined by equations (15)-(18). Then, the number of stages is found by iterating y_p and x_r concentrations along the cascade using equations (7) and (8) until $y_p \geq y_{p,N}$ and $x_r \leq x_{r,M}$. In these equations, the X_f term is determined

by the feeding concentration (equation (2)). The numerical implementation of these equations is presented and discussed in section 3.

$$F_{p,N} = \frac{1}{EF} \frac{RF}{100} F_{f,inj} \quad (15)$$

$$F_{r,M} = F_{f,inj} - F_{p,N} \quad (16)$$

$$y_{p,N} = x_{f,inj} EF \quad (17)$$

$$x_{r,M} = \frac{F_{f,inj} x_{f,inj} - F_{p,N} y_{p,N}}{F_{r,M}} \quad (18)$$

The feed flows at the inlet of each stage along the cascade are determined from the stage-cut values v_i . The stage-cut value is defined by the permeate-to-feed flows ratio, and it can be calculated using the feed, permeate and retentate concentrations at stage i according to equation (19) (refer to Appendix B for derivation) [17]. Since $F_{p,i-1} = v_{i-1} F_{f,i-1}$ and $F_{r,i+1} = (1 - v_{i+1}) F_{f,i+1}$, equation (1) can be re-written as equation (20). The application of equation (20) to each stage results in a matrix, given by equation (21), that enables the calculation of the feed flows at the inlet of each cascade stage. The corresponding permeate and retentate flows can be calculated using, respectively, equations (22) and (23).

$$v_i = \frac{F_{p,i}}{F_{f,i}} = \frac{x_{f,i} - x_{r,i}}{y_{p,i} - x_{r,i}} \quad (19)$$

$$F_{f,i} = F_{f,inj} \delta_{i,inj} + (1 - v_{i+1}) F_{f,i+1} + v_{i-1} F_{f,i-1} \quad (20)$$

$$\begin{bmatrix} \vdots \\ F_{f,i-1} \\ F_{f,i} \\ F_{f,i+1} \\ \vdots \end{bmatrix} = \begin{bmatrix} \vdots & \vdots & 0 & 0 & 0 \\ -v_{i-2} & 1 & -(1 - v_i) & 0 & 0 \\ 0 & -v_{i-1} & 1 & -(1 - v_{i+1}) & 0 \\ 0 & 0 & -v_i & 1 & -(1 - v_{i+2}) \\ 0 & 0 & 0 & \vdots & \vdots \end{bmatrix}^{-1} \begin{bmatrix} \vdots \\ \vdots \\ F_{f,inj} \\ \vdots \\ \vdots \end{bmatrix} \quad (21)$$

$$\mathbf{F}_p = v \mathbf{F}_f \quad (22)$$

$$\mathbf{F}_r = \mathbf{F}_f - \mathbf{F}_p \quad (23)$$

$$P_{\text{tot}} = \frac{W_{\text{min}}}{RT\eta} \left(\sum_{i=1}^M F_{p,i} + \sum_{j=1}^{N-1} F_{p,j} \right) \quad (27)$$

2.6. Stages surface area

The required surface area A_i (m²) for each stage is determined according to equation (24). This quantity depends on the total permeating flow $F_{p,i}$, feed and permeate pressures (i.e., γ and p_p), and gas permeance Π (mol m⁻² s⁻¹ Pa⁻¹) through the membrane. The permeance is dependent on the gas type, membrane's material and temperature. The value of the permeance used to calculate the surface area is determined using the linear combination of the permeances for the two species a and b present in the stream, according to equation (25) [18].

$$A_i = \frac{F_{p,i}}{\Pi p_p (\gamma - 1)} \quad (24)$$

$$\Pi = \Pi_a x_{f,a} + \Pi_b x_{f,b} \quad (25)$$

2.7. Evaluation of the compression power

As presented in figure 1, compressors are employed between stages to increase the pressure of the flow routed from the permeate side of stage i to the feed side of stage $i + 1$, and maintain a sufficient pressure difference across the membranes of the different stages. The compression power required is dependent on the minimum molar energy W_{min} (J mol⁻¹) required to isentropically compress a gas from p_p to p_f [19]. W_{min} is determined by equation (26), where c_p and c_v are the gas specific heat constants (J K⁻¹ kg⁻¹), T is its temperature (K) and R is the constant for ideal gases (J K⁻¹ mol⁻¹).

$$W_{\text{min}} = RT \frac{c_p}{c_p - c_v} \left[\left(\frac{p_f}{p_p} \right)^{\frac{c_p}{c_p - c_v}} - 1 \right] \quad (26)$$

The total compressing power P_{tot} (W) required to run the membrane cascade can be calculated by summing all the contributions for the $N + M - 1$ stages (the permeate flow of the N -th stage is not compressed), as expressed by equation (27). The electrical-to-mechanical conversion efficiency η is also considered, which is typically around 70% [20].

3. Numerical implementation

The objective of this work is to determine numerically the number of stages required to achieve a defined set of separation performance, given by the enrichment factor and recovery fraction defined in equations (13) and (14). In addition, the gas concentrations and flows along the cascade, the stages' surface area and power consumption are also desired outputs of the numerical calculations (since they have a direct impact on the physical dimensioning and operating costs). These parameters are calculated relying on a simple algorithm. The required input parameters for these calculations are: enrichment factor (EF) and recovery fraction (RF), feed flow at the injection stage ($F_{f,\text{inj}}$) and concentrations ($x_{f,\text{inj}}$) of the binary mixture feeding flow, feed-to-permeate pressures (γ) and permeances ratio given by the selectivity α . The numerical implementation follows the steps below:

- with EF , RF , $F_{f,\text{inj}}$ and $x_{f,\text{inj}}$, determine the permeate and retentate concentrations of the most permeating species: $y_{p,N}$ (equation (17)) and $x_{r,M}$ (equation (18));
- using α , γ and $y_{p,N}$, estimate the stage separation factor S_{stg} , given by equation (12);
- with S_{stg} and $x_{f,\text{inj}}$, determine the permeate ($y_{p,\text{stg}}$) and retentate ($x_{r,\text{stg}}$) concentrations of the injection stage, with equations (7) and (8);
- using the feed, permeate and retentate concentrations for the injection stage, calculate its cut-value v_{stg} , using equation (19);
- using $y_{p,\text{stg}}$ as feed concentration for stage 1 of the enriching section, use $X_f = \frac{y_{p,\text{stg}}}{1 - y_{p,\text{stg}}}$ to determine $y_{p,1}$ and $x_{r,1}$ (as in step c)). Repeat these calculations until the permeate concentration has an equal or larger value than $y_{p,N}$. The number of iterations required is equal to the number of stages N in the enriching section. For each stage determine the cut-values as described step d);

- 275 f) using $x_{r,1}$ as feed concentration for stage 1 of the stripping
 276 section, use $X_f = \frac{x_{r,1}}{1-x_{r,1}}$ to determine $y_{p,1}$ and $x_{r,1}$ (as in step
 277 c)). Repeat these calculations until the retentate concentra-
 278 tion has an equal or lower value than $x_{r,M}$. The number of
 279 iterations required is equal to the number of stages M in the
 280 stripping section. For each stage determine the cut-values as
 281 described in step d);
- 282 g) the feed flows along the cascade are determined using the
 283 cut-values v_i in the matrix equation (21). The permeate and
 284 retentate flows are then calculated using, respectively, (22)
 285 and (23);
- 286 h) using the permeate flows determined in g), the gamma-value,
 287 the permeate pressure, and the permeances of the permeat-
 288 ing gases, the surface area of each stage can be determined
 289 with equation (24). The total compressing power is also
 290 calculated using the permeate flows, according to equation
 291 (27).

