

Analysis of high flux membranes for desalination in waste-heat driven vacuum membrane distillation plants: Experimental validation and techno-economic analysis

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HIGHLIGHTS

- Evaluation of vacuum membrane distillation for seawater desalination.
- Hyper-concentrated brine at 20 wt% and limited thermal consumption are possible.
- Fitting of numerical models with experimental data for ceramic membranes.
- LCOW of 3–8 \$/m³ for cascade plant layout and next generation ceramic membranes.

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ABSTRACT

Vacuum membrane distillation is a promising technology for seawater desalination, as it enables high recovery ratios, with reasonable thermal and minimal electric consumption. Ceramic membranes can offer notable advantages over polymeric membranes, mainly due to their robust thermal and mechanical stability, yet they have limited representation in the literature. Accordingly, this work investigates ceramic membranes and their techno-economic performance for large scale desalination plants, with target recovery ratios above 85 %. First, a one-dimensional model for multilayered membranes was developed in MATLAB, validated against experimental data of water fluxes with different membrane materials, including ceramic ones, specifically collected within this work. This also enabled the fitting of the membrane characteristic parameters with the collected experimental data. With the aim of reducing the thermal consumption, a full-scale plant layout was defined with various stages in cascade and a sensible waste heat source at 90 °C. Results demonstrate that thermal consumption level in the 180–250 kWh/m³ range is possible, with average water fluxes around 20 kg/(m²·h). With reasonable assumptions on capital costs and plant availability, the levelized cost of water was found to be between 3 and 8\$/m³.

1. Introduction

To address the growing demand for sustainable water resources in various regions, it is essential to develop desalination technologies that are cost-effective, energy-efficient and can achieve high recovery ratios (RR) to avoid issues related to the management of high-salinity brines. Current commercial desalination technologies include pressure-driven reverse osmosis (RO), which accounts for 69 % of the global desalination capacity, along with thermally driven technologies, such as multi-stage flash (MSF) and multi-effect distillation (MED) that account for

25 % of the global desalination capacity [1]. With only 1 % of the world's desalination plants powered by renewable energy and global desalination capacity on the rise, the CO₂ emissions and environmental impact of seawater desalination are expected to increase significantly [2]. At the same time, the development of zero liquid discharge (ZLD) technologies has emerged to address the need for sustainable brine management of high-concentration desalination byproducts [3].

Membrane distillation (MD) is an emerging thermally-driven desalination technology that is not only compatible with low grade waste heat energy, but also has low sensitivity to high salinities in the feed [4,5]. On the other hand, compared to RO, which requires 2.5–4 kWh/

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Nomenclature			
Acronyms			
AAO	Anodic Alumina Oxide	J_W	Mass water permeated flux [$\text{kg m}^{-2} \text{s}^{-1}$]
αAO	Alpha Alumina Oxide	$\dot{m}$	Permeate vapor mass flow rate [kg s^{-1}]
CRF	Capital Recovery Factor	$\dot{m}_{\text{feed}}$	Feed mass flow rate [kg s^{-1}]
HTF	Heat Transfer Fluid	$\dot{m}_{\text{permeate}}$	Permeated mass flow rate [kg s^{-1}]
LCOW	Levelized Cost Of Water production	M_m	Molar mass of water [kg kmol^{-1}]
KPI	Key Performance Indicator	N	Molar water permeated flux [$\text{kmol m}^{-2} \text{s}^{-1}$]
MSF	Multi-Stage Flash	P	Pressure [Pa]
MED	Multi-Effect Distillation	ΔP^*	Driving force of the VMD process [Pa]
MD	Membrane Distillation	P_m	Pressure of the vapor along the membrane [Pa]
PTFE	Polytetrafluoroethylene	$P_{\text{interface}}$	Pressure of the vapor at the interface between two layers [Pa]
RecR	Recirculation Rate of each VMD stage [–]	P_{vacuum}	Vacuum pressure imposed by vacuum pump [Pa]
RR	Recovery Ratio	P_{sat}	Saturation pressure of water [Pa]
RO	Reverse Osmosis	$\dot{q}$	Heat flux across the membrane [W m^{-2}]
STEC	Specific Thermal Energy Consumption	$\dot{Q}$	Heat transferred [W]
VMD	Vacuum Membrane Distillation	r	Average radius of the membrane pore [m]
ZLD	Zero Liquid Discharge	R	Universal gas constant [$\text{m}^2 \text{kg s}^{-2} \text{kmol}^{-1} \text{K}^{-1}$]
		t	Membrane thickness [m]
		T	Temperature [$^{\circ}\text{C}$ or K]
		ΔT_{STAGE}	Feed temperature difference across each VMD stage [$^{\circ}\text{C}$]
		$x_{\text{H}_2\text{O}}$	Molar fraction of water in the feed [–]
Symbols		Greek symbols	
a_0	Structural parameter [–]	α	Convective heat transfer coefficient [$\text{W m}^{-2} \text{K}^{-1}$]
A_{permeate}	Effective mass transport area of membrane [m^2]	$\gamma_{\text{H}_2\text{O}}$	Activity coefficient of water in the feed [–]
b_0	Viscous flow structural parameter [m]	ϵ	Membrane porosity [–]
C_A	Transmission coefficient [$\text{kmol m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$]	μ	Viscosity of the vapor permeated [Pa s]
C_B	Viscous flow coefficient [$\text{kmol s m}^{-2} \text{kg}^{-1} \text{Pa}^{-1}$]	τ	Membrane tortuosity [–]
$C_{p,\text{feed}}$	Feed heat capacity [$\text{J kg}^{-1} \text{K}^{-1}$]		
d	Diameter of the circular membrane [m]		
Δh_{eva}	Evaporation enthalpy of water [J kg^{-1}]		

m^3 of electrical energy [6], MD has demonstrated a significantly lower electrical energy demand (around one order of magnitude lower) [7,8]. Additionally, RO necessitates extensive chemical pre-treatment processes, which not only increase operational complexity but also amplify the environmental impact associated with brine disposal. In contrast, MD employs hydrophobic membranes with a larger pore size and operates under lower transmembrane pressures than RO. This may lessen the need for extensive feed pre-treatment prior MD and reduce the compaction of the scale layer that may form. As a result, MD is less susceptible to irreversible scaling and, consequently, has a lower chemical footprint [9,10]. These notable features position MD as a promising sustainable ZLD technology for concentrating brines to levels

above the 8 wt% salinity limit of the RO technology [3].

While MD is a potential sustainable desalination technology, its low water permeate flux and intensive thermal energy consumption are among the major challenges that must be addressed to enable its commercial deployment [11–13]. Since the thermal energy requirement, for the evaporation of water in MD plants, estimated at 90–98 % of the total thermal input provided [14], the primary strategy to enhance thermal energy efficiency is recovering latent heat from condensing warm permeate vapor streams [15]. Among the four major MD configurations (i.e., Air Gap, Direct Contact, Sweeping Gas and Vacuum), vacuum membrane distillation (VMD) experiences the least conductive losses through the membrane [5,11,16,17]. This translates to higher thermal

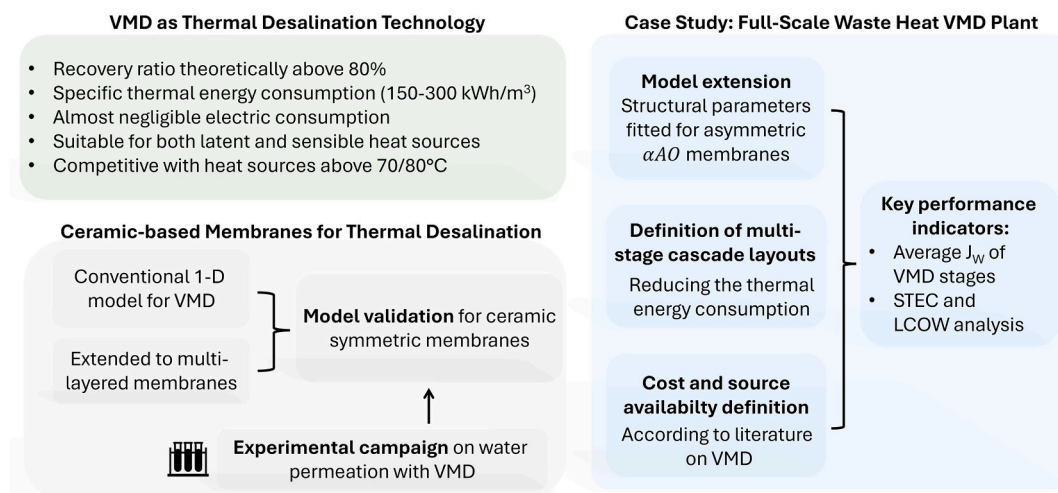


Fig. 1. Flowchart of the methodology developed in this work.

efficiencies, where a higher fraction of the input thermal power is exploited in the evaporation of the feed water. The water flux, on the other hand, is highly dependent on the selected MD configuration [18]. Specifically, comparative experimental studies have shown that VMD has the potential for higher water fluxes than other MD configurations, such as direct contact membrane distillation, primarily due to the low permeate operating pressures in VMD that improve the driving force [19,20].

To date, polymeric membranes have been extensively considered for MD applications, yet the potentially low conductive losses and high fluxes of the VMD configuration make ceramic membranes a viable option despite their intrinsic high thermal conductivity, mass transfer resistance and high production costs. Ceramic membranes offer key advantages due to their superior thermal, chemical and mechanical stability [21]. Moreover, asymmetric α -alumina membranes are particularly suitable for long-term and high-performance VMD applications. For instance, their feasibility for treating hypersaline aqueous feeds (e.g. 35 wt% NaCl) has already been demonstrated in the literature [22], along with their superior long-term stability over 500 h of VMD operation with a feed solution comprising 3 wt% NaCl [23]. However, only a handful of outdated economic analyses have focused on VMD and were based on polymeric membranes [24,25], whereas little emphasis has been placed on evaluating the techno-economic viability of ceramic membranes for VMD desalination applications. Furthermore, the vast majority of membrane material development studies have considered water flux as a key performance indicator when evaluating novel MD membrane materials [26]. In this context, a research gap exists regarding the techno-economic performance of VMD utilizing ceramic membranes. Bridging this knowledge gap will enable the integration of MD with low-grade waste heat sources, making it a viable desalination system for treating high-salinity feeds while potentially reducing the environmental impact of desalination.

Specifically, this work focuses on modelling both developed and commercial membranes and on enhancing the economic viability of the VMD process. The studied membrane materials represent the most extensively evaluated materials for VMD, including a PTFE membrane, an asymmetric α -alumina membrane along with a novel symmetric anodic alumina membrane. Fig. 1 demonstrates the methodology developed in this work, where a one-dimensional model is first implemented based on common theoretical principles and then validated against the experimental water fluxes of two flat-sheet membrane materials: among them, one is anodic aluminum oxide, whose modelling in VMD as membrane material is unprecedented. To model the performance of asymmetric α -alumina membranes, a practical approach to a known challenge in modelling membranes performance is proposed using the validated model and is based on the experimental water fluxes. This approach addresses the challenges associated with determining the characteristic parameters for each membrane layer of the asymmetric α -alumina membrane. Eventually, this study integrates the technical and economic analysis of a novel multistage waste-heat driven VMD plant layouts, based on commercial tubular asymmetric α -alumina membranes. For a preset recovery ratio of 85.5 %, enabled via the incorporation of a recirculation stream over each VMD stage, energy and mass balances were deployed to evaluate the water flux across each stage, and assess the viability of the plant layouts through key performance indicators (KPI), such as the levelized cost of water production (LCOW) and the specific thermal energy consumption (STEC). Consequently, this study adds to the sparse literature on the techno-economic evaluation of VMD for ceramic membranes that have a relatively high-water flux.

2. Modelling of VMD

This section focuses on the description of conventional models for VMD membranes, implemented in this work in MATLAB with a 1-D discretization. The model, which is capable of handling permeation of water across multilayer membranes, is then adopted for both the

comparison with experimental data from this work, and in the simulations of the full-scale VMD plant.

