

Membrane distillation - producing high quality water from saline streams by deploying waste heat

A.E. Jansen*, J.W. Assink, J.H. Hanemaaijer and J. van Medevoort

Keywords: membrane technology, hybrid, distillation, desalination, seawater, dewatering, brine, mass transfer, waste heat, ultrapure water

Abstract

Membrane distillation has been developed to stage that commercial applications will become feasible in the near future, aiming at fresh water extraction from seawater. Pilots tests and bench scale tests over thousand hours, using various configurations of membranes, spacers and condenser materials, have shown the potential of membrane distillation. Pilots have demonstrated the excellent product water quality, the need for little water pretreatment and a thermal energy requirement of approx. 520 MJ/m³ water. Evaluations for large scale applications, using inexpensive waste heat, are indicating an energy requirement of 300-400 MJ/m³ and costs between 0.30 and 0.50 dollar per m³. Future development work will focus on further reduction of the energy requirements, allowing new applications.

Introduction

The need for fresh water is connected to all land-based biological organisms, including mankind and its food chain. Industrial processes and especially the population in megacities have developed very rapidly in the last five decades, leading to overexploitation of groundwater and nearby rivers systems. Also the climate change and the ever growing agricultural production of food and other types of biomass has given raise to the concern of shortage of available freshwater to mankind and, as a result, natural biosystems.

Consequently, fresh water produced from seawater has become an increasingly important source. Just over 1 % of the drinking and process water produced worldwide has been extracted from seawater and brackish sources. The total desalination capacity is 40 million m³/day in 2006. However, an annual growth of approx. 10% in the production capacity is expected in the period 2005-2015 and larger growth rates are expected in the years after 2015 (Media Analytics Limited, 2006). State-of-the-art techniques for water extraction (usually called desalination techniques) are multistage flashing (MSF), multi-effect distillation (MED) and reverse osmosis (RO). In addition, electro dialysis (ED) as ion removal process may be applied to brackish water sources. The main driving force for these techniques is heat (in MSF or MED) or electricity (for creating a high pressure difference in RO processes or polarity for ionic transport in ED). Reverse osmosis has become a very competitive alternative since the end of the 1990's, because the energy requirements of RO were strongly reduced. Nowadays values around 3.1 kWh per m³ water produced are not uncommon and even values below 2 kWh per m³ are reported, because of improved energy recovery concepts (Fritzmann et al 2007). In addition, the costs of commercial RO module systems have been drastically reduced by standardization and the high market volumes. Most desalination (water extraction) projects nowadays follow the RO concept, allowing water prices of roughly 0.50-0.60 US dollar per m³ produced in large scale plants.

Many new variants have been added to the aforementioned state-of-the-art technologies, but most of them have not shown to be competitive in (large scale) water extraction applications up to now. An exception of high practical interest is membrane distillation (MD), which is now on the brink of commercialization.

Membrane distillation has been recognized an alternative for water extraction for already a long time, but insufficient performance, related to costs for membranes and high energy consumption, has been a show stopper several times.



First publications on MD date back to the sixties of the last century (Findley, 1967). MD uses a membrane with gas filled pores, in contrast to usual membrane techniques for water treatment, such as ultrafiltration and RO. Water vapor is transported through the MD membrane, using a water pressure difference -i.e. a temperature difference- as the main driving force. The water vapor pressure should be taken at the evaporating and condensing surfaces. A correction for the effect of dissolved salts on the water pressure is needed at the evaporating side of the membrane.

Major advantages of a MD membrane are the short travel distance between the evaporating surface and the condensing surface, allowing very compact installations, and a full segregation of the feed stream and the product stream, which makes a very good salt retention possible.

MD technology holds a number of other advantages, potentially making it an important alternative for state-of-the-art techniques for seawater desalination. MD may use low top temperatures, making it suitable for using waste heat or solar heat. MD essentially uses no additives to prevent (bio)fouling of the membrane in the MD module. MD produces only up to approx. 10% of the feed stream as distillate. The environmental impact of the retentate discharge will therefore in most cases be low or neglectable.