292 4. Sensitivity analysis for membrane cascade dimensioning

293 4.1. Input and output parameters

294 In the sensitivity study presented below, we are interested in
 295 determining the impact of the required performance (EF , RF),
 296 operating conditions (γ) and membrane properties (α) on the:

- 297 • number of stages N and M ;
- 298 • stages flows $F_{f,i}$, $F_{p,i}$ and $F_{r,i}$;
- 299 • stages surface areas A_i ;
- 300 • total power consumption P_{tot} .

301 These four characteristics of the membrane cascade were
 302 selected as they provide key information regarding its design,
 303 dimensions and cost-effectiveness, required to evaluate the fea-
 304 sibility of membrane technology for a given application.

305 For the sensitivity analysis a reference case is considered,
 306 whose scenario is presented in table 1. The values $EF = 20$
 307 and $RF = 90\%$ were proposed in an earlier publication for the

Symbol	Value	Unit
EF / RF	20 / 90%	–
H_2 / He	0.2 / 99.8	mol%
Π_{H_2}	0.72	$\mu\text{mol m}^{-2}\text{s}^{-1}\text{Pa}^{-1}$
Π_{He}	0.34	$\mu\text{mol m}^{-2}\text{s}^{-1}\text{Pa}^{-1}$
α	2	–
$F_{f,inj}$	10^4	$\text{m}^3 \text{h}^{-1}$ at STP

Table 1: Reference case parameters used for the sensitivity analysis based on the requirements for the TER system of the EU-DEMO [21, 22]. The permeances for H_2 and He were obtained experimentally with zeolite membranes [23]. STP: standard conditions for temperature and pressure.

308 performance of the membrane cascade in DEMO-relevant con-
 309 ditions for the HCPB TER system [21]. The composition of
 310 the purge gas downstream of the breeding blanket is expected
 311 to consist mainly of H_2 and He , with 0.1 wt.% H_2/He (which
 312 translates into 0.2 mol% H_2/He) [22]. In addition, $\alpha = 2$ is rep-
 313 resentative of the selectivity value obtained by calculating the
 314 ratio of the experimental permeances of H_2 and He obtained
 315 for a MFI zeolite-type membrane, also given in table 1 [23].
 316 From this reference case, EF , RF , α and γ are varied in ranges
 317 of interest and their impact on the dimensioning of the cascade
 318 is discussed. For the sensitivity analysis, the stages are num-
 319 bered $1, 2, \dots, N_{stg}$, where N_{stg} corresponds to the total number
 320 of stages (i.e., $N_{stg} = N + M + 1$).

321 4.2. Influence of selectivity and pressures-ratio

322 4.2.1. Number of stages

323 In figure 5, the number of stages as a function of α , obtained
 324 for $EF = 20$ and $RF = 90\%$ is presented. This plot was ob-
 325 tained for $\gamma = 20$ (i.e., upper limit of the realistic pressure-ratio
 326 values discussed in section 2.3). The total number of stages de-
 327 creases from 27 for $\alpha = 1.5$ down to 6 for $\alpha = 10$. It should
 328 be noticed that the number of stages sharply decreases from 27
 329 down to 9 when the selectivity increases from $\alpha = 1.5$ to $\alpha = 4$.
 330 When the selectivity is increased further to 10, only three stages
 331 are spared.

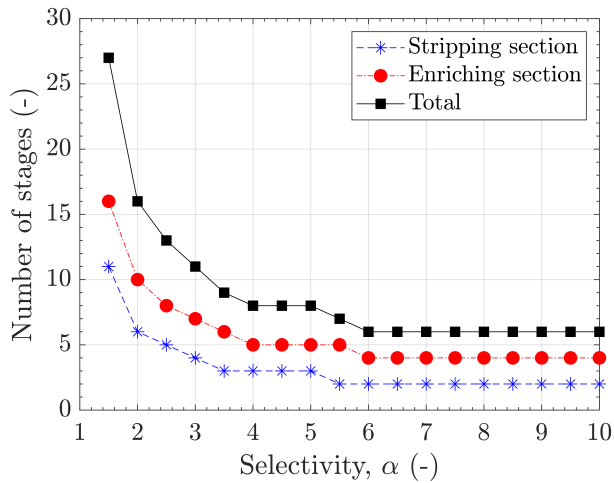


Figure 5: Number of stages as a function of the selectivity. Obtained for $\gamma = 20$, $EF = 20$, $RF = 90\%$. Stages in stripping section: dashed blue line with stars; stages for enriching section: dot-dashed red line with circles; total number of stages (i.e., enriching plus stripping): solid black line with squares.

332 The number of stages obtained with $\gamma = 20$ is the smallest
 333 that would be required for practical applications. However, this
 334 value corresponds to the largest power consumption required
 335 which directly impacts the feasibility of the system. There-
 336 fore, an investigation was done to determine, for each selec-
 337 tivity α , what is the minimum value γ_{\min} which keeps the num-
 338 ber of stages at a minimum. The results are compiled in table
 339 2. For each selectivity, a range $[\gamma_{\min}, \gamma_{\max}]$ was determined,
 340 corresponding to the pressure ratios that can be applied with-
 341 out changing the number of stages. The resulting range for the
 342 stage separation factors $[S_{\text{stg,min}}, S_{\text{stg,max}}]$, calculated with (3),
 343 are also presented.

344 For selectivities between 6 and 10, the minimum number of
 345 stages required to achieve $EF = 20$ and $RF = 90\%$ is the same
 346 and equal to 6. Nevertheless, γ_{\min} decreases when the selectiv-
 347 ity increases. For instance, $\gamma_{\min} = 7.5$ for $\alpha = 10$. Therefore,
 348 although there is no reduction of the total number of stages, the
 349 increasing of selectivity relaxes the required pressure-ratio. It is
 350 also interesting to appreciate that for low selectivities, S_{stg} and
 351 α are very similar, whereas, for instance, for $\alpha = 10$ S_{stg} is not
 352 larger than 6.9. This illustrates the two regimes introduced in
 353 section 2.3 (pressure and selectivity limited).

α	N_{stg}	γ_{\min}	γ_{\max}	$S_{\text{stg,min}}$	$S_{\text{stg,max}}$
(-)	(-)	(-)	(-)	(-)	(-)
1.5	27	19.0	20.0	1.462	1.464
2	16	11.5	20.0	1.843	1.906
3	11	9.5	20.0	2.489	2.734
4	8	16.0	20.0	3.383	3.491
5	8	8.5	20.0	3.431	4.188
6	6	16.0	20.0	4.606	4.831
7	6	11.5	20.0	4.650	5.425
8	6	9.5	20.0	4.668	5.977
9	6	8.5	20.0	4.708	6.491
10	6	7.5	20.0	4.624	6.971

Table 2: Range of γ , and corresponding range of S_{stg} , for which the lowest practical number of stages is ensured for $EF = 20$, $RF = 90\%$ and $x_{f,i} = 0.2$ mol%.