2.1. Mass transfer modelling

The governing equations used to describe the VMD process across a porous membrane are presented here reported. These equations are derived from the work of Zhang [27] and that of Kast and Hohenthanner [28], which were published in the open literature and subsequently cited and applied by other authors for multistage systems [29,30].

For membranes conventionally used in VMD, the pores size is in the range of 0.1 μm to 0.45 μm , where Knudsen numbers are typically in the range of 0.2 to 1.0 [22,31]. This range indicates that mass transfer of freshwater across the membrane pores is jointly dominated by Knudsen diffusion and viscous flow, which aligns with the typical mass transfer characteristics of VMD membranes. Hence, the permeated water flux N is equal to the sum of these two components [32]:

$$N \left[\frac{\text{kmol}}{\text{m}^2\text{s}} \right] = N_K + N_P \quad (1)$$

Where [32]

$$N_K \left[\frac{\text{kmol}}{\text{m}^2\text{s}} \right] = \frac{8}{3} \frac{\epsilon r}{\tau t} \sqrt{\frac{1}{2\pi R M_m T_{\text{wall}}^{\text{feed}}}} \Delta P_s \quad (2)$$

And [32]

$$N_P \left[\frac{\text{kmol}}{\text{m}^2\text{s}} \right] = \frac{1}{8\mu} \frac{\epsilon r^2}{\tau t} \frac{P_m}{R T_{\text{wall}}^{\text{feed}}} \Delta P_s \quad (3)$$

And, in a more compact form:

$$N = (a_0 C_A + b_0 C_B P_m) \Delta P_s \quad (4)$$

Having

$$a_0 [-] = \frac{\epsilon r}{\tau t} \quad (\text{i}) \text{ and } C_A \left[\frac{\text{kmol}}{\text{m}^2\text{sPa}} \right] = \frac{8}{3} \sqrt{\frac{1}{2\pi R M_m T_{\text{wall}}^{\text{feed}}}} \quad (\text{ii}) \quad (5)$$

And

$$b_0 [\text{m}] = \frac{\epsilon r^2}{\tau t} \quad (\text{i}) \text{ and } C_B \left[\frac{\text{kmol} \cdot \text{s}}{\text{m}^2\text{kgPa}} \right] = \frac{1}{8\mu R T_{\text{wall}}^{\text{feed}}} \quad (\text{ii}) \quad (6)$$

Specifically, $T_{\text{wall}}^{\text{feed}}$ [K] is the feed temperature at the surface of the membrane, M_m [$\text{kg} \cdot \text{kmol}^{-1}$] is the water molar mass, R [$\text{m}^2\text{kg} \cdot \text{s}^{-2}\text{kmol}^{-1}\text{K}^{-1}$] is the universal gas constant, μ [$\text{Pa} \cdot \text{s}$] is the viscosity of the vapor across the pores, P_m [Pa] is the mean pressure in the pores, defined as the mathematical average between the pressure on the permeate side (i.e., the vacuum pressure, $P_{\text{vacuum}}^{\text{permeate}}$ [Pa]) and the vapor pressure of water at the wall temperature of the feed side ($P_{\text{sat}}(@T_{\text{wall}}^{\text{feed}})$ [Pa]).

Moreover, a_0 and b_0 are parameters defined as functions of the characteristics of the membrane, and hence are not dependent on the vacuum distillation process itself. In particular, r [m] is the average pore radius of the membrane, ϵ [-] is the membrane porosity, τ [-] is the tortuosity of the membrane pores and t [m] is the membrane thickness.

Eventually, the pressure drop (ΔP_s [Pa]), which is the driving force of the VMD process, is given by:

$$\Delta P_s = P_{\text{sat}}(@T_{\text{wall}}^{\text{feed}}) x_{\text{H}_2\text{O}} \gamma_{\text{H}_2\text{O}} - P_{\text{vacuum}}^{\text{permeate}} \quad (7)$$

being the difference between the saturation pressure of pure water at the wall temperature of the feed ($P_{\text{sat}}(@T_{\text{wall}}^{\text{feed}})$), multiplied by both the molar fraction of water in the feed ($x_{\text{H}_2\text{O}} \left[\frac{\text{kmol}_{\text{H}_2\text{O}}}{\text{kmol}_{\text{feed}}} \right]$) and its activity coefficient ($\gamma_{\text{H}_2\text{O}}[-]$), and the vacuum pressure on the permeate side

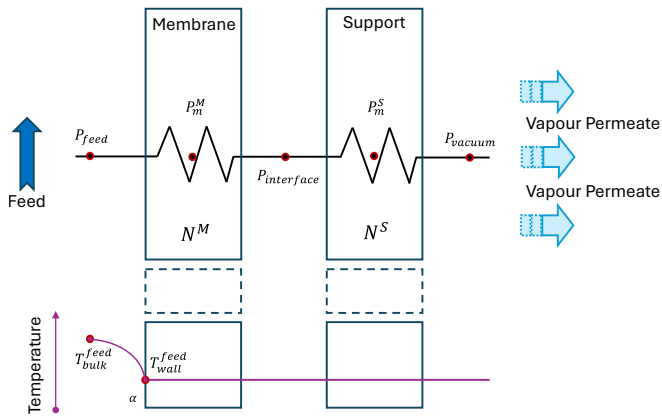


Fig. 2. Schematic of mass and energy transfer processes within both membrane and support layers.

($p_{vacuum}^{permeate}$) imposed by the vacuum pump.

The two factors (i.e., x_{H_2O} and γ_{H_2O}) account for a drop in permeate flux for feeds with high salinity content. A more detailed description of the salinity effects on the driving force ΔP_* is reported in Appendix A.

It's also possible to consider configurations, depending on the membrane characteristics, where the mass transfer process is characterized by two layers in series (Fig. 2), or even more. This typically occurs when a support is added on top of the membrane to mechanically sustain it. Under these circumstances, the formulation of the flux across both the membrane and the support is modelled as follows (M: Membrane and S: Support):

$$N^M \left[\frac{\text{kmol}}{\text{m}^2 \text{s}} \right] = (a_0^M C_A^M + b_0^M C_B^M P_m^M) \Delta P_*^M \quad (8)$$

And

$$N^S \left[\frac{\text{kmol}}{\text{m}^2 \text{s}} \right] = (a_0^S C_A^S + b_0^S C_B^S P_m^S) \Delta P_*^S \quad (9)$$

Specifically, P_m^M is the average pressure in the pores across the membrane layer, P_m^S is the average pressure in the pores across the support layer. Analogously, ΔP_*^M is the driving force between the feed

and the pressure at the interface ($P_{interface}$), while ΔP_*^S is the driving force between the pressure at the interface and the vacuum pressure. As the vapor flux N has to be the same for the whole permeation process, i.e., across both the membrane and the support layer, the numerical model iterates on the pressure of the permeate at the interface between the membrane and support layer $P_{interface}$, such that N^M and N^S are equal.

Similarly, for concentric layers (as in a tubular membrane), the pressure at the interface must be found iteratively, such that:

$$N^1 \cdot d^1 = \dots = N^i \cdot d^i = N^{Last} \cdot d^{Last} \quad (10)$$

is satisfied. Where d is the diameter of the circular section at the interface between two membrane layers, the superscript "1" stands for the inner layer (in contact with the feed), while the superscript "Last" indicates the outer layer, normally intended as support layer, directly in contact with the vacuum pressure.

2.2. Heat transfer modelling

To properly simulate the mass transfer problem across the membrane, the wall temperature on the feed side (T_{wall}^{feed}) must be determined. This value can be determined by solving the energy balance across the membrane, expressed as follows:

$$\dot{q} = \alpha (T_{bulk}^{feed} - T_{wall}^{feed}) = J_w \cdot \Delta h_{eva} \quad (11)$$

Where $\dot{q} [\text{Wm}^{-2}]$ is the thermal power per unit of membrane area required by the VMD process, $\alpha [\text{Wm}^{-2}\text{K}^{-1}]$ is the convective heat transfer coefficient between the feed and the membrane wall, $T_{bulk}^{feed} [\text{K}]$ is the bulk temperature of the feed, $J_w [\text{kg m}^{-2}\text{s}^{-1}]$ the water flux that permeates across the membrane (being $N \cdot M_m$) and $\Delta h_{eva} [\text{J kg}^{-1}]$ is the mass enthalpy of evaporation of water at the feed wall temperature, T_{wall}^{feed} .

In the energy balance, all the thermal power exchanged is provided to the permeate for its evaporation. Moreover, as already assumed in analogous literature works, the thermal problem is approached assuming a negligible contribution of the conductive thermal resistance of the membrane (and the support, if present), due to very limited

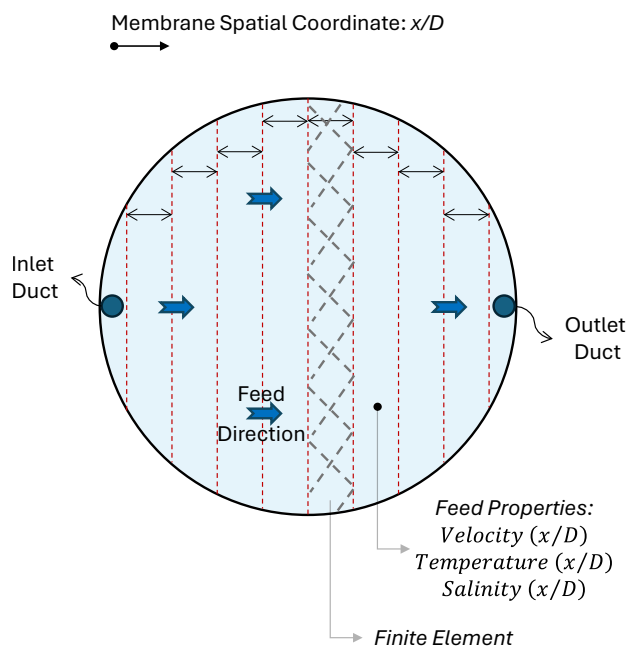


Fig. 3. Qualitative representation of the 1-D discretization of the two geometries of membranes.

thickness, high porosity, and negligible contribution of its thermal conductivity [17,33–35]. To be added the negligible contribution of the convective heat transfer coefficient in the permeate side, due to its extremely low pressure (vacuum pressure) and extremely low permeate flow rate (i.e., the absence of fluid inherently eliminates the possibility of having a convective heat transfer phenomenon). The permeate flux, on the permeate side, is assumed to instantaneously evacuate the system at constant pressure and temperature, without affecting the heat and mass transfer process. A qualitative representation of the temperature profile across the membrane is proposed in Fig. 2 along with the mass resistances: even if the representation refers to the condition with a support of the membrane, it holds validity when the support is not present.

Conventional heat transfer correlations are adopted in this work, reported in Appendix B, whereas the thermodynamic and transport properties of saline water as function of temperature and salinity are adopted according to literature [36].

Therefore, the convective heat transfer coefficient can be determined for each finite-volume considered in the geometry, both for the flat-sheet circular membrane of the experimental setup and for the tubular membrane used in the full-scale desalination plant. In each section of the membrane, all the thermodynamic and transport properties of the feed (including temperature, pressure, salinity, density, etc.) are univocally defined, and the heat transfer correlations are applied.

2.3. Model implementation

The experimental campaign developed in this work adopts flat membranes with circular base, whereas tubular membranes are selected to represent the state-of-the-art of the commercial membranes. In both configurations, the numerical analysis of the VMD process has been carried out with 1-D Matlab model discretizing the geometry in 50 finite elements. As proposed in Fig. 3, the circular membrane is discretized into finite elements along the membrane spatial coordinate (equidistant between each other), perpendicular to the flow direction: in each finite element the feed has defined characteristics in terms of flow rate and properties. For the commercial tubular membrane, on the other hand, a more conventional geometrical discretization along the axis of the straight circular channel has been adopted.