Theoretical backgrounds

MD has developed into different configurations. Lawson et al (1997), Guijt (2002), Alklaibi et al (2004) and others distinguish the following major configurations, as developed and researched until the start of 21st century:

- a) Direct contact MD. Both fluids contact the membrane. The product stream circulates over an heat exchanger to remove the condensing heat of produced water and to maintain a driving force.
- b) Air gap MD. The air gap is used to reduce the 'leakage' of heat by conduction through the membrane; the conductive leakage has a negative effect on the energy efficiency of the process. Disadvantage of this configuration is the additional resistance of both the air gap and the layer of condensing water to the transport of water vapor, leading to low fluxes (i.e. the production rate per m² of membrane).
- c) Sweep gas MD. The produced water vapor is transported to an external heat exchanger, where the water is condensed, the sweep gas is usually recycled to the MD unit.
- d) Vacuum MD. The resistance of the air gap is strongly reduced by applying a vacuum. The produced water vapor is led to a condensing surface, usually downstream of the vacuum pump.

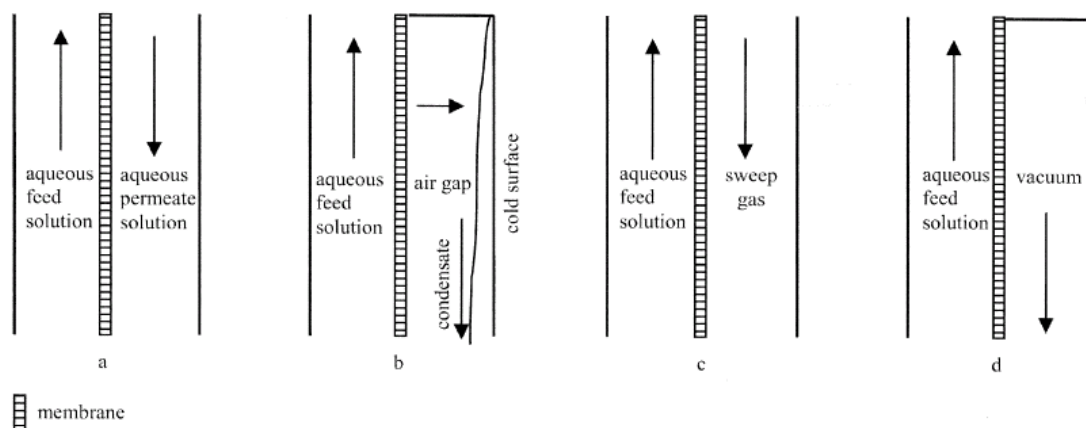


Figure 1 Various MD configurations: a) direct contact MD, b) air gap MD, c) sweep gas MD and d) vacuum MD. Source: Meindersma et al (2006)

A more detailed survey of the work on these concepts can be found in the aforementioned publications, Lawson et al (1997), Guyt (2002) and Alklaibi et al (2004), and others.

The MD concept described in this publication is new in that it combines direct contact MD with an internally placed heat exchanger to recover virtually all evaporation heat. Originally, the new process was based on air gap MD and offered an extra large condensing surface (Hanemaaijer, Van Heuven, 2000). The saline feed water was led twice through the module. The first time through a set of small pipes that forms the condenser. The water flow in the tubes is preheated by condensation of the product water at the outside surface. The second time, the preheated water flows countercurrently through a set of membrane fibers. Waste heat, or another heat source of choice, will give the water a temperature rise of 2-10 kelvin at the high temperature end of the module (i.e. the top temperature) before the water is returned. From this point the water is called retentate, in analogy with other membrane processes. The temperature difference between the two streams is sufficient to maintain a driving force for evaporation throughout the whole length of the module. The evaporated water is condensed at the small condenser pipes with relative cold feed water. Separation of module in many temperature segments helps to form an near ideal countercurrent process, resulting in important energy savings. However, this original concept proved to be difficult to scale up to full-scale installations, after which it was decided to switch to a concept with essentially no air gap. The second concept avoids the need for building strong and rigid structures to hold all module items (pipes and fibers) in place. Additionally, the second concept uses flat membranes and condensing surfaces, again avoiding the problems with flexible membrane fibers in the original concept. Hanemaaijer et al (2006a) gives more details. The so-called Memstill® process is based on this latest concept and has recently been tested in pilots under real-world conditions.

Heat transfer and mass transfer are coupled in MD processes. As water evaporates at the interface with the membrane, the temperature sinks at this point. The water vapor condenses at the other side of the membrane, and the heat of condensation will be transported to the relatively cold feed water. As a result a temperature profile will develop (figure 1). Mass and heat transfer relations can be developed for MD and solved in an analytical or numerical way.

The aforementioned maximum in the production rate of MD is based on the heat balance. The sensible heat content of heated water, which essentially drives the evaporation process, is maximally 15% of the heat of evaporation of the same mass of water.