4.2.2. Flows and concentrations

As discussed in section 2.1, the classical membrane cascade is operated under flows circulating between stages. As a consequence, there is a build-up of flows along the cascade, whose steady-state profile can be determined from the equations presented before. The profile for the feed flows is presented in figure 6. This plot was obtained for the conditions presented in table 1, which provides a 16-stages cascade, and the feeding stage is the number 7. It should be noticed that, as a result of the build-up of flows, the flow at the injection stage is around 5.4×10^4 m³/h, which is roughly five times higher than the initial feeding flow. However, the retentate and permeate flows $F_{r,1}$ and $F_{p,16}$ are, respectively, 9626 m³/h and 374 m³/h, and thus the mass-balance is respected. This asymmetric profile of the feed flows is explained by the fact that a small fraction of the feeding flow permeates through the membrane stages, where a large fraction is recycled back in the retentate sides of the stages (i.e., $v_i < 0.5$). As a result, larger flows exist at the stripping section in comparison to the enriching section.

The plot of figure 7 shows the total feed flow at the injection stage as a result of the recycling of flows between stages,

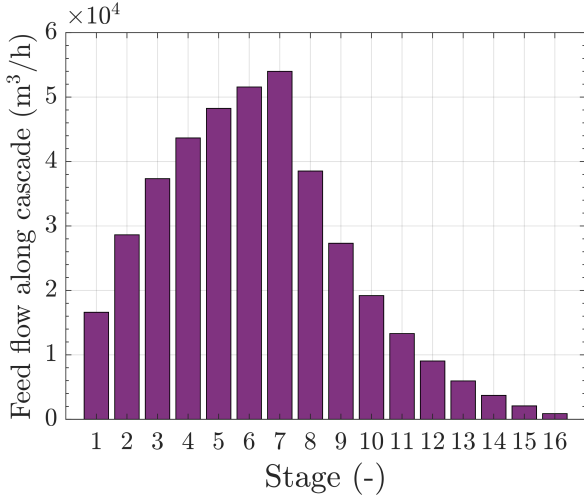


Figure 6: Feed flow along the cascade stages. Obtained for $EF = 20$, $RF = 90\%$, $F_{f,inj} = 10^4 \text{ m}^3/\text{h}$, $\gamma = 20$ and $\alpha = 2$. The 7-th stage is the injection stage.

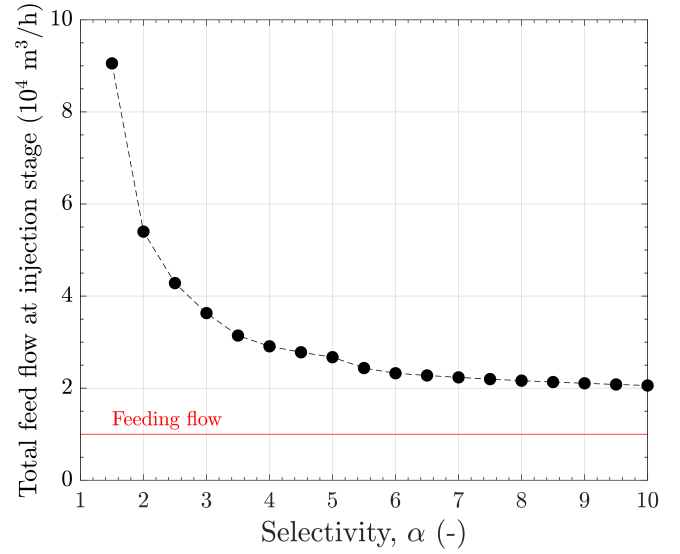


Figure 7: Feed flow at the injection stage as a function of the selectivity. The red solid line indicates the original feed flow: $F_{f,inj} = 10^4 \text{ m}^3/\text{h}$. Obtained for $EF = 20$, $RF = 90\%$ and $\gamma = 20$.

375 obtained for $F_{f,inj} = 10^4 \text{ m}^3/\text{h}$. Since the number of stages de-
 376 creases with the selectivity (figure 6), the build-up of flows also
 377 decreases. While for $\alpha = 1.5$ the feed flow amounts to roughly
 378 9 times the initial feeding flow, for $\alpha = 10$ the total feed flow is
 379 only 2 times higher than $F_{f,inj}$. Thus, considering that the initial
 380 feeding flow foreseen to purge the breeding blanket is already
 381 large, the use of a membrane cascade would increase this flow
 382 at least two times ($2 \times 10^4 \text{ m}^3/\text{h}$ in this case). This fact raises
 383 questions regarding to the dimensions and footprint of the sys-
 384 tem (i.e., components, pipework) but also to tritium inventory
 385 (even though membrane cascades can be operated continuously
 386 and do not require a redundant system). These aspects are dis-
 387 cussed in section 5.

388 The plot of figure 8 shows the profiles of the feed, permeate
 389 and retentate concentrations along the cascade for the species
 390 with the highest permeance. These results were obtained for
 391 the reference case. As expected, the concentrations increase
 392 along the cascade, towards the last stage of the enriching sec-
 393 tion, where the highest concentrations are obtained. The reten-
 394 tate concentration at stage 1 and the permeate concentration at
 395 stage 16 meet the input requirements presented in section 2.5:
 396 the permeate concentration at stage 16 is 4.79 mol%, which is
 397 higher than $y_{p,N} = 4 \text{ mol}\%$ (using equation (13)); the reten-

tate concentration at stage 1 is 0.015 mol%, which is lower
 than $x_{r,M} = 0.021 \text{ mol}\%$ (using equation (18)). Furthermore,
 it should be noticed that the permeate and retentate concentra-
 tions of stages $i - 1$ and $i + 1$, respectively, are equal to the feed
 concentration at stage i (as expected from the definition of ideal
 cascade, given by equation (2)).

4.2.3. Permeate flow at the last stage

In view of the applicability of a membrane cascade in a
 complex system such as the fuel cycle of a fusion reactor, with
 plenty of interfaces, the permeation flow $F_{p,N}$ at the last stage
 (and/or the retentate flow $F_{r,M}$ at the first stage) is a very im-
 portant parameter to take into account. This flow will have
 a direct impact on the dimension of the next system, or, con-
 versely, the existence of an interface may impose important
 constraints on the flows and concentrations which have to be
 reached downstream of the cascade. In the plot of figure 9,
 the ratio $F_{p,N}/F_{f,inj}$ is plotted as a function of the selectivity for
 $EF = 20$, $RF = 90\%$ and $\gamma = 20$. The dependency of the
 relative permeate flow with the selectivity is not monotonic be-
 cause the number of stages required to fulfill EF and RF also
 changes with the selectivity (refer to section 4.2.1 and figure 5).