As the process is always dominated by the predetermined vacuum pressure on the permeate side ($P_{\text{vacuum}}^{\text{permeate}}$) the modelling of the problem across finite elements is focused on the permeate side. An iterative calculation is carried out for each finite element, by initially guessing the temperature at the wall of the feed ($T_{\text{wall}}^{\text{feed}}$) based on the respective bulk temperature ($T_{\text{bulk}}^{\text{feed}}$) to solve the mass transfer problem. As J_W is computed, the energy balance on each finite-volume is determined (Eq. (11)). As a last step, the convective heat transfer coefficient is determined through the heat transfer correlations (cf. Appendix B) and the heat flux is compared with the one calculated through Eq. (11). Eventually, the initial guess of $T_{\text{wall}}^{\text{feed}}$ is updated, for each finite volume, up to convergence.

It is important to underline that if a support layer is present, the convergence on the assumed value of $T_{\text{wall}}^{\text{feed}}$ and the convergence on the abovementioned $P_{\text{interface}}$ (the permeate pressure at the interface between the support layer and the membrane), are solved simultaneously, for each finite-volume of the geometry analyzed. The ΔP_*^M and ΔP_*^S defined as:

$$\Delta P_*^M = P_{\text{sat}} \left(@T_{\text{wall}}^{\text{feed}} \right) \cdot x_{\text{H}_2\text{O}} \cdot \gamma_{\text{H}_2\text{O}} - P_{\text{interface}} \quad (12)$$

and

$$\Delta P_*^S = P_{\text{interface}} - P_{\text{vacuum}} \quad (13)$$

Table 1

Characteristics of adopted AAO and PTFE membranes, which were supported on a perforated stainless-steel disc.

Parameter	Properties of membrane top layer	
	AAO membrane	PTFE membrane
Material	Anodic Alumina ^a	PTFE ^a
Average Pore Diameter	200 nm ^a	220 nm ^a
Porosity	20–50 % ^a	85 % ^a
Thickness	55 ± 0.1 μm ^b	78 ± 1 μm ^b
Tortuosity	1 ^c (Assuming cylindrical pores)	1.2 ^c (Estimated as 1/ε [17])

^a Reported by supplier.

^b Measured [37].

^c Estimated.

highlighting the dependency of both of them on the interface pressure as shown in Fig. 2.

Finally, once a solution is reached on the single finite-element, the permeate flux J_W is used to identify the permeate vapor mass flow rate $\dot{m}_{\text{permeate}} [\text{kg} \cdot \text{s}^{-1}]$ by knowing the area of the membrane where permeation occurs, $A_{\text{permeate}} [\text{m}^2]$, according to:

$$\dot{m}_{\text{permeate}}(i) = A_{\text{permeate}}(i) \cdot J_W(i) \quad (14)$$

Hence, the feed mass flow rate on the subsequent finite-element, $\dot{m}_{\text{feed}}(i+1)$, can be computed as:

$$\dot{m}_{\text{feed}}(i+1) = \dot{m}_{\text{feed}}(i) - \dot{m}_{\text{permeate}}(i) \quad (15)$$

Solving the energy balance on the i-th element according to:

$$\dot{Q}(i) = A_{\text{permeate}}(i) \cdot J_W(i) \cdot \Delta h_{\text{eva}} = \dot{m}_{\text{permeate}}(i) \cdot \Delta h_{\text{eva}} \quad (16)$$

the feed temperature on the subsequent element $T_{\text{bulk}}^{\text{feed}}(i+1)$ is computed with

$$T_{\text{bulk}}^{\text{feed}}(i+1) = T_{\text{bulk}}^{\text{feed}}(i) - \frac{\dot{Q}(i)}{C_{p,\text{feed}}(i) \cdot \dot{m}_{\text{feed}}(i)} \quad (17)$$

assuming the feed heat capacity $C_{p,\text{feed}}(i)$ as function of the feed temperature and salinity.

3. Experimental methods

To validate the numerical VMD model, an experimental campaign was conducted to evaluate the water flux J_W at various feed temperatures and for a range of membrane materials, including symmetric and asymmetric membranes. The results of the models are then compared to the experimental water flux values in the next chapter.

3.1. Membrane materials

Three categories of membranes were adopted in the experimental campaign. First, flat-sheet anodic aluminum oxide membranes (Ano-disc25, Cytiva Life Sciences) supplied with a polypropylene support ring were hydrophobized using a silane grafting method previously described [37]. The membranes had a diameter of 21 mm and will be referred to as the AAO membranes. Secondly, a flat-sheet polytetrafluoroethylene (PTFE) membranes (FGLP02500, Merck Millipore Ltd) supplied with a polyethylene support mesh were also evaluated. The membranes had a diameter of 25 mm and will be referred to as the PTFE membranes. To mechanically support the AAO and PTFE membranes during the VMD experiments and enable precise estimation of their effective transport area, the polypropylene supports were replaced by a stainless-steel 316 support disc with a diameter and thickness of 25 mm and 3 mm, respectively (Figure S. 1 and Figure S. 2). The stainless-steel

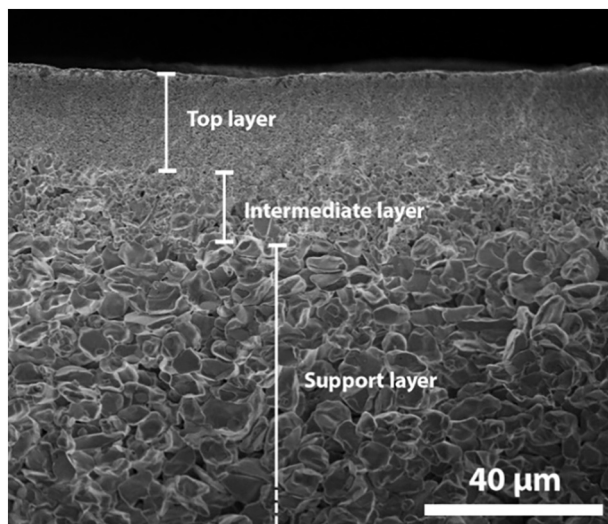


Fig. 4. Cross-sectional SEM image of the layers making up the α AO membranes.

support disc had 65 pores each with a diameter of 1 mm. Table 1 lists the characteristics used for the VMD performance modelling of these two membranes.

Finally, asymmetric flat-sheet membranes (Fraunhofer IKTS, Germany) with a diameter of 25 mm and thickness of either 1 mm or 3 mm (herein referred to as the α AO-1 and α AO-3 membranes) were also evaluated and hydrophobized using a silane grafting method previously described [37]. These membranes consist of three layers, a top layer, an intermediate layer and a support layer (Fig. 4), whose surface topography has been characterized through scanning electron microscopy (SEM) and atomic force microscopy (AFM) in previous work [23,37], and their properties are listed in Table 2. These asymmetric membranes are also adopted in our VMD full scale plant simulation, where tubular α AO membranes are considered instead of flat sheet ones. Table 2 lists the properties of the flat-sheet and tubular membranes, as reported by the supplier (Fraunhofer IKTS, Germany).

3.2. VMD experiments

Two VMD experimental setups were utilized to evaluate the flux

performance of the selected membranes, which mainly differ in the permeate circuit design and the method for flux estimation. Both setups had the feed water circulated using a pump and heated to the set temperature using either heating cords or a heat exchanger connected to a heating circuit. Vacuum was maintained within the permeate circuit using either a dry multi-stage roots pump (ACP 40, CV version, Pfeiffer Vacuum GmbH, Germany) or a diaphragm pump (N 920 G, KNF, Germany). In both setups, the permeate pressure was monitored using electronic pressure transducers. For the setup utilizing the dry multi-stage roots pump, the membrane flux was estimated online using a mass spectrometer (MS, GAM 400 – gas analysis system, InProcess Instruments GmbH, Germany) and using the weight of the collected permeate during 5 min time intervals. The setup utilizing the diaphragm pump collected the vapor drawn from the membrane module in a vessel under vacuum. Its weight was recorded through a PLC program, thus allowing for real-time estimation of flux over time. Full details of the experimental setups were previously described [23,37], and their schematic diagrams are provided in the supplementary data (Figure S. 1 and Figure S. 2).

The membranes were tested in two modules designed for flat-sheet membranes with a diameter up to 25 mm. The geometry of the feed channel in both modules is schematized in Fig. 5, where the feed entered through an inlet duct on the left side, flowed through a circular channel with a height of 2 mm, and exited through an outlet duct on the right side. Permeation occurred only through the top side of the channel that is in contact with the membrane surface. The membrane was sealed using rubber gaskets and supported with a stainless-steel disc, for the case of the AAO and PTFE membranes. Figure S. 1 shows the stainless-steel support disc having a well-defined permeation area (51 mm^2), which was proved to reduce the effective transport area used for the membrane flux estimation [37].

The permeate water flux (J_w) across the three membranes was estimated at three different feed water temperatures (around 60°C , 70°C and 80°C) with a feed flow rate at $30 \times 10^{-3} \text{ m}^3/\text{h}$ ($8.33 \times 10^{-3} \text{ kg s}^{-1}$). The AAO and PTFE membranes were evaluated with the setup having a dry multi-stage roots vacuum pump and with a permeate pressure set at 2 kPa. Within the membrane module, a thermocouple monitored the temperature within the feed channel. In contrast, the α AO membrane was evaluated using the setup utilizing the diaphragm vacuum pump, where the permeate pressure was set at 5 kPa and thermocouples were placed within the inlet and outlet ducts of the membrane feed channel.

Table 2

Characteristics of adopted asymmetric α AO membranes.

Parameter	Membrane top layer	Membrane intermediate layer	Membrane support layer	Source
Flat-sheet α AO-3 membrane				
Average Pore Diameter	200 nm	800 nm	2.5 μm	Reported by supplier
Porosity	43 %	38 %	38 %	
Tortuosity	1.5	3.9	3.2	
Thickness	$21 \pm 0.1 \mu\text{m}^a$	$13 \pm 0.1 \mu\text{m}^a$	3 mm	Measured
Flat-sheet α AO-1 membrane				
Average Pore Diameter	200 nm	800 nm	2.5 μm	Reported by supplier
Porosity	43 %	38 %	38 %	
Tortuosity	1.5	3.9	3.2	
Thickness	$25 \pm 0.2 \mu\text{m}^b$	$11 \pm 0.8 \mu\text{m}^b$	1 mm	Measured
Tubular α AO membrane				
Average Pore Diameter	200 nm	800 nm	3 μm	Reported by supplier
Porosity	39 %	38 %	33 %	
Tortuosity	1.5	3.9	3.2	
Thickness	30 μm	15 μm	1.5 mm	

^a Measured [37].

^b Measured [23].

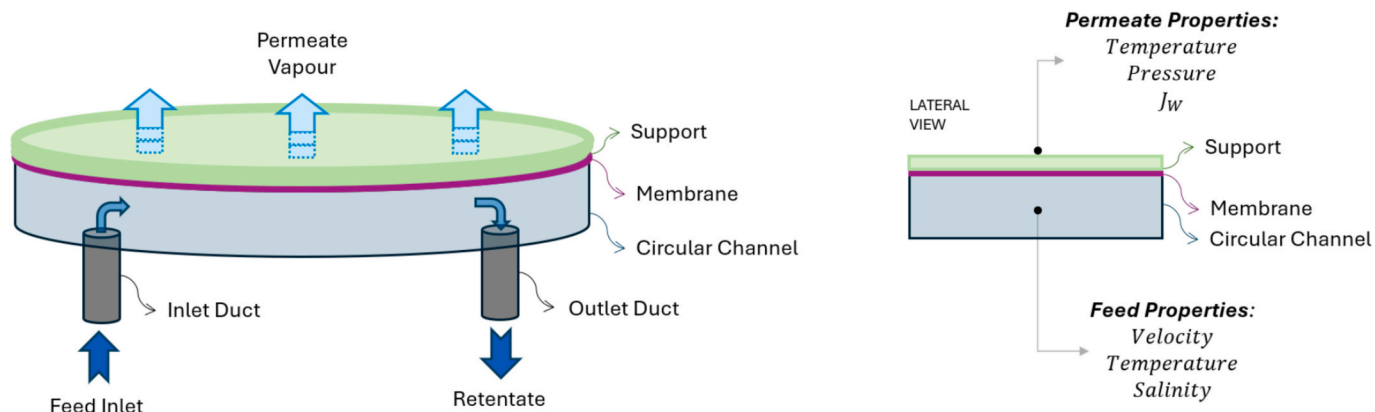


Fig. 5. Schematic diagram of the feed channel in the membrane modules.