Mass transfer relations are based on differences in partial pressure of water across the membrane:

$$J = K_m \cdot \Delta P \quad [1]$$

K_m is related to the membrane characteristics and includes transport by convection and diffusion, the latter being composed of Knudsen diffusion and molecular diffusion. It is obvious that the mass transfer of water vapor will be hindered by the presence of inert gasses in the membrane, for instance air. Degassing of the feed stream may therefore be beneficial to the performance of MD. Potentially, the degassing process could be integrated in the MD process at the low temperature side of the MD module, by increasing the total membrane surface by 2-10%. The ΔP has to be corrected for the actual salt content near the evaporating surface, i.e. the membrane¹. No correction was made for potential salt polarization near the membrane (Chernyshov et al, 2003), in contrast to the temperature polarisation.

The heat transfer across the membrane is based on the temperature difference, which is actually needed to create a partial water pressure difference:

$$Q = (K_h / \delta) \Delta T_m \quad [2]$$

Where K_h is the sum of the thermal gas conductivity and the solid conductivity in the membrane ($K = \epsilon K_g + (1-\epsilon) K_s$, with ϵ the porosity of the membrane), δ the membrane thickness and ΔT_m the temperature difference across the membrane.

¹ A thermodynamic model of OLI Systems inc. was used to estimate the water pressure of saline solutions.

The mass and temperature relations will not be described here in further detail. Reference can be made to literature, for instance Chernyshov and Guijt, who present numerical and analytical solutions. Some interesting phenomena can be noted, however, in relation to temperature polarization and the temperature difference between the hot and cold stream.

A higher flux will have a strong impact on the temperature polarization θ , which is defined by the ratio of temperature difference between the evaporation and condensing surface compared to that of the bulk temperature difference:

$$\theta = \frac{T_{hm} - T_{pm}}{T_h - T_p} \quad [3]$$

Here, T_h is the temperature of the relative hot stream (the retentate) and T_p is the temperature of the product water. Schofield (1987) stated a range of 0.4 to 0.7 for θ in well designed systems. Very low values of θ should be avoided, which can be achieved by increasing the liquid velocity in the channels or by using heat transfer promoting spacers. Higher liquid velocities however will increase the pressure drop along the liquid channel.

A higher temperature difference between the feed stream and the retentate will result in a higher water flux, leading to a smaller required membrane area at a given production capacity. But a higher temperature difference also means a higher energy input at the top side of the module. In this way, the energy consumption and the required membrane area of the process are partly exchangeable for a given module concept.

The performance of MD is often expressed as specific flux, which is related to the vapor pressure of water:

$$J_s = \frac{dM}{A \cdot dt} \cdot \frac{1}{\Delta P_{in}} \quad [4]$$

A is here the total membrane area (m^2), i.e. it is not corrected for areas that do not contribute to the water transport; dM/dt is the produced amount of water (kg/s) and ΔP_{in} is the average water vapor difference along the membrane. For this, the logarithmic average is used:

$$\Delta P_{in} = \frac{(\Delta P_2 - \Delta P_1)}{\ln(\Delta P_2 / \Delta P_1)} \quad [5],$$

based on T_{in} and T_{out} values in the mixed water flows just outside the MD module to calculate the average water vapor pressures with an Antoine relation. ΔP_2 is calculated at the high temperature end of the module and ΔP_1 at the low temperature side of the module.

It should be noted that the flux will exponentially increase with increased temperature, but the specific flux will decline, due to temperature polarization.

The MD module itself has several points for optimization, especially in the case of the Memstill® concept:

- type of membrane (thickness, type of material, porosity, tortuosity)
- type of condenser material (thickness, specific heat of conductivity)
- type of spacer (assisting the heat transfer and mass transfer in both liquid filled channels)
- the hydraulic diameter of the channel, which is related to the type of spacer and construction technique (determines the pressure drop along the flow channel and heat transfer)
- a thin layer of product water, and heat recovery upon discharge of the product

Memstill® is based on commercially available materials, in order to avoid high development costs. Optimization of the MD process should be focused on maximal transfer of sensible heat in the liquid filled parts of the MD module and the condenser (i.e. a series of low heat resistances from the bulk of the hot liquid flow to the surface where the water evaporates and similarly the series of heat resistances to the heat sink, the cold feed stream), but in the same time a maximal blocking of sensible heat in the membrane itself, where heat is preferentially transported as water vapor. For both high energy efficiency in MD and high vapor fluxes, the membrane should have a high ϵ/τ ratio, ϵ is the porosity of the membrane and τ the tortuosity of the pore system.