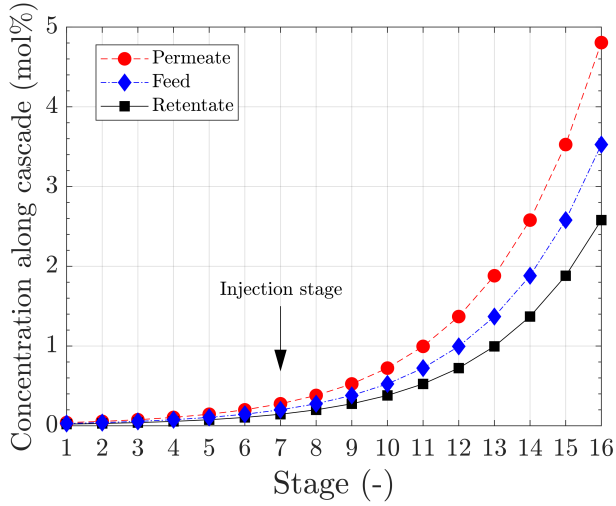


Figure 8: Absolute concentrations of the most permeating species along the cascade stages, obtained for $EF = 20$, $RF = 90\%$, $\gamma = 20$, $\alpha = 2$ and $x_{r,i} = 0.2 \text{ mol\%}$. Feed: blue dot-dashed line with diamonds; Permeate: red dashed line with circles; Retentate: black solid line with squares.

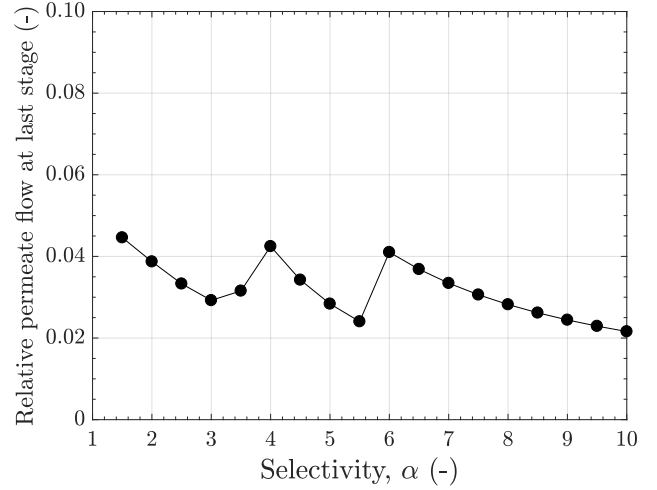


Figure 9: Relative permeate flow at last stage, in respect to the feed flow F_r , as a function of the selectivity α . Obtained for $\gamma = 20$, $EF = 20$, $RF = 90\%$, $F_{f,inj} = 10^4 \text{ m}^3/\text{h}$.

419 For selectivity ranges associated to a constant number of stages, 442
 420 ($\alpha = 6 - 10$, with 6 stages), the relative permeate flow at the 443
 421 last stage decreases with the selectivity. As a matter of fact, a 444
 422 higher selectivity induces a decrease of the permeate flow (and
 423 an increase of the retentate flow). 445

424 For α values between 6 and 10 the total number of stages is 446
 425 constant, and equal to 6 (refer to figure 5). Therefore, when α 447
 426 increases, the concentration of the most permeating species in 448
 427 the permeate flow increases, leading to a decrease of the total 449
 428 permeate flow: from roughly 4% down to 2% of the injection 450
 429 feed flow. Likewise, for $\alpha = 4 - 5$, the number of stages is 451
 430 also constant (equal to 8) resulting in a decrease of $F_{p,N}$ with α . 452
 431 For selectivities between 1.5 and 3.5, the relative permeate flow 453
 432 at last stage globally decreases but is impacted by additional 454
 433 effects, as the number of stages is reduced from 27 to 9. 455

434 Regardless of the selectivity, the permeate flow at the last 456
 435 stage of the enriching section represents only a small percent- 457
 436 age of the initial feeding flow (in the range 2% - 5%), whereas 458
 437 the retentate flow of the first stage contribution is above 90%. 459
 438 Nevertheless, these values change considerably with EF , as it is 460
 439 shown in section 4.3.2. In view of the application of the mem- 461

brane cascade to the TER system of the HCPB, the depleted, 440
 441 large flow at the retentate side of the stripping section is re-used
 for further purging of the ceramic beds. The enriched, small
 442 flow of the enriching section is used as input for the membrane
 reactor, which must be kept at reasonable size and costs.

4.2.4. Surface area

The total surface area of each membrane cascade system 446
 as a function of the selectivity is presented in the figure 10
 447 for $\gamma = 20$. A monotonic decrease of the area exists from
 448 $\sim 1 \times 10^4 \text{ m}^2$ for $\alpha = 1.5$ down to $\sim 3 \times 10^2 \text{ m}^2$ for $\alpha = 10$,
 449 mainly due to the decreasing number of stages. If the mini-
 450 mized pressure-ratios (γ_{\min} in table 2) would have been used in-
 451 stead, the surface area would be greater (up to five times). This
 452 fact demonstrates the trade-off existing between the pressure-
 453 ratio (i.e., compression power) and the surface area. Moreover,
 454 calculations with different injection flows highlight its strong
 455 impact on the surface area: a flow decrease by a factor of 10,
 would lead to 10 times less area.

4.2.5. Power consumption

The total power consumption of each membrane cascade 460
 system as a function of the selectivity was calculated using
 equation (27) and is presented in figure 11 for $\gamma = 20$. A 461

sharp decrease exists from ~ 120 MW for $\alpha = 1.5$ down to ~ 4 MW for $\alpha = 10$, which corresponds to a power reduction by a factor of 30. The order of magnitude for the power consumption is essentially determined by the flows. Therefore, the flow-independent power consumption reduction relative to $\alpha = 1.5$ is also presented in figure 11. From $\alpha = 1.5$ to $\alpha = 3.5$, the power consumption is reduced by one order of magnitude. Further increasing of α to 10, the power required is only reduced by a factor 3. If the γ_{\min} values (table 2) would have been used as input, the power consumption could be reduce by, at best, a factor of 2. As for the surface-area, if the injection flow is decreased by a factor 10 the power also reduces by the same factor (equation 27).

4.3. Influence of EF and RF

4.3.1. Number of stages

The number of stages as a function of both EF and for different values of RF , both used as performance requirements defined in equations (13) and (14), is presented in the plot of figure 12 for $\alpha = 2$ and $\gamma = 20$. The total number of stages (top plot) varies considerably with both EF and RF . On the one side, for higher values of EF a higher purification is required, and thus more stages are required in the enriching section (middle plot in the same figure). For instance, for $RF = 90\%$, the number of stages increases from 14, for $EF = 10$, to 22, for

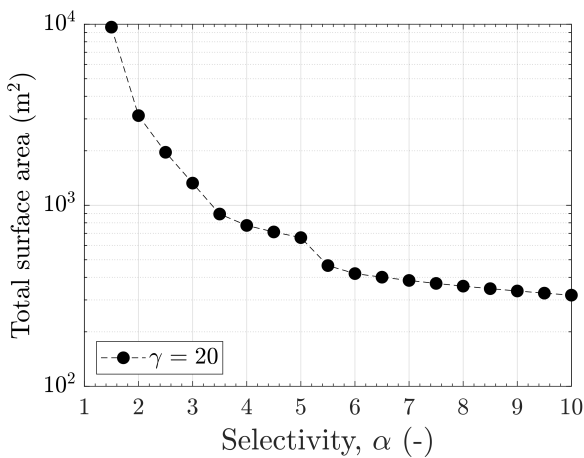


Figure 10: Total surface area as a function of the selectivity for $\gamma = 20$. Obtained for $EF = 20$, $RF = 90\%$, $\alpha = 2$ and $F_{f,inj} = 10^4$ m³/h.