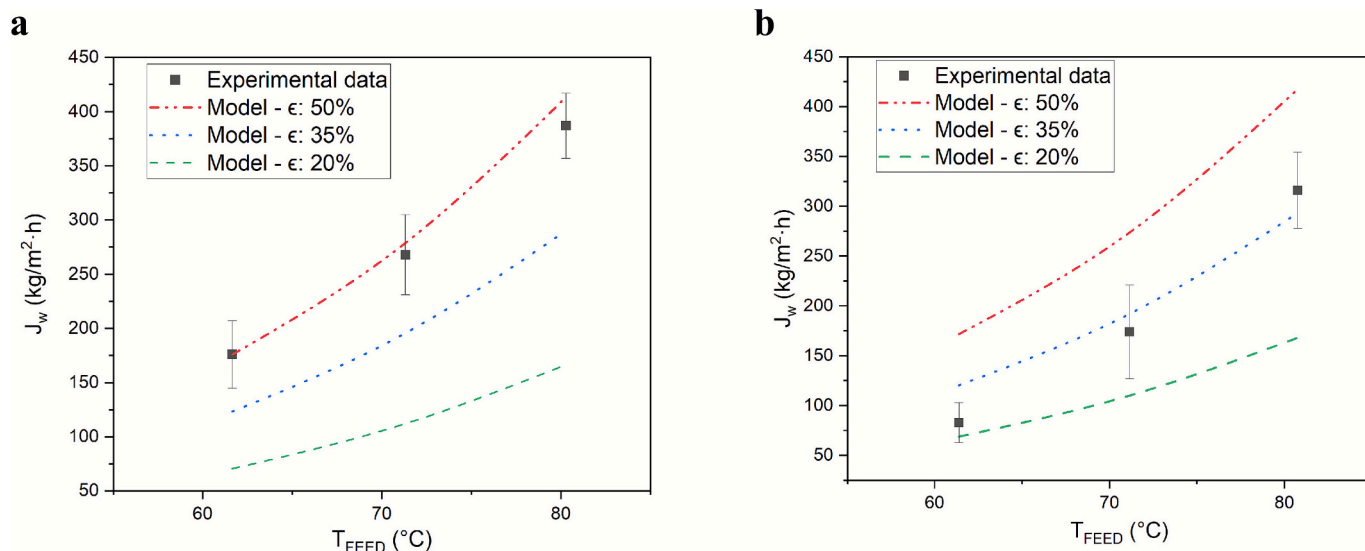


Fig. 6. Modelled (dotted lines) and measured (squares) water fluxes of AAO membranes for a feed of (a) ultra-pure water and (b) an aqueous solution of 3 wt% NaCl. Results are provided with respect to the feed temperature, for feed flow rate at 30 L/h and the $P_{vacuum}^{permeate}$ at 50 mbar. The error bars depict the standard deviation of the respective average flux values.

4. Experimental and model results

4.1. Model validation with symmetric membranes

The experimental permeate water fluxes J_w were used to validate the 1-D model described in Section 2. To demonstrate the wide applicability of the model, commonly used in literature to describe and predict the water flux in VMD, this was validated across a wide range of membrane materials with different characteristic properties. Accordingly, the water fluxes of symmetric and asymmetric ceramic membranes were modelled, along with a polymeric membrane, to be compared against the water fluxes experimentally measured within this work. The experimental results, correlating the J_w to their measured conditions, are tabulated in the supplementary materials (Table S.1 and Table S.2).

The water flux J_w of the AAO membranes was previously measured [37], and modelled as a function of the feed temperature for both an ultra-pure water feed and an aqueous feed solution of 3 wt% NaCl (Fig. 6). While the porosity of the membrane was specified as a range by the manufacturer (50 %–20 %), the model was evaluated at porosities of 50 %, 35 % and 20 %. For a feed of ultra-pure water, the modelled water flux increased with the increase in porosity and the experimentally measured water fluxes were in good agreement with the modelled flux at a porosity of 50 % (Fig. 6(a)). The increase of the NaCl concentration in

the feed water lowers the water partial pressure on the feed side of the membrane by 2 %. While the water flux is linearly related to the partial pressure difference across the membrane, the water partial pressure reduction was reflected on the modelled VMD flux. For instance, modelled water flux of the AAO membrane for an aqueous feed solution of 3 wt% NaCl at 50 °C was only 2 % lower than for an ultra-pure feed at a similar temperature (Fig. 6(b)). In contrast, the experimentally measured water fluxes of the AAO membrane were lower with a flux reduction range of 18–53 % for the three feed-temperatures tested. With the increase of the NaCl concentration from 0 to 3 wt% in the feed water, the experimental water flux of the AAO membrane was reduced from 176 ± 31 kg/(m²·h) to a value of 83 ± 20 kg/(m²·h) for a feed at approx. 60 °C. This large effect of reduction of the experimental flux during MD operation may be attributed to surface scaling and fouling [38,39]. In particular, scaling may be induced by concentration polarization, whose effect is more prominent for membranes with higher fluxes [40]. Considering the relatively high-water flux of the AAO membrane, surface scaling is highly probable and may be responsible for the flux reduction observed experimentally. However, helium permeance decreased by 14 % after these VMD tests, confirming internal pores obstruction, despite that the membrane achieved NaCl rejection as high as 99.8 % [37]. Long-term operational VMD tests revealed that surface erosion and structural degradation were responsible for the internal

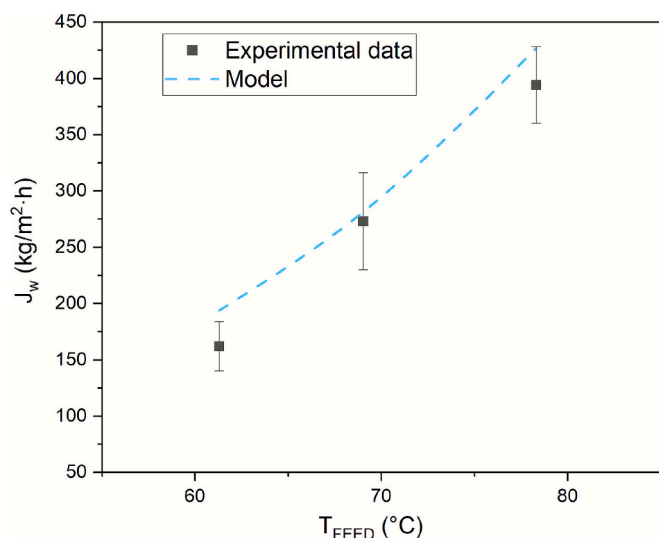


Fig. 7. Modelled (dotted lines) and measured (squares) fluxes of PTFE membrane for a feed of ultra-pure water. Results are provided with respect to the feed temperature, and for the feed flow rate at 30 L/h and the $P_{\text{vacuum}}^{\text{permeate}}$ at 20 mbar. The error bars depict the standard deviation of the respective average flux values.

membrane pores blockage of the AAO membrane, as observed through SEM imaging [23]. While the membrane permeability is directly related to the membrane porosity, reduced permeability due to pores blockage appears here as though the membrane porosity is reduced. In fact, the experimental fluxes for an aqueous feed solution of 3 wt% were closer to those modelled with a membrane porosity of 35 % and 20 %. Accordingly, the evaluation of the water flux for a range of porosities also enabled illustrating the significance of the membrane porosity on its water flux. Details on the experimental testing conditions of the AAO membrane, such as of the measured feed temperature and the imposed permeate pressure are provided along with the measured water flux values (J_w) in Table S.1 and Table S.2.

For similar feed temperatures, the PTFE membrane was experimentally tested [37]. Yet only for an ultra-pure water feed due to the compromised NaCl rejection observed for similar commercial PTFE membranes [17]. The modelled water flux values for the PTFE membrane were also consistent with those experimentally measured (Fig. 7). Modelled and experimental water fluxes increased with the increase in the feed temperature, as expected by the theoretical background of VMD. Moreover, the modelled water flux values for the PTFE membrane were slightly higher (Fig. 7) than those of the AAO membrane (Fig. 6 (a)). For instance, the modelled water flux of the PTFE membrane was at 193 kg/(m²·h), while that of the AAO membrane was estimated at 176 kg/(m²·h) for a feed water temperature of approx. 60 °C. This may be attributed to the slightly higher feed temperature for the PTFE membrane (Table S.1), but also due to its larger pore size and higher porosity (Table 1). In contrast, the cylindrical pores of the AAO membrane offered a shorter path for vapor transport and contributed to its higher water flux, despite the lower porosity compared to the PTFE membrane. Therefore, it may be inferred that the membrane pores tortuosity has a significant contribution to enhancing the water flux of hydrophobic VMD membranes.

4.2. Model extension for asymmetric alumina membranes

Regardless of the application, the asymmetric structure of asymmetric α AO membranes posed challenges for modelling its performance due to the difficulty in determining the characteristic parameters for each membrane layer (namely the porosity, tortuosity, and pore size [41]). Different approaches were reported in literature to quantify these

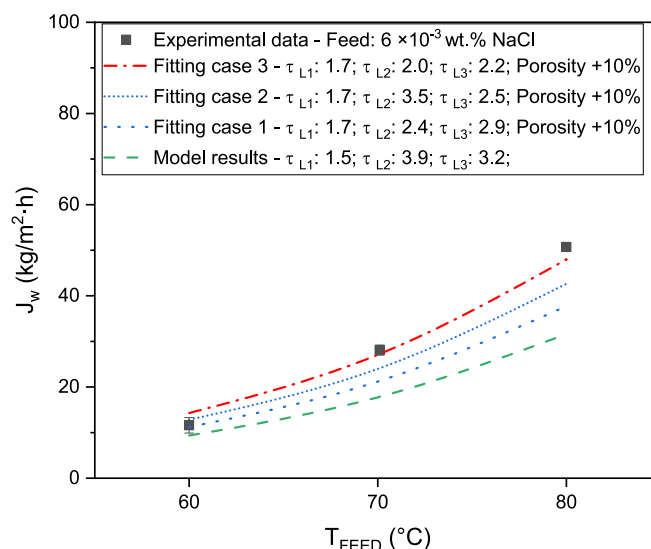


Fig. 8. Modelled (dotted lines) and measured (squares) fluxes of α AO membranes. Results are provided with respect to the feed temperature, and for the feed flow rate at 30 L/h, $P_{\text{vacuum}}^{\text{permeate}}$ at 20 mbar and an aqueous feed solution of 6×10^{-3} wt% NaCl.

parameters with the aim of describing the overall transport of these membranes for pervaporation applications. One method to simplify describing the overall transport of these membranes is to neglect the composite character of these asymmetric membranes, which seemed reasonable considering that Bruijn et al. observed a difference of a maximum 6 % for pervaporation applications [41]. While this method simplifies the modelling of asymmetric inorganic membranes, it overlooks assessing the effect of each layer's characteristic parameters on the overall vapor mass transfer through these membranes. The effect of these characteristic parameters is of particular interest for tailoring the development of ceramic membranes for VMD applications. For that, another method involves quantifying these parameters simultaneously to the production process of these membranes. Utilizing uncoated support along with supports coated with one or two microfiltration layers, researchers could determine the characteristic parameters of each layer by utilizing gas permeation tests [42,43]. However, parameters determined using this approach for describing the water flux of VMD membranes required the introduction of a correction factor to enhance the fit between the modelled and experimental water fluxes [22]. By using average values that lack confidence intervals as input for semi-empirical equations, this method also limits the model into generating specific water flux values rather than a range [22]. Using this method, it is particularly challenging to determine the tortuosity of each layer for asymmetric membranes, where each additional layer may impact the tortuosity of the preceding layer. The α AO membranes supplier utilized this method to determine the tortuosity of each layer (Table 2). Using these input values into the model, the experimental water flux of the α AO-3 membranes was in agreement with the modelled values only at the 60 °C feed temperature and deviation from the model fluxes was observed with increasing feed temperatures (Fig. 8). Therefore, an iterative approach was adopted in this work within a reasonable range of tortuosity values for similar ceramic membrane materials [22,41,42].

The flat-sheet α AO-3 membranes consisted of three layers, a top, an intermediate and a support layer (Fig. 4), where each had distinct characteristic parameters and a different thickness (Table 2). Enhancements of the model fit may be achieved for lower tortuosity values of the different membrane layers (*top tortuosity* τ_{L1} , *intermediate layer tortuosity* τ_{L2} , and the *support layer tortuosity* τ_{L3}). Three fitting procedures (ref. to Fig. 8) were performed to assess this effect:

Table 3

Fitted characteristic parameters for simulation of asymmetric tubular α AO membranes.

Parameter	Membrane top layer	Membrane intermediate layer	Membrane support layer
Material	Alpha Alumina	Alpha Alumina	Alpha Alumina
Average Pore Diameter	0.2 μm	0.8 μm	3 μm
Porosity	43 %	42 %	36 %
Tortuosity	1.7	2.0	2.2
Thickness	30 μm	15 μm	1500 μm

- Fitting Case 1: With a decrease of the support tortuosity τ_{L3} from the initial value of 3.2 to 2.9 increased the model fluxes by approximately 20 % relative to the original modelled values and the overall modelled flux error decreased to 17 % (Relative to the experimental flux values). The enhanced fitting may seem like a combined effect of decreasing τ_{L3} along with the tortuosity of the intermediate layer τ_{L2} from 3.9 to 2.4.
- Fitting Case 2: When the support tortuosity τ_{L3} was further decreased to a value of 2.5 (with an increase of the intermediate layer tortuosity τ_{L2} to a value of 3.5), the model fluxes still increased by approx. 35 % relative to the original modelled values. This also reduced the modelled flux error to 14 % with respect to the experimental flux values. Variation in the top layer tortuosity also had a similar marginal effect on the membrane water flux. The marginal influence of τ_{L1} and τ_{L2} may be attributed to their lower thickness, as compared to the support.
- Fitting Case 3: With a further reduction of the support layer tortuosity τ_{L3} to a value of 2.2, a further error reduction was observed to a value of 11 % compared to the experimental flux values. Therefore, this fitting procedure reveals the strong effect of the support layer tortuosity, considering its significantly higher thickness.

For a 3 wt% NaCl aqueous feed solution at 80 °C, the experimentally measured water flux of the α AO-3 membrane was at $50 \pm 5 \text{ kg}/(\text{m}^2\cdot\text{h})$ (Table S.2) [23]. Using fitting case 3, the modelled flux for the α AO-3 membrane was $47 \text{ kg}/(\text{m}^2\cdot\text{h})$, which is in good agreement with the experimental flux.

For a 6×10^{-3} wt% NaCl aqueous feed solution at 80 °C, the experimentally measured water flux of the α AO-1 membrane was $107 \pm$

$2 \text{ kg}/(\text{m}^2\cdot\text{h})$ (Table S.1). Applying the fitting procedure for this membrane enhanced the fitting between the modelled and experimental flux. For instance, when the characteristic properties provided by the supplier were used, the modelled flux for the α AO-1 membrane was at $75 \text{ kg}/(\text{m}^2\cdot\text{h})$. In contrast, when fitting case 3 properties were used, the modelled flux for this membrane was at $104 \text{ kg}/(\text{m}^2\cdot\text{h})$, which is in good agreement with the experimental flux.

The proposed model fitting procedure, for the α AO membrane tortuosity values, enhanced the characteristic parameters of the α AO membranes. Accordingly, the tortuosity and porosity of the tubular α AO membranes were adapted based on the results of the flat sheet membranes. In fact, the tubular α AO membranes' porosity was increased by 10 % with respect to the characteristics provided by the supplier, which is within the 10 % confidence interval indicated by the supplier. According to the fitting procedure carried out in this work, the top layer tortuosity was increased by 10 %, the intermediate layer tortuosity was reduced by 50 % and the support layer tortuosity was reduced by 31 %. Considering the marginal effect of the top layer tortuosity on the modelled water flux, it was adapted to be in the range of similar membranes reported in the literature [22,42]. The adapted characteristic parameters of the tubular α AO membranes for the subsequent plant simulation are summarized in Table 3.

The asymmetric α AO membrane demonstrated a superior long-term stability over 500 h of VMD operation with 3 wt% NaCl in the aqueous feed solution [23]. Even though the superior long-term flux stability of the asymmetric α AO membrane was not demonstrated at the highest feed salinity levels simulated in the plant (up to 20 wt%), the long-term durability results show the strong potential of these ceramic membranes. Considering their potential and scalability prospect, the aim of simulations presented in the next chapter is to carry out a preliminary techno-economic analysis of a VMD plant based on the tubular asymmetric α AO membrane. Despite the small effect of the model fitting on the flux values, this procedure may be useful as a guide for the future developments of ceramic membranes for VMD applications. It also highlighted the need for a robust, yet simplified, approach to enable determining the characteristic parameters and modelling of asymmetric ceramic membranes.

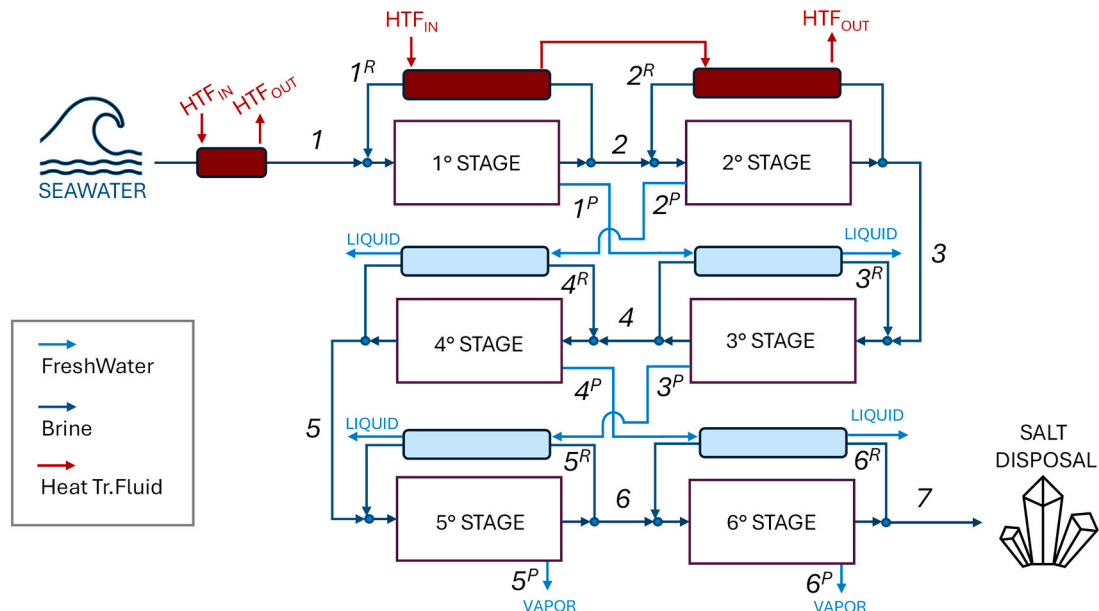


Fig. 9. Plant layout of the full scale VMD plant with 6 stages for $RR = 85.5 \%$.

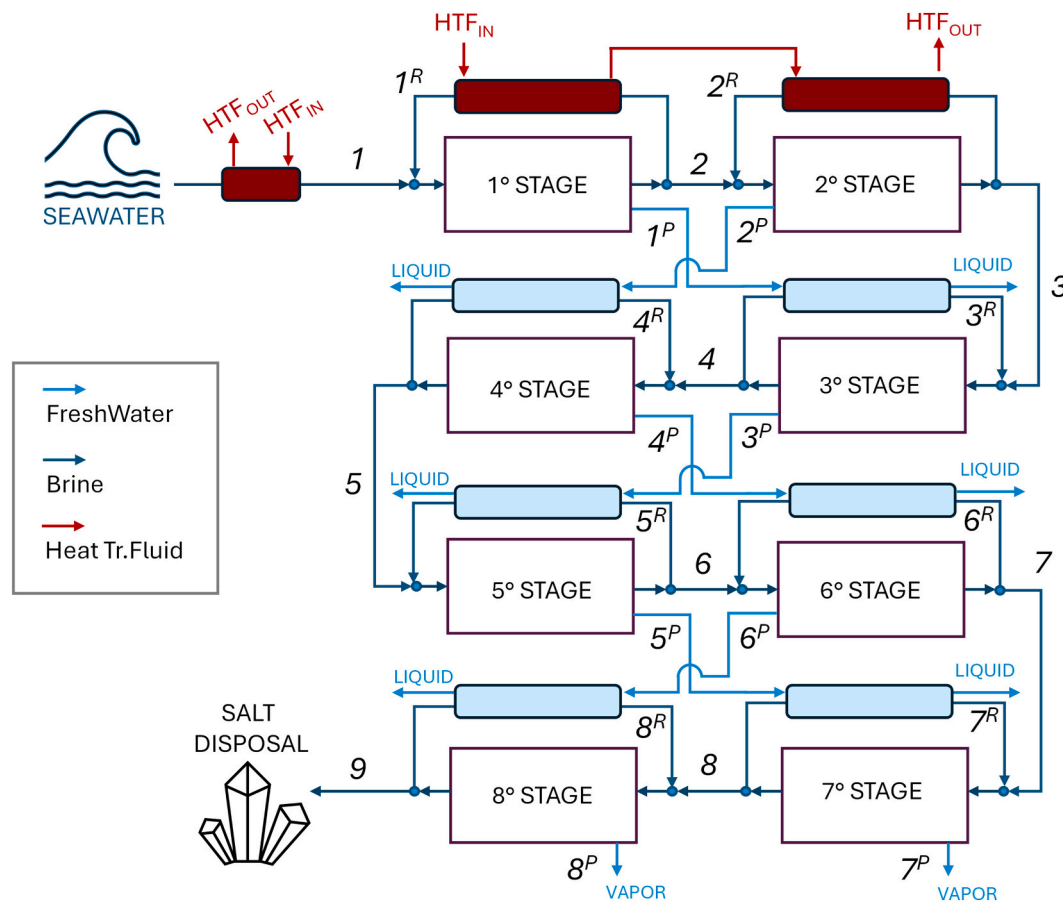


Fig. 10. Plant layout of the full scale VMD plant with 8 stages for $RR = 85.5\%$.

5. Case study: techno-economic analysis of a waste-heat driven VMD plant

In this section, two multi-stage VMD plant layouts were investigated in a techno-economic perspective. The objectives were to reduce the plants STEC through latent heat recovery, and to achieve a high recovery ratio, possibly reaching near-zero liquid discharge conditions. For the economic analysis, it must be underlined that the high production cost of ceramic membranes is often regarded economically prohibitive for the MD processes [44,45]. This is expected considering that these membranes are not yet available for large-scale production. The capital costs for the primary plant equipment were estimated at the preliminary engineering stage for scope estimation purposes. That is relying on information from equipment manufacturers and reported data in peer-reviewed literature. A sensitivity analysis for the LCOW was also conducted to evaluate the impact of variations in the annual plant availability.

5.1. Investigated VMD plant layouts for near-zero liquid discharge

Single stage thermal desalination processes entail an STEC of around $660 \text{ kWh}_{\text{th}}/\text{m}^3$ (the evaporation enthalpy of water at temperatures in the range $40\text{--}80^\circ\text{C}$). Therefore, multiple stages in cascade are normally adopted in order to boost the thermal performance by reducing the STEC [15]. In this work, two VMD plant layouts considered had either 3 cascades of effects or 4 cascades of effects (Fig. 9 and Fig. 10), where each cascade consisted of two stages. A stage was composed of 10 commercial asymmetric tubular αAO membranes in series, whose characteristics are summarized in Table 3. The feed flow rate was defined in the tubular section so that turbulence is fully experienced, with a mass flow per unit of cross flow area of $1000 \text{ kg}/(\text{s}\cdot\text{m}^2)$. The brine

concentration is increased from an inlet concentration of 3.5 wt% NaCl to an outlet concentration of 20 wt% (RR = 85.5 %). A subsequent ZLD plant, not considered in this work, can be adopted to push the overall RR from 85.5 % to the crystallization point at 89 %, or eventually even further through salts precipitation.