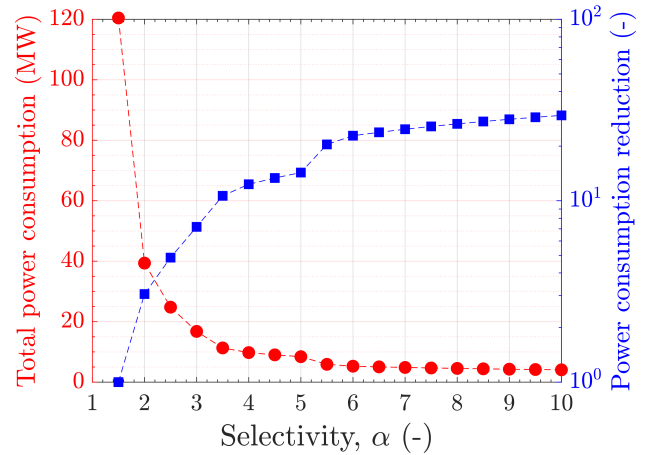


Figure 11: Total power consumption (red dashed-line with circles) and power reduction (in respect to the value for $\alpha = 1.5$, blue dashed-line with squares) as a function of the selectivity α for $\gamma = 20$. Obtained for $EF = 20$, $RF = 90\%$, $F_{f,inj} = 10^4$ m³/h.

$EF = 90$. On the other side, for a constant value of EF and a given injection flow, a higher RF can only be reached by increasing the permeate flow at the output of the cascade, which is achieved by increasing the number of stages to promote an increase of the build-up flows (equation 14). As EF is constant, the number of stages in the enriching section is kept constant, and hence the number of stages in the stripping section increases. This behavior can be appreciated in the middle and bottom plots of figure 12. The number of stages in the stripping section is mainly driven by RF , whereas the stages in the enriching section is independent of RF . Moreover, it should be noted that the impact of RF on the total number of stages is greater when $RF \rightarrow 100\%$. In fact, by increasing RF from 80% to 90%, the total number of stages for $EF = 20$ increases by 3, while from 90% to 99% the increase in number of stages is 7. Increasing further RF the number of stages required increases dramatically. At the limit of 100% an infinite number of stages would be required. This is justified by the fact that the cascade consists of membranes permeable for the feeding species, and thus a complete recovery of the desired product is not possible.

In the plot of figure 13, the total number of stages for $\alpha = 10$ as a function of both EF for different RF values is presented. It can be observed that the impact of EF and RF on the number

509 of stages is significantly lower than it is for $\alpha = 2$, since the
 510 stages have a high selectivity: for $RF = 90\%$, the number of
 511 stages increases from 5 to 7 when EF increases from 10 to 90,
 512 respectively.

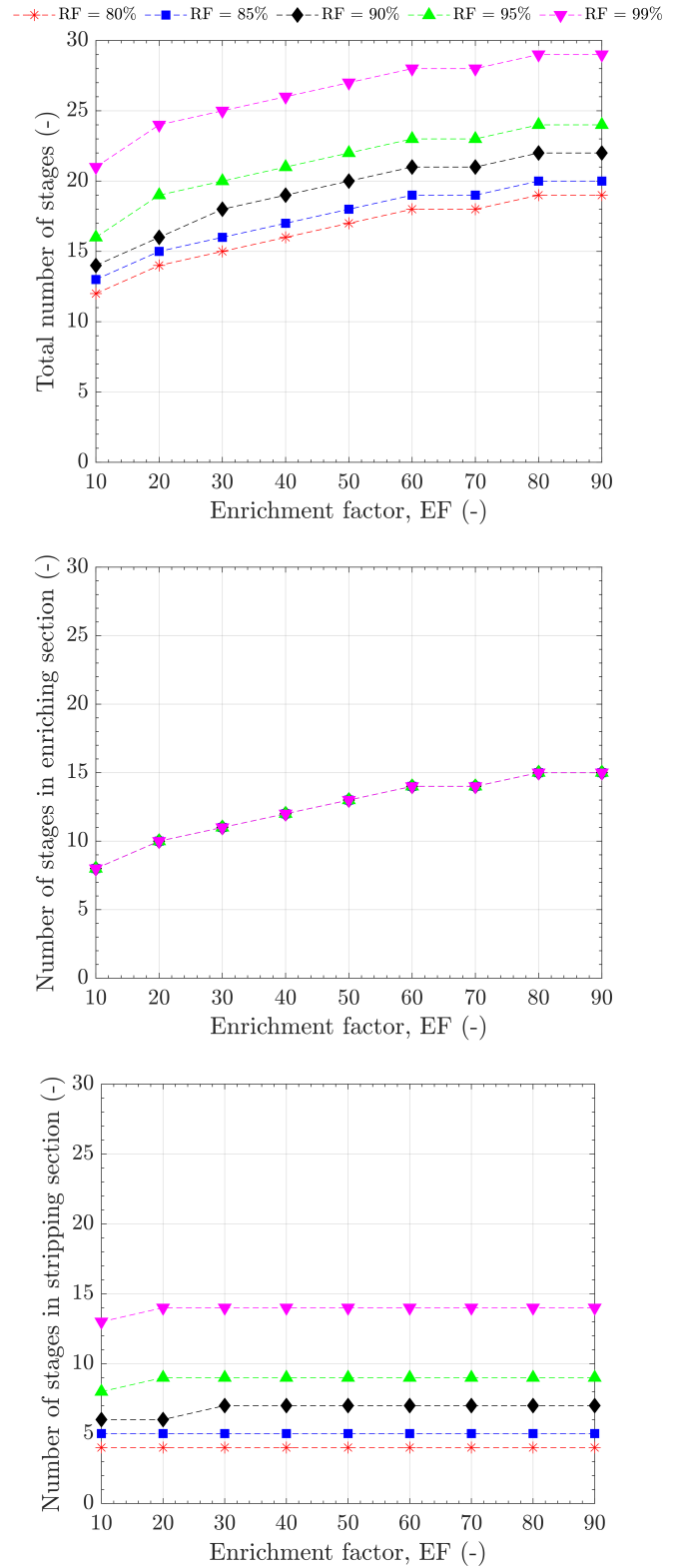


Figure 12: Total number of stages (top), and number of stages in the enriching (middle) and stripping (bottom) sections as a function of EF and RF , obtained for $\gamma = 20$, $\alpha = 2$. $RF = 80\%$: dashed red line with stars; $RF = 85\%$: dashed blue line with squares; $RF = 90\%$: dashed black line with diamonds; $RF = 95\%$: dashed green line with upward triangles; $RF = 99\%$: dashed pink line with downward triangles.

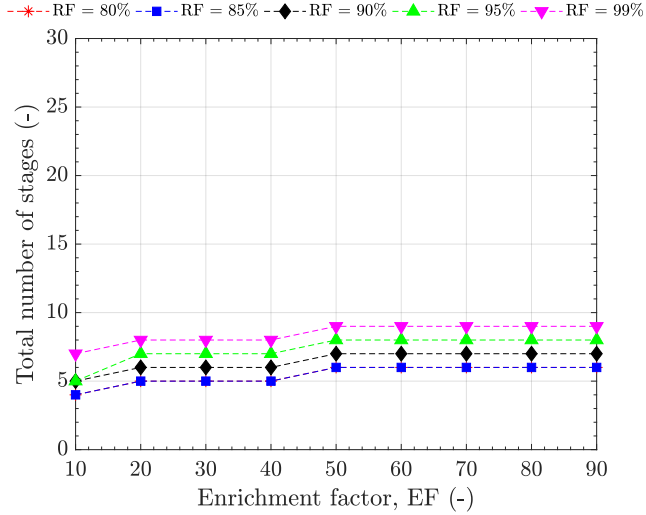


Figure 13: Total number of stages as a function of EF and RF , obtained for $\gamma = 20$, $\alpha = 10$. $RF = 80\%$: dashed red line with stars; $RF = 85\%$: dashed blue line with squares; $RF = 90\%$: dashed black line with diamonds; $RF = 95\%$: dashed green line with upward triangles; $RF = 99\%$: dashed pink line with downward triangles.

4.3.2. Permeate flow at the last stage

The permeate flow at the last stage of the cascade ($F_{p,N}$) is presented as a function of EF and RF for $\gamma = 20$, $\alpha = 2$ and

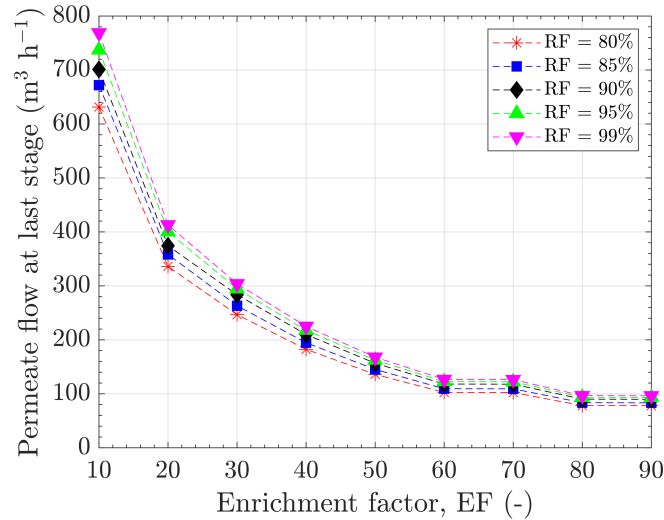


Figure 14: Permeate flow at last stage as a function of EF and RF , obtained for $\gamma = 20$, $\alpha = 2$, $F_{f,inj} = 10^4 \text{ m}^3/\text{h}$. $RF = 80\%$: dashed red line with stars; $RF = 85\%$: dashed blue line with squares; $RF = 90\%$: dashed black line with diamonds; $RF = 95\%$: dashed green line with upward triangles; $RF = 99\%$: dashed pink line with downward triangles.

$F_{f,inj} = 10^4 \text{ m}^3/\text{h}$ in figure 14. The dependency of $F_{p,N}$ with both EF and RF is consistent with the variation of the number of stages in the enriching section: EF significantly impacts $F_{p,N}$, whereas RF only has a limited influence. Increasing EF for a fixed RF leads to an increase of the number of stages in the enriching section, and thus the permeate flow of stage N decreases: for $RF = 90\%$, $F_{p,N}$ decreases from $700 \text{ m}^3 \text{ h}^{-1}$ to $100 \text{ m}^3 \text{ h}^{-1}$ for $EF = 10$ and $EF = 90$, respectively. These results are consistent with the number of stages presented in figure 12.

Increasing the EF requirement brings the benefit of decreasing the permeate flow at the last stage (i.e., interface with the next system). This may be desired as for instance in the case for a tritium extraction system based on a membrane cascade followed by a palladium-based membrane reactor. On the other side, the lower flow is obtained at the expense of a higher number of stages, which leads to other constraints, such as the space occupied by the system or the power required to run it.

4.3.3. Power consumption

In figure 15, the power consumption as a function of EF and for different RF values relative to the reference case ($EF = 20$, $RF = 90\%$) is presented. This plot was obtained for $\alpha = 2$ and $\gamma = 20$. Increasing EF or RF leads to an increase of the power consumption to operate the cascade. These results correlate well with the results presented on figure 12. The stronger dependency of the power consumption with RF is explained by the increasing of the number of stages which results in larger flows along the cascade. Moreover, it should be noticed that this plot gives the relative evolution of the power consumption with EF and RF that applies to any given injection flow.

For higher selectivities, the number of stages is less sensitive to EF and RF and therefore for a high-performing system the increase in power consumption is mitigated.

5. Discussion on the feasibility of a membrane cascade

The results presented in the previous section show the impact of the different design (i.e., EF , RF), operating (i.e., γ) and

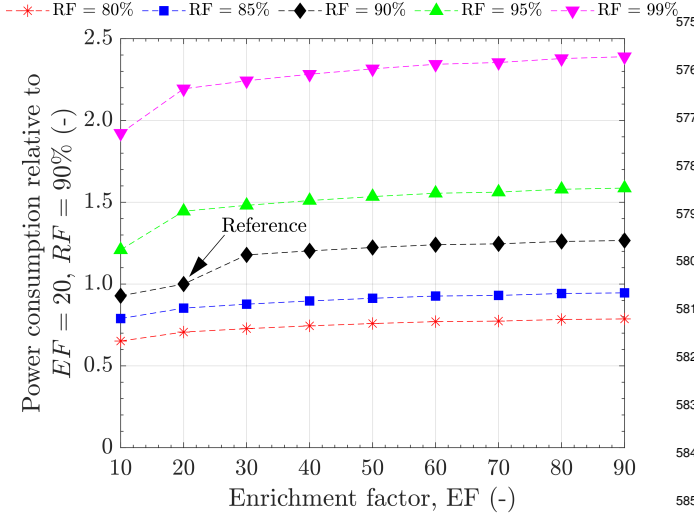


Figure 15: Power consumption relative to the reference case with $EF = 20$ and $RF = 90\%$ as a function of EF and RF . Obtained for $\alpha = 2$ and $\gamma = 20$.

separation (i.e., α) parameters on the number of stages, processing flows, surface area and power consumption of a membrane cascade. These are key information to discuss the feasibility and the interest related to membrane technologies, as fusion requires systems which are as simple as possible, compact, reliable and demonstrate a reasonable energy consumption.

As mentioned above, a multi-stage cascade is required when the separation of a single-membrane is not enough to meet the performance requirements. In the fuel cycle of a fusion reactor, the molecules required for separation (e.g., He, H₂, Ar, N₂, Xe) are rather small in contrast to the pore sizes of the porous (ceramic) membranes available. Therefore, the selectivity should not exceed the so-called Knudsen selectivity, which is determined by the ratio of the molecular masses of the two gas species: $\sqrt{\frac{M_{\text{He}}}{M_{\text{H}_2}}} = 1.41$, $\sqrt{\frac{M_{\text{Ar}}}{M_{\text{DT}}}} = 2.82$, $\sqrt{\frac{M_{\text{Xe}}}{M_{\text{DT}}}} = 5.11$ [24, 25]. From figure 5, this would mean that for H₂/He more than 27 stages would be required, whereas for DT/Xe 8 stages would suffice (for $EF = 20$, $RF = 90\%$). However, improvement of the selectivity is possible by exploring other transport mechanisms by surface modification of the membrane. A typical example is the synthesis of molecular-sized surfaces such as zeolites [26]. A modest improvement of the selectivity to 2 was obtained experimentally for H₂/He with MFI zeolite mem-

branes at 298 K (used as reference in this work), due to its adsorption properties, decreasing the number of required stages down to 16 [23]. Higher selectivity values could be obtained for SOD-type zeolite membranes with a pore diameter of 0.27 nm, which is in between the kinetic diameters of He (0.26 nm) and H₂ (0.289 nm) [27, 28]. At these conditions, molecular-sieving mechanism (i.e., separation by size) would occur, decreasing further the number of stages. An example of molecular-sieving with small, non-condensable (permanent) molecules has been reported for H₂/N₂ with carbon membranes (0.3–0.5 nm), with a very high selectivity of 725 at 298 K [29]. Since the kinetic diameter of Ar (3.54 nm) is similar to that of N₂ (3.72 nm), similar selectivities would be expected for DT/Ar [27]. With these selectivities, only 4 stages would be required for $EF = 20$ and $RF = 90\%$. In fact, this number of stages would already be reached for $\alpha \geq 17$.