Heat was supplied to the VMD plant layouts through the primary heat exchangers (red boxes in Fig. 9 and Fig. 10) from a sensible heat source at 90 °C, represented by the heat transfer fluid (HTF). The heat source characteristics are not specified to present a general case that can be of use in various contexts and in different applications. The heat source is assumed to be available in a steady state, while its intermittence is accounted for through a sensitivity analysis on the annual plant availability. The temperature of the feed to the first stage was at 87 °C and the pinch between the HTF and the plant feed was 3 °C. The lowest saturation temperature was at the last cascade effects, which was at least 25 °C. All odd-numbered stages (1st, 3rd, 5th, 7th) are in cascade with respect to each other, to ensure a minimum temperature difference guaranteed by the presence of the even-numbered stages (2nd, 4th, 6th, 8th), that alternates with the first with respect to the feed flow direction. Analogously, all the even-numbered stages (2nd, 4th, 6th, 8th) are in cascade with each other, for the same reason.

The heat source temperature variation (ΔT_{HTF}) is adopted as a sensitivity parameter, and it was assumed ranging between 6 °C and 18 °C. This range is representative of many applications, including heat recovery from exhaust gases at the stack of power generation or chemical plants. Heat source temperature variation directly influences the temperature differences of the feed across each stage (ΔT_{STAGE}), which was assumed to be constant over all the stages and was defined as:

$$\Delta T_{STAGE} = T_1 - T_2 = T_2 - T_3 = T_{n-1} - T_n = T_{last-1} - T_{last} \quad (18)$$

The heat source temperature variation was evaluated assuming a

Table 4

Full scale VMD plant characteristics.

Parameters	Value	Input/Output
Seawater salinity	3.5 wt%	Input
Salinity at outlet of VMD	20 wt%	Input
Recovery ratio	85.5 %	Input
Inlet seawater temperature	20 °C	Input
Inlet temperature of HTF	90 °C	Input
Feed temperature at inlet (T_1)	87 °C	Input
ΔT_{STAGE}	3 °C to 9 °C	Input (Sensitivity analysis)
$T_2, T_3, T_4, \dots T_n$	Computed from ΔT_{STAGE}	Input (Sensitivity analysis)
ΔT_{HTF}	Computed from ΔT_{STAGE}	Input (Sensitivity analysis)
	6 °C to 18 °C	
Pinch point at condenser (i.e: $T_{1P}-T_{3R}$)	1 °C	Input
Recirculation rate at first stage ($\dot{m}_{R1}$)	Computed	Output
Recirculation rate at second stage	As $RecR_1$	Output
Recirculation rate at subsequent stages	Computed	Output
STEC	Computed	Output
Average J_W per stage	Computed	Output
Membrane area per stage	Computed	Output

constant pinch at the cold end and hot end of the two primary heat exchangers. Thus, resulting in twice the ΔT_{STAGE} , as the recirculation rate in the first and second stage is assumed to be equal.

With respect to the feed flow rate (stream 1), the vapor permeation and freshwater separation of each stage were enhanced by adding recirculation streams ($1^R, 2^R, \dots N^R$ in Fig. 9 and Fig. 10) over each stage. These recirculation streams introduce a higher thermal power as input to each stage. Therefore, a much higher RR (85.5 %) was possible in this work for the same number of stages and STEC. This study assumes that adequate pre-treatment is implemented to minimize membrane fouling. Consequently, the effects of fouling were not considered in this work. It should be noted that this assumption may limit the applicability of the

results to cases where effective fouling control is achieved. For simplicity, the recirculation rate at the first stage ($\dot{m}_{R1}$) and at the second stage ($\dot{m}_{R2}$) were assumed equal and were defined according to:

$$RecR_1 = \frac{\dot{m}_{R1}}{\dot{m}_1} = RecR_2 = \frac{\dot{m}_{R2}}{\dot{m}_2} \quad (19)$$

Values of the recirculation rates were defined for each VMD plant simulation, computed iteratively to meet the plant target outlet salinity at the set energy input from the primary heat exchangers. The recirculation rates of subsequent stages were computed based on the permeate flow rate of the upstream stage, along with the temperature gain of the recirculation streams. In fact, as the energy balance at the condensers of the

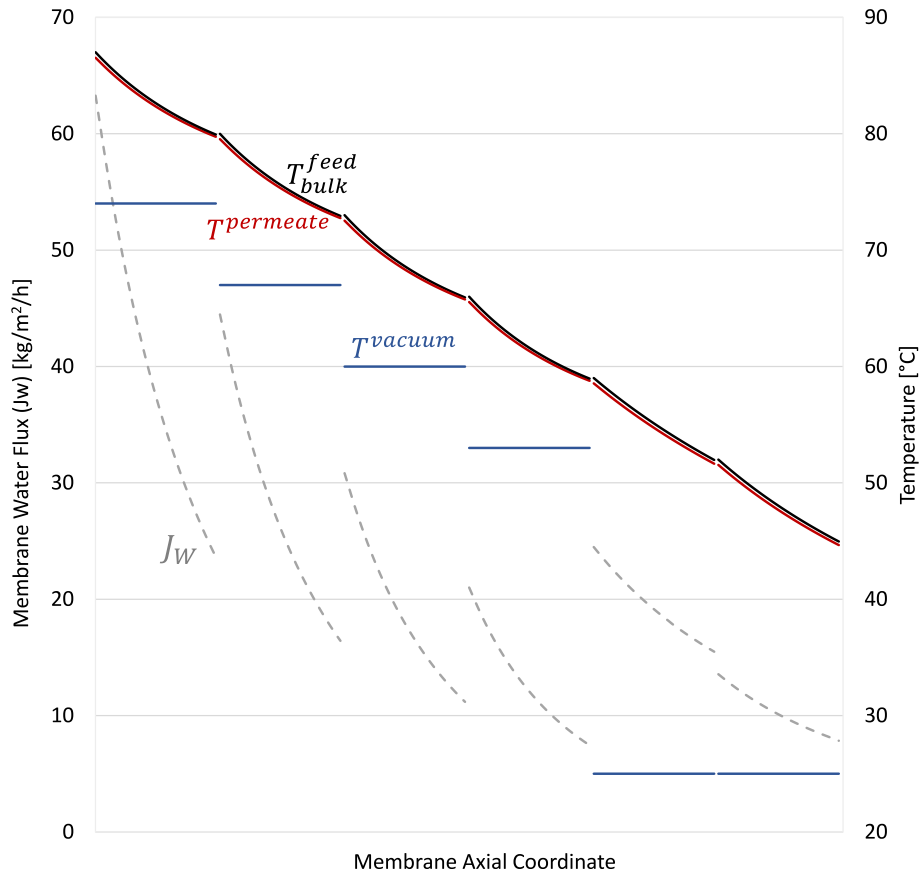


Fig. 11. Axial trend of temperature and water flux across the 6 stages layout with $\Delta T_{STAGE} = 7$ °C.

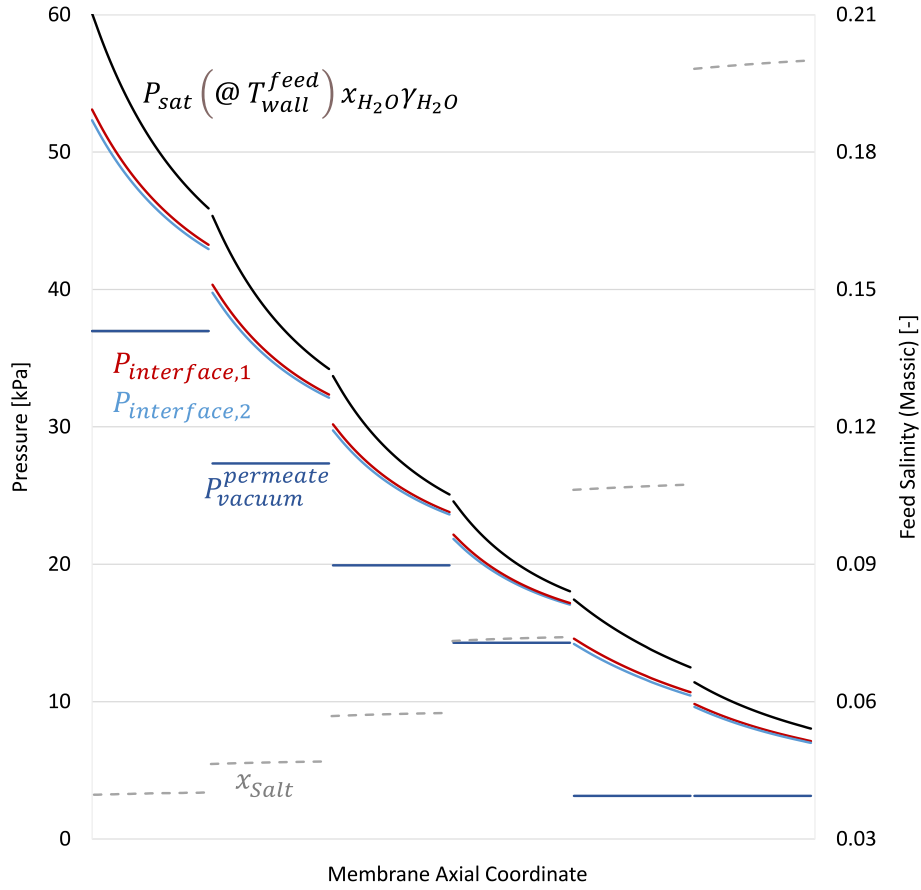


Fig. 12. Axial trend of feed salinity and pressures across the 6 stages layout with $\Delta T_{\text{STAGE}} = 7^\circ\text{C}$.

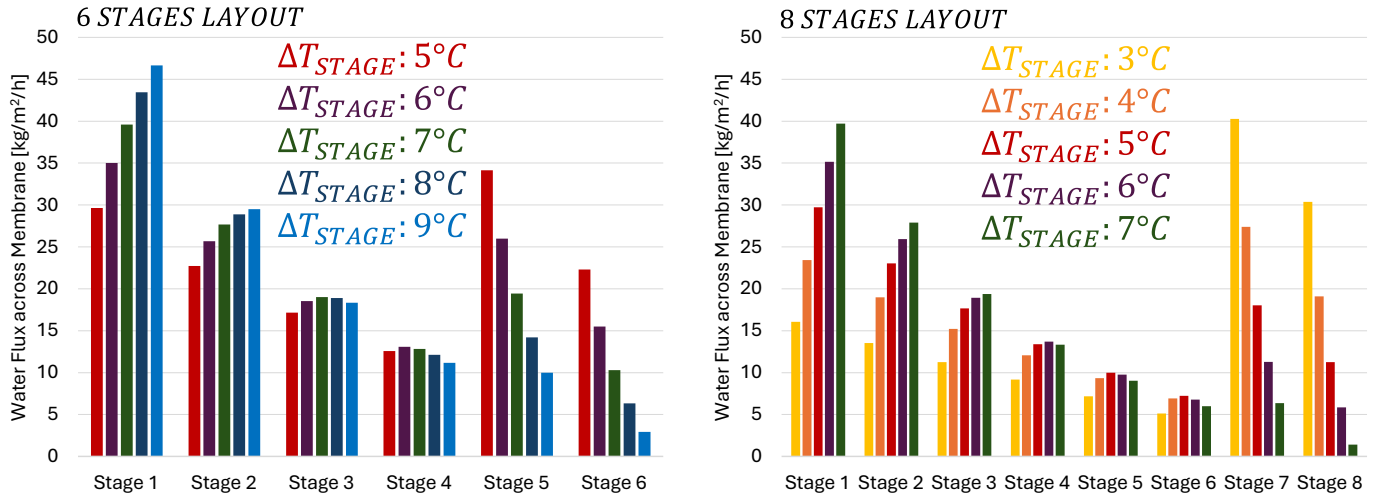


Fig. 13. Average water flux across each stage of the simulated plants.

each stage (blue HX in Fig. 9 and Fig. 10) is defined, the pinch point temperature difference in the condenser was set at 1°C . Across the simulations, recirculation streams mixing was assumed to be isothermobaric (with $T_1 = T_{1R}, \dots, T_N = T_{NR}$), even if the salinity of the mixing flow is different due to water separation at each stage (i.e: $x_1 < x_{1R}, \dots, x_N < x_{NR}$). Therefore, a sudden salinity increment is exhibited at each mixing point, where the recirculated streams had a higher mass

flow rate than the feed stream of the respective stage. The overview of both input and output of VMD plant is reported in Table 4.