Besides the selectivity, the other two factors which have a direct impact on the number of stages and processing components are EF and RF . In the fuel cycle, high-performance cascades are required for a continuous delivery of the desirable tritiated-enriched stream to the interfacing systems. Although $EF = 20$ and $RF = 90\%$ have been considered as reference case (as proposed in [21]), stricter requirements may be required in the future which would lead to a larger number of stages. A direct implication of the number of stages is the space required to accommodate the stages, connecting pipework and process equipment. More importantly, the volume of gas which is continuously processed by the cascade system impacts the tritium inventory. This aspect is very important since one of the arguments in favor of membranes technology (in contrast to more mature technologies) is its potential to minimize tritium inventory, which is of paramount importance in any technology integrating the fuel cycle as discussed in [30, 31]. Therefore, further studies should be conducted to estimate the actual amount of tritium inside the membrane cascade for various number of stages.

Clearly, another aspects impacting the tritium inventory is the initial feeding flow and tritium concentration. As presented

613 previously, the gas flow downstream of the HCPB breeding
614 blanket is expected to be $10^4 \text{ m}^3 \text{ h}^{-1}$, which is equivalent to
615 121 mol s^{-1} . In contrast, the flow for the separation of the plasma
616 enhancement gases is expected to be 0.14 mol s^{-1} [10]. How-
617 ever, since the flow coming from the blanket is expected to have
618 few ppm of HT and the flow from the plasma exhaust is ex-
619 pected to have 99% of DT, a higher tritium inventory may be
620 expected despite the lower flow for the latter case (provided
621 they have the same number of stages) [32]. Moreover, the feed
622 flow has also a direct impact on the compression power. The
623 values presented in the plot of figure 11 show that, for $\alpha < 3$, the
624 power required is more than 1% of the net power expected for
625 the EU-DEMO (3 GW [5]), which may not be feasible. Never-
626 theless, a strong power reduction would be attained with higher
627 selectivity membranes (for $\alpha > 10$ the power required would
628 be below 0.2%). Furthermore, the feed flow has a direct im-
629 pact on the required surface area per stage which will impact
630 the volume of the vessel accommodating the membrane.

631 Last but not least, the input flows impact directly the perme-
632 ate and retentate flows downstream of the cascade. Regardless
633 of the selectivity, the same order of magnitude is expected for
634 the downstream flows (for $10^4 \text{ m}^3 \text{ h}^{-1}$, the highest flow reduc-
635 tion on the permeate side is around 100). Thus, if the interfac-
636 ing systems require smaller flows for optimum sizing, operation
637 and costs, the feeding flow of the cascade should be reduced.
638 However, this flow reduction is limited by the requirements of
639 the systems upstream (e.g., purging of the breeding blanket for
640 tritium recovery).

641 In sum, the main limiting factors for a cost-efficient, small,
642 yet high-performing, membrane cascade are the selectivity α ,
643 feed flow $F_{f,\text{inj}}$ and required performance parameters EF and
644 RF . Moreover, the design of an optimized cascade has to take
645 into account the requirements of the interfacing systems in the
646 fuel cycle of the fusion reactor.

647 6. Conclusions

648 This paper presents a numerical code developed to size a
649 multi-stage membrane system depending on the following in-

put parameters: selectivity α , pressure-ratio γ enrichment fac-
tor EF and recovery fraction RF . A sensitivity study was per-
formed using this tool to determine the most impacting param-
eters on the dimensioning of membrane cascade systems in view
of the fuel cycle of fusion reactors.

The selectivity α , given by the ratio of the permeances of the
two gas species, and the pressure-ratio γ across the membrane
have a direct impact on the achievable separation of a single-
stage and thus on the number of stages needed to meet certain
performance parameters. These parameters are the enrichment
factor EF and RF , and the higher these values the more num-
ber of stages is required. In the case of the TER for the HCPB
blanket, $EF = 20$ and $RF = 90\%$ have been used as reference,
which would lead to a minimum number of 16 stages for $\alpha = 2$
and 6 for $\alpha = 10$. For larger selectivities, the number of stages
required would be reduced to 4. The injection flow has a direct
impact on the membrane surface-area and compression power
necessary to run a cascade. For instance, with $\alpha = 10$ (i.e.,
6 stages) injection flows of $10^4 \text{ m}^3 \text{ h}^{-1}$ may require 10^2 m^2 and
4 MW. Furthermore, the higher the injection flows the larger is
the footprint of the system and hence the higher is the tritium
inventory, which is a key aspect for the overall tritium manage-
ment in the fuel cycle of the fusion reactor.

Due to the similar sizes of the molecules expected in a fu-
sion reactor, the selectivities are rather limited for the majority
of the membranes available. The increase of the selectivities
towards the range of interest (i.e., lowest number of stages) can
be achieved by either exploring the different adsorption prop-
erties of those molecules onto materials or molecular-sieving.
These separation mechanisms could be achieved with micropor-
ous materials such as zeolite or carbon membranes.

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pressed herein do not necessarily reflect those of the European

687 Commission.

688 Appendix A. Separation factor dependency with α , γ and

689 y_p

690 A two-component (a and b) gas stream feeding a given mem-
691 brane permeates through it with a total flow F_p . Assuming a
692 module with perfect mixing, where the concentrations in the
693 lumen ($x_{r,a}$ and $x_{r,b}$) and permeate ($y_{p,a}$ and $y_{p,b}$) sides are con-
694 stant, equations (A.1) and (A.2) apply for the component flows
695 permeating the membrane.

$$y_{p,a}F_p = \frac{\Pi_a p_f}{t}(x_{r,a} - \gamma^{-1}y_{p,a})A \quad (\text{A.1})$$

$$y_{p,b}F_p = \frac{\Pi_b p_f}{t}(x_{r,b} - \gamma^{-1}y_{p,b})A \quad (\text{A.2})$$

696 Since $y_{p,a} = 1 - y_{p,b} \equiv y_p$ and $x_{r,a} = 1 - x_{r,b} \equiv x_r$, using
697 equation (9), and dividing equation (A.1) by (A.2), the relation
698 (A.3) is obtained. Then, x_r , written as function of S_{stg} and y_p
699 with equation (3), must be replaced in (A.3). After some alge-
700 bra, equation (A.4) is finally obtained which expresses S_{stg} as a
701 function of α , γ and y_p .

$$\frac{y_p}{1 - y_p} = \alpha \frac{x_r - \gamma^{-1}y_p}{(1 - x_r) - \gamma^{-1}(1 - y_p)} \quad (\text{A.3})$$

$$S_{\text{stg}} = \frac{\alpha - y_p(\alpha - 1)\gamma^{-1}}{1 + (1 - y_p)(\alpha - 1)\gamma^{-1}} \quad (\text{A.4})$$

702 Appendix B. Determination of cut using species concentra- 703 tions

704 From the global mass-balance ($F_f = F_p + F_r$) and the com-
705 ponent mass-balance ($F_f x_f = F_p y_p + F_r x_r$), equations, relation
706 (B.1) follows.

$$\frac{F_p}{F_r} = \frac{x_f - x_r}{y_p - x_f} \quad (\text{B.1})$$

707 Since $\nu = \frac{F_p}{F_p + F_r}$, then $\frac{F_p}{F_r} = \frac{\nu}{1 - \nu}$. Thus, by equating the latter
708 with (B.1), equation (19) is obtained after some algebra.

709 References

- 710 [1] I. Cristescu, et al., Commissioning of water detritiation and cryogenic
distillation systems at TLK in view of ITER design, *Fusion Engineering
and Design* 82 (2007) 2126–2132.
- 711 [2] M. Glugla, et al., The ITER tritium systems, *Fusion Engineering and De-
sign* 82 (5-14) (2007) 472–487.
- 712 [3] A. Ciampichetti, et al., Conceptual design of tritium extraction system for
the european HCPB test blanket module, *Fusion Engineering and Design*
87 (6-6) (2012) 620–624.
- 713 [4] D. Demange, et al., A new combination of membranes and membrane
reactors for improved tritium management in breeder blanket of fusion
machines, *Fusion Engineering and Design* 86 (9-11) (2011) 2312–2316.
- 714 [5] G. Federici, et al., European DEMO design strategy and consequences for
materials, *Nuclear Fusion* 57 (2017) 092002.
- 715 [6] A. Brunetti, et al., Membrane technologies for CO₂ separation, *Journal
of Membrane Science* 359 (2010) 115–125.
- 716 [7] V. Violante, et al., Membrane separation technologies: their application
to the fusion reactor fuel cycle, *Fusion Engineering and Design* 22 (3)
717 (1993) 257–263.
- 718 [8] A. Basile, et al., Membrane integrated system in the fusion reactor fuel
cycle, *Catalysis Today* 25 (3-4) (1995) 321–326.
- 719 [9] S. Tosti, A. Pozio, Membrane processes for the nuclear fusion fuel cycle,
Membranes 8 (4) (2018) 1–14.
- 720 [10] S. Tosti, et al., Ceramic membranes for processing plasma enhancement-
gases, *Fusion Engineering and Design* 124 (2017) 928–933.
- 721 [11] D. Demange, et al., Zeolite membranes and palladium membrane reac-
tor for tritium extraction from the breeder blankets of ITER and DEMO,
Fusion Engineering and Design 88 (2013) 2396–2399.
- 722 [12] A. F. Ismail, et al., *Gas Separation Membranes: polymeric and inorganic*,
Springer, 2015.
- 723 [13] K. K. Sirkar, *Separation of Molecules, Macromolecules and Particles:
Principles, Phenomena and Processes*, Cambridge University Press, 2014,
Ch. 9. Cascades, pp. 812–818.
- 724 [14] R. W. Baker, *Membrane Technology and Applications*, John Wiley &
Sons Ltd, 2012, Ch. 8. Gas Separation, pp. 325–378.
- 725 [15] S. Weller, W. A. Steiner, Separation of gases by fractional permeation
through membranes, *Journal of Applied Physics* 21 (1950) 279–283.
- 726 [16] T. C. Merkel, et al., Power plant post-combustion carbon dioxide capture:
An opportunity for membranes, *Journal of Membrane Science* 359 (2010)
727 126–139.
- 728 [17] M. Ohno, et al., Gas separation performance of tapered cascade with
membrane, *Journal of Nuclear Science and Technology* 15 (6) (1978)
729 411–420.
- 730 [18] M. Binns, et al., Strategies for the simulation of multi-component hollow
fibre multi-stage membrane gas separation systems, *Journal of Membrane
Science* 497 (2016) 458–471.
- 731 [19] G. Towler, R. Sinnott, *Chemical Engineering Design: Principles, Practice*

- 756 and Economics of Plant and Process Design, 2nd edition, Elsevier, Ltd.,
757 2013, pp 1221 - 1222.
- 758 [20] H. Nguyen-Schaefer, Rotordynamics of Automotive Turbochargers,
759 Springer-Verlag, 2012, Ch. Thermodynamics of Turbochargers, pp. 21–
760 24.
- 761 [21] O. Borisevich, et al., Zeolite membrane cascade for tritium extraction and
762 recovery systems, *Fusion Science and Technology* 67 (2) (2015) 262–265.
- 763 [22] F. Hernández, et al., A new HCPB breeding blanket for the EU DEMO:
764 Evolution, rationale and preliminary performances, *Fusion Engineering
765 and Design* 124 (2017) 882–886.
- 766 [23] O. Borisevich, et al., Experimental study of permeation and selectivity of
767 zeolite membranes for tritium processes, *Fusion Engineering and Design*
768 98-99 (2015) 1755–1758.
- 769 [24] K. Li, Ceramic Membranes for Separation and Reaction, John Wiley &
770 Sons, 2007, Ch. 4. Transport and Separation of Gases in Porous Ceramic
771 Membranes, pp. 97 – 134.
- 772 [25] National Institute of Standards and Technology, NIST
773 Chemistry WebBook, online, visite on 04/11/2018, URL:
774 <https://webbook.nist.gov/chemistry/mw-ser/>.
- 775 [26] N. Kosinov, et al., Recent developments in zeolite membranes for gas
776 separation, *Journal of Membrane Science* 499 (2016) 65–79.
- 777 [27] S. M. Auerbach, et al. (Eds.), Handbook of Zeolite Science and Technol-
778 ogy, Marcel Dekker, Inc., 2003.
- 779 [28] S. Khajavi, et al., Preparation and performance of H-SOD membranes: a
780 new synthesis procedure and absolute water preparation, in: R. Xu, et al.
781 (Eds.), *From Zeolites to Porous MOF Materials - the 40th Anniversary of
782 International Zeolite Conference*, 2007.
- 783 [29] M. A. L. Tanco, et al., Composite-alumina-carbon molecular sieve mem-
784 branes prepared from novolac resin and boehmite. Part II: Effect of the
785 carbonization temperature on the gas permeation properties, *International
786 Journal of Hydrogen Energy* 40 (2015) 3485–3496.
- 787 [30] B. Bornschein, et al., Tritium management and safety issues in ITER
788 and DEMO breeding blankets, *Fusion Engineering and Design* 88 (2013)
789 466–471.
- 790 [31] Y. Hörstensmeyer, et al., Analysis of the EU-DEMO fuel cycle elements:
791 Intrinsic impact of technology choices, *Fusion Engineering and Design* in
792 press.
- 793 [32] F. Franza, et al., Tritium transport analysis in HCPB DEMO blanket with
794 the FUS-TPC code, *Fusion Engineering and Design* 88 (9-10) (2013)
795 2444–2447.