Along with the water flux J_w , the results also include the STEC and GOR of the plant, defined as:

$$\text{STEC} = \frac{Q_{in}}{V_{\text{freshwater}}} \quad (20)$$

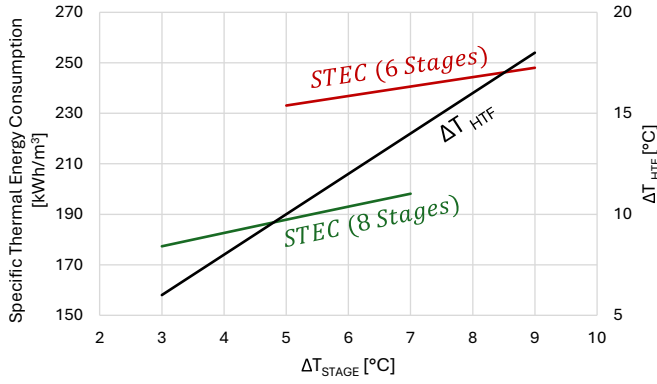


Fig. 14. STEC and ΔT_{HTF} (i.e. heat source temperature difference) vs. ΔT_{STAGE} .

where,

$$Q_{in} = m_{1R} \cdot C_{p,1R} \cdot (T_1 - T_2) + m_{2R} \cdot C_{p,2R} \cdot (T_2 - T_3) \quad (21)$$

and

$$V_{freshwater} = \frac{m_{1P} + m_{2P} + m_{3P} + \dots + m_{lastP}}{\rho_{water}} \quad (22)$$

and

$$GOR = \frac{V_{freshwater} \cdot \rho_{water} \cdot \Delta h_{eva}}{Q_{in}} \quad (23)$$

5.2. Technical analysis of VMD plant layouts

As indicated, sensitivity analyses on the ΔT_{STAGE} are carried out to understand the performance and characteristics of the VMD plant layout with 3 cascades and 4 cascades of effects. Fig. 11 and Fig. 12 summarize the results of the detailed 1-D simulations for one simulation case. Namely, the 6 stages plant layout for a ΔT_{STAGE} of 7 °C and with 14 °C of temperature difference on the heat source. The axial membrane coordinate in Fig. 11 and Fig. 12 correspond to the stages, each containing 10 membrane modules connected in series. Based on the required membrane area per stage, multiple membranes may be placed within each module.

As visible in Fig. 11, the temperature of the permeate (i.e. the temperature at the wall of the feed) is close to the bulk temperature of the feed (with temperature differences between 0.45 and 0.2 °C), and the temperature polarization effects are not so relevant. The saturation temperature of the vacuum, where pure water permeates, is decreasing in the first 4 stages (first 2 cascades), while it is capped at the minimum value (25 °C), in the last two stages, since no additional recovery is

foreseen. The permeate flux, J_W , is decreasing as the temperature driving force decreases along each stage, decreasing also across different stages, penalized at low feed temperatures. The water flux increases in the last two stages, as they are characterized by the minimum admissible vacuum degree, increasing the driving force. In this example, the STEC is 240.6 kWh/m³, and the recirculation ratio in the first two stages is 9.55. The heat source was assumed to be always available at 90 °C, thus heating the feed of the first stage to 87 °C. The STEC was evaluated by taking into account the thermal consumption of heating up the feed from 20 °C to this temperature. The temperature of the brine at the outlet of the present VMD plant configuration (at 20 % salinity) is 45 °C (Fig. 11). This temperature varies between 57 °C (with 6 stages and $\Delta T_{STAGE} = 5$ °C) to 31 °C (with 8 stages and $\Delta T_{STAGE} = 7$ °C) based on the number of stages and the assumed ΔT_{STAGE} .

To justify the assumption of negligible conductive losses across the membrane, heat transfer by conduction was estimated based on the material's thermal conductivity (0.9–1.0 W/m/K) and the membrane porosity [46]. Flux of the convective heat from the bulk feed to the wall temperature was about 40 kW/m². This was computed based on the convective heat transfer coefficients on the feed side, which ranges from 98,000 to 110,000 W/m²/K across all the stages (modelled as in Appendix B), and the average bulk-to-wall temperature difference of approximately 0.4 °C across all stages. Therefore, by computing a membrane thermal resistance of about 0.001 m²·K/W, conductive losses below 2 % (i.e., thermal efficiency >98 %) would be due to a trans-membrane temperature difference of less than 1 °C, a condition compatible with the observed small convective temperature gradient.

The axial trend of the salinity increases along the stages' axial direction, and from stage to stage (Fig. 12). That is due to the high recirculation ratio, up to 20 % at the VMD plant outlet. The driving force ΔP^* is graphically visualized by presenting the feed pressure, the pressure $P_{interface,1}$ at first interface layer of the membrane (top layer), the pressure $P_{interface,2}$ at second interface layer of the membrane (intermediate layer), and the vacuum pressure (across the support layer).

Fig. 13 shows the average water flux J_W across each stage of the plant for the plant layout with 6 stages (left) and 8 stages (right), where estimated fluxes are always higher in the higher-temperature stages (first stages). In particular, assuming a large ΔT_{STAGE} increases the average driving force across each stage. The driving force is boosted as the J_W increases drastically from the third-to-last stage to the second-to-last stage. That is due to the lower vacuum temperature imposed in the second-to-last stage (25 °C). The average J_W of the whole plant is relatively constant between the various configurations. That is because, when high ΔT_{STAGE} values were assumed, the higher J_W of the first stages is counterbalanced with the lower J_W of the last stages, and vice versa if lower ΔT_{STAGE} are considered.

The results in Fig. 14 depict the trend of the specific thermal consumption of the two plant layouts, depending on the number of stages

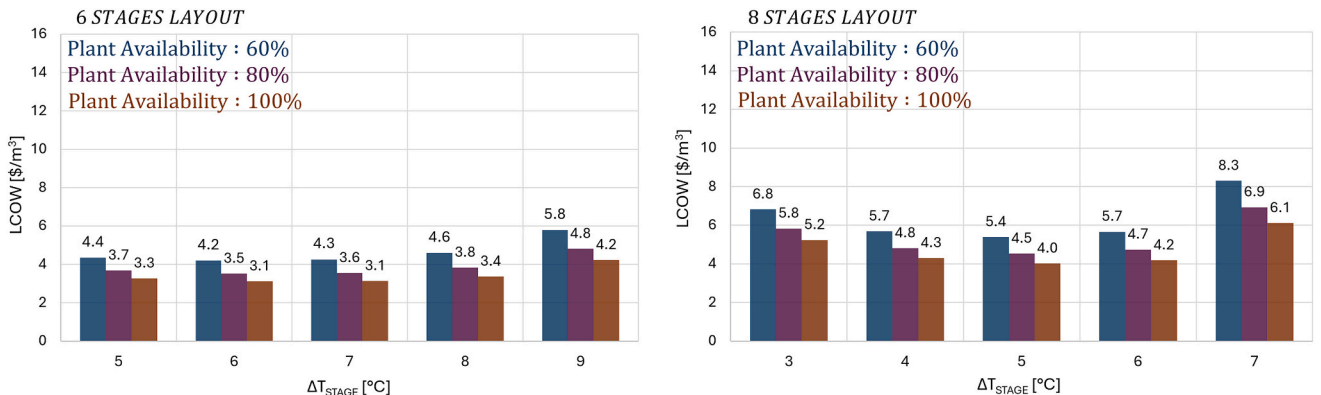


Fig. 15. Estimated LCOW of the plants with a feed flow rate of 3.6 m³/h, for different ΔT_{STAGE} . Results are shown for the percentage of annual plant availability.

and the ΔT_{STAGE} , including the foreseen temperature difference across the heat source. When sensible heat source exploitation is essential (up to 18 °C, in this work), it is noticeable that 6 stage layouts are likely to be adopted, with STEC in the order of 240 kWh_{th}/m³. For more isothermal (latent-like) heat sources, in contrast, VMD layouts with 8 stages in cascade are more energy efficient (or possibly even 10, a configuration not investigated in this work), with STEC in the range of 180 kWh_{th}/m³. In comparison with commercial thermally driven systems, the membrane technology proposed in this work aims at achieving desalination close to ZLD, which comes at a cost of a STEC higher than the one of mature technological solutions suitable for lower RR. For instance, assuming the same heat source of this work, a MED plant theoretically achieves STEC in the range of 80–110 kWh_{th}/m³, depending on the number of stages (from 6 to 8). However, in practice MED systems cannot allow for RR exceeding 50 % due to the detrimental effects of high salinity flows to the heat exchangers involved in the process. STEC values of MD plants are dependent on how the heat input is recuperated and recirculated in cascade between the stages: considering that the evaporation enthalpy of water at 50–80 °C is in the range between 650 and 660 kWh_{th}/m³, this value can be considered as the STEC for the single-stage VMD plant without any internal heat recovery.

In addition to the STEC, also the values of the GOR of the plant are reported. For the plant layout with 6 stages, the GOR ranges linearly from 3.4 (ΔT_{STAGE} of 5 °C) to 3.8 (ΔT_{STAGE} of 9 °C), whereas for the plant layout with 8 stages, the GOR varies linearly, analogously, from 4.6 (ΔT_{STAGE} of 3 °C) to 5.4 (ΔT_{STAGE} of 7 °C).

Accordingly, the criteria for selecting the most suitable configuration does not only consider the thermal analysis and the heat source characteristics but must be an outcome of a techno-economic analysis. On this regard, the next sub-section describes a preliminary economic analysis conducted for the various plant layouts proposed in this chapter, with the aim to minimize the LCOW of the VMD plant.

5.3. Economic analysis of VMD plant layouts

In this work, the LCOW is proposed as a key performance indicator to assess the effect of varying the plant layout configuration and annual availability. That is to account for potential fluctuations in the waste heat availability that may impact the plant annual production capacity and economic performance, thus adding depth and improving the validity of our analysis for practical applications. Therefore, capital costs and annual operating and maintenance costs were estimated for each of the two plant layouts presented in this study. Capital costs were evaluated for the major plant equipment and other sub-cost elements. The major plant equipment were the membranes and their modules, water pumps and heat exchangers. These were expected to have a major contribution to the capital costs and were acquired through either personal communication with suppliers or using the capacity method that accounts for the economy of scale with respect to a base case [47]. The other sub-costs included the piping, instrumentation, installation, indirect costs and contingencies, estimated as a percentage of the capital cost. The main annual operating and maintenance costs considered were the energy and membranes replacement costs which were also a function of the plant annual availability.

This section highlights the estimated LCOW for the two plant layouts shown in Fig. 9 and Fig. 10, for annual plant availability of 60 %, 80 % and 100 %. Specifically, the case study of this work presents techno-economic results of the plant (i.e. LCOW) by assuming a 3.6 m³/h feed flowrate and a freshwater production capacity of around 3 m³/h. The cost of the tubular α AO membranes was estimated at 1000 \$/m². The capital costs, annual operating and maintenance costs and LCOW were calculated following the approach detailed on Appendix C.

For the 6 stages layout, the LCOW followed an increasing trend with the ΔT_{STAGE} (Fig. 15). That is due to the lower water flux of the last two stages for the high ΔT_{STAGE} layouts (Fig. 13a), which required a larger membrane area and hence a higher capital cost. While the 8 stages

layout exhibited a lower specific thermal energy consumption, estimated LCOW was almost twice that of the 6 stages layouts over the viable range of ΔT_{STAGE} values. Thus, showing a trade-off between the plant energy consumption (STEC) and its corresponding LCOW. However, the LCOW did not increase linearly with the ΔT_{STAGE} and an optimum ΔT_{STAGE} value at 4 °C is found. That is due to its higher overall water flux of the plant layout, hence requiring a lower membrane area and capital costs. Overall, the plant availability (spanned between 60 % and 100 %) has an important but not drastic effect on the LCOW. It is important to note that the intermittence of the waste heat source would affect the membrane permeate flux, reflected as reduced water production and lower annual plant availability. Herein, the plant annual availability reflects the effect of intermittence of the waste heat source on the LCOW and economic performance of the plant. While the membranes' integrity remains unaffected during downtime, i.e. if the membrane dried out [40], a stable waste heat source is economically advantageous.

Most importantly, the resulting numerical values of LCOW reported in this study is shown as a key performance indicator (along with STEC) to evaluate the feasibility of the plant layouts and the use of ceramic membranes, without the goal of setting a benchmark price for VMD. As a matter of fact, analysis of LCOW values reported in literature for VMD plants can be found in an extremely wide range, from 0.6 to 18 \$/m³, as the case with other MD configurations [48]. The large variation in the water production costs is related to differences in the membrane materials, plant capacity, feed salinity, target recovery ratio and energy source considered in the respective studies. Furthermore, it is also important to point out that economic analyses in literature are often based on unique assumptions and application-based case scenarios, which often hinder legitimate comparison and benchmarking.

6. Conclusions

This study focused on VMD plants with near-zero liquid discharge for seawater desalination, with brines close to the crystallization point. The developed model was successfully validated with experimental water flux values of symmetric membranes of different materials. Simulation of innovative asymmetric tubular α AO membranes was enabled by numerically fitting the membrane parameters (mainly the tortuosity of each layer) to match the experimental vapor flux of the same membranes.

The techno-economic analysis of plant layouts based on the modelled asymmetric tubular α AO membranes, demonstrated that the exploitation of a sensible heat source available at 90 °C is effectively possible with a series of effects in cascade, achieving STEC in the range between 180 and 250 kWh/m³. Finally, the annual economic analysis revealed a reasonable LCOW range between 3 and 8 \$/m³, based on conventional assumptions regarding capital and operational costs. This analysis also considered a low-cost ceramic membrane, representative of a future scenario with high commercialization levels. The economic results are encouraging, especially given the very high recovery ratio (above 85 %) achieved by the simulated VMD plant—a recovery ratio level not attained by other thermal desalination technologies.

CRedit authorship contribution statement

Nadin Al-Jariry: Writing – review & editing, Writing – original draft, Visualization, Validation, Investigation, Conceptualization. **Ettore Morosini:** Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Formal analysis. **Frank Lipnizki:** Writing – review & editing, Conceptualization. **Igor Matteo Carraretto:** Writing – review & editing, Writing – original draft, Software, Methodology, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A

In this work the driving force of the VMD process is the pressure difference between the saturation pressure of pure water at the wall conditions (where the evaporation firstly occur) multiplied by a correction factor (the product between the molar fraction of water in the feed and the activity coefficient of water in the feed) and the vacuum pressure applied on the other side of the membrane by the vacuum pump:

$$\Delta P^* = P_{sat} (@T_{wall}^{feed}) x_{H2O} \gamma_{H2O} - P_{vacuum}^{permeate} \quad (A.1)$$

According to the literature model of Zhang [27], the saturation pressure of the water is considered without any correction terms related to its salt concentration, whereas subsequent works included the penalizing effects of the salt content in the feed to the driving force. For example, a literature work of Chung [29] proposed to introduce the activity coefficient of water in the feed, γ_{H2O} , to correct the saturation pressure of water.

Subsequently, a literature work of Li [17] introduced both the molar fraction of water and its activity coefficient as correction factors, that was modelled as:

$$\gamma_{H2O} = 1 - 0.5 \cdot (1 - x_{H2O}) - 10 \cdot (1 - x_{H2O})^2 \quad (A.2)$$

Other studies of Alawad [49,50] assumed the same correction terms of Li (i.e: $x_{H2O} \cdot \gamma_{H2O}$), as assumed in this work in Eq. (12).

Finally, literature works of Triki [33] or Asiri [51] adopted a similar correction factor $1/A^*$, where A^* is expressed as:

$$A^* = 1 + 0.57257 \cdot \left(\frac{y_{NaCl}}{1 - y_{NaCl}} \right) \quad (A.3)$$

and it has validity from 0 to 200 gNaCl/kg [36], to correct the vapor pressure of water at the temperature T_{wall}^{feed} , being y_{NaCl} is the mass fraction of salt in the feed.

The comparison between the three different models shown, adopted to introduce a correction factor to the VMD driving force ΔP^* to account for the feed salinity, is graphically shown in Fig. A1: as visible, the model adopted in this work from Li and Alawad (considering $x_{H2O} \cdot \gamma_{H2O}$) is coherent with the one of Triki ($1/A^*$) for a wide range of water mass fraction in the feed (from 100 % to 85 %), while the models of Chung (γ_{H2O}) and the sole adoption of the water molar fraction (x_{H2O}) both have a more limited effect on the reduction of the driving force ΔP^* .

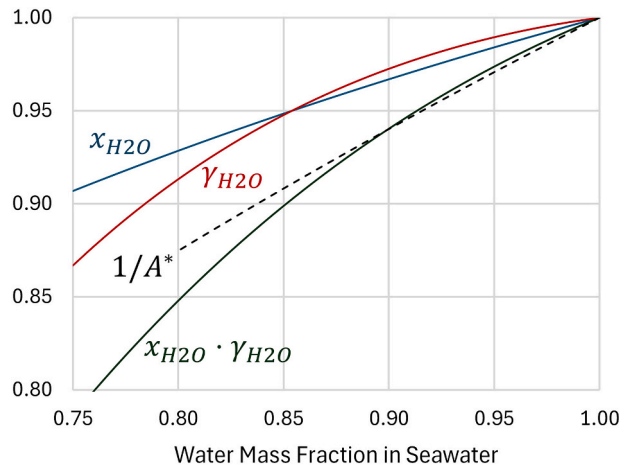


Fig. A1. Seawater salinity effect on the driving force.

Appendix B

The correlations for the convective heat transfer coefficient of the feed in contact with the membrane are reported in this section.

In the case of internal flow on the flat sheet circular membrane, adopted in Sections 2.3, 4.1 and 4.2 for the comparison between the model and the experimental data collected, the correlations for the Nusselt number are for a flat plate:

$$Nu = 0.023 Re^{0.8} Pr^{0.3} \quad (B.4)$$

$$\alpha = Nu \cdot \frac{k_{feed}}{L_c} \quad (B.5)$$

In the case of internal flow in the channel of the tubular membrane, used in Section 5 for the simulations of the full-scale VMD plant, the correlations for Nusselt are for a channel, as shown in Eq. (B.6), where $d_h[m]$ is the hydraulic diameter of the channel.

$$Nu = \frac{(f/8)(Re_{dh} - 1000)Pr}{1 + 12.7(f/8)^{1/2}(Pr^{2/3} - 1)} \text{ and } \frac{1}{\sqrt{f}} = (1.82 \log Re_{dh} - 1.64) \quad (B.6)$$

for $3000 \leq Re_{dh} \leq 5 \cdot 10^6$

$$\alpha = Nu \cdot \frac{k_{feed}}{d_h} \quad (B.7)$$

Appendix C

C.1. Capital costs

- Membranes and membrane modules

The plant simulation was based on tubular α AO membranes, whose properties are summarized in Table 3. Their cost was estimated at 1000 \$/m², based on consideration of an additional production step that adds 500 \$/m² to the current market price of microfiltration membranes that are in the range of 400–500 \$/m². While the membranes are not available for a large-production scale yet, their cost is expected to be reduced upon wide adoption in the future.

Considering the low diameter to length ratio of the simulated tubular membranes and high salinity of processed water, the membrane modules must offer high flexural strength and resistance to corrosion. In this study, the modules' capital cost estimation was based on polymeric producible and tested MD modules as the reference. The capacity method was used, and the reference capacity was the membrane surface (m²) [52]:

$$C = n \times C_{ref} \left(\frac{Q}{n \times Q_{ref}} \right)^f \quad (C.8)$$

where C_{ref} and Q_{ref} are the cost (76,193 \$) and capacity (300 m²) of the reference module, respectively, and f is the scaling factor (0.8) that is also known as degression coefficient.

- Heat Exchangers

The plant layouts shown include heat exchangers for heating the feed stream with heat supplied by a waste heat source and condensers were used for recovering the latent heat from the vapor streams. The number of heat exchangers varied depending on the plant layout in consideration, where 8 and 10 heat exchangers were designated for the 3 and 4 cascade layouts, respectively. The cost of heat exchangers was taken as 157 \$/m² [53], with the heat exchange area (A) is estimated as:

$$A = \frac{Q_{exchanger}}{U \Delta T} \quad (C.9)$$

where $Q_{exchanger}$ is the heat duty (W) of each heat exchanger, U is the overall heat transfer coefficient (1000 W/m²·K) [54] and ΔT is the mean logarithmic temperature difference along the heat exchanger.

- Pumps

Cost of water circulation pumps was also estimated according to the capacity method with respect to available data, where the reference capacity was specified as the water volumetric flow. The reference pump cost (\$17,841), capacity (250 m³/h), and the scaling factor (0.14) were adopted from literature [55,56]. The number of water pumps varied depending on the plant layout in consideration, where 12 and 16 water pumps were designated for the 3 and 4 cascade layouts, respectively.

- Other capital costs

Apart from the major plant equipment, capital costs also include sub cost elements such as costs for piping, sensors, electrical subsystems, installation etc. Specific costs for such elements are difficult to obtain and are commonly assumed as a fraction of the major plant equipment capital costs, which facilitates their cost estimation as a function of the number of modules and their interconnections. Considering the relatively high cost of ceramic membranes, the sub-costs were assumed to be 10 % of the major plant equipment's capital costs.

C.2. Annual operating and maintenance costs

In this study, the operating and maintenance costs accounted for the membrane replacement cost, the plant maintenance cost, water pretreatment chemicals and the electric energy cost of water pumps. The assumed membrane life is 10 years, which corresponds to an annual membrane

replacement factor of 10 % the membranes capital cost [57], starting from the first year of operation. The annual plant maintenance cost was considered as 0.5 % of the total capital costs [24]. The chemicals cost for the water pretreatment was taken as 0.018 \$/m³ of feed water [58]. The electric energy cost of the water circulation pumps was estimated for a pump efficiency of 0.8 [59], a pressure head of 0.7 bar per stage [58], and a unit price of electricity as 0.1 \$/kWh [52].

C.3. Levelized cost of water (LCOW)

As all the capital and operative cost are introduced, the work showcases the LCOW as the sole economic indicator of the full-scale plant. It was estimated based on the amortized capital and annual operating costs, considering different operating conditions such as the plant availability and the waste heat cost. The capital recovery factor (CRF) is assumed at 8 %; for an annual discount rate of 5 % and an amortization period of 20 years. The LCOW was evaluated for each of the plant layouts and the variable operating conditions according to:

$$LCOW \left[\frac{\$}{m^3} \right] = \frac{\sum CAPEX \bullet CRF + \sum OPEX}{W_p \bullet t_{eff}} \quad (C.10)$$

where W_p is the annual production capacity of the plant (m³/year) and t_{eff} is the percentage of annual plant availability assumed to be at 60 %, 80 % or 100 %.

Appendix D. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.desal.2025.119627>.

Data availability

Data will be made available on request.

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