A high ϵ value ensures a low leakage of sensible heat via the solid mass of the membrane (heat losses by conduction), but also a high ϵ/τ value will give a low resistance to the vapor flow through the membrane. The membrane thickness should be optimized because thicker membranes will result in lower heat losses via the membrane, but also in lower water vapor transport. Both processes are proportional to the membrane thickness, as demonstrated by Bandini et al (1991) for pure water, and hence, related to the intrinsically properties of the membrane. However, the major resistance to mass and heat transport may also be outside the membrane, as one will easily see when very thin membranes were chosen and the resistance of the membrane will be virtually zero.

As one can also easily understand, the layer of product water in the Memstill® process should be as thin as possible. The heat resistance of this layer will become minimal by applying the right pressures on all liquid channels. The applied pressures resulted in a 'milking effect', as shown in many bench-scale and pilot tests: the product water leaves the module every 2-5 seconds in small volumes. It is assumed that the product water channel has reached virtually zero thickness by this approach. Heat losses in MD are related to the total configuration of the module of which a part is shown in figure 2.

The overall heat balance of the process ideally contains the following factors: a) the enthalpy of the feed stream, b) the enthalpy of the retentate, c) the enthalpy of the product stream and d) the enthalpy added at the high temperature side of the module (preferentially by using waste heat). Inside the module heat is mainly transported as latent heat from the retentate to the product stream. In addition, a part of the driving force of the MD process will be lost due to the internal and external heat conduction processes.

Total heat transfer includes three routes:

a) heat transfer by conduction (sensible or perceptible heat) from the warm retentate to the cold channel with feed water and the product stream, in a rectangular channel:

$$Q_c = nW \int_0^L q_{m,x} dx = W \int_0^L k_m (T_{h,x} - T_{p,x}) dx \quad [6]$$

(k_m = heat conductivity of the membrane, W = width of the channel, n = number of membranes in the module, $(T_{h,x} - T_{p,x})$ = temperature difference between retentate and product stream at point x , and L is the length of the module; see also figure 2)

b) heat losses Q_{loss} to the surroundings:

$$Q_{loss} = A_{ext} \overline{k_{case}} (\overline{T_c} - T_{env}) \quad [7],$$

(A_{ext} is the external surface of the module, k_{case} the heat conductivity of the module case and T_{env} the external temperature) and

c) heat transfer by water vapor transport (Q_l latent heat)

$$Q_l = \Delta H_v \overset{\circ}{m}_p \quad [8],$$

with ΔH_v = heat of evaporation and m_p = mass flow of product water.

The relative influence of Q_{loss} (relation 7) on the total heat flow will become very small and may be neglected, especially by sufficient isolation and by scaling up the modules.

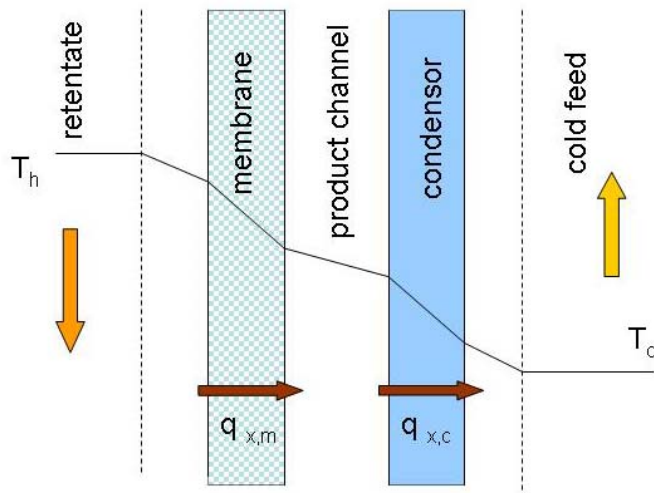


Figure 2: The MD module, according to the 'Memstill®' configuration. The dotted vertical lines indicate the border layers of the feed stream and retentate stream near the condenser and the membrane. The condenser wall is impermeable for gasses and liquids. Vertical arrows indicate the flow directions in both liquid channels. Spacers in these channels are not shown. The temperature profile and the local heat fluxes q in the membrane and condenser are additionally shown.

The MD membrane should fully reject salts from the saline feed. Hence, the membrane should not be wetted by tensides in the feed and the MD membrane should be free of large pores ('pin holes'). The pressure difference across the membrane should be preferentially below 1 bar to avoid 'leakage' through these large pores.

Tests of Drioli et al (1986) showed that $0.2 \mu\text{m}$ nominal pore sizes or smaller are required for obtaining full rejection of salt or glucose. This is related to the Laplace equation:

$$\Delta P = \frac{4\gamma}{d} \cos \varphi \quad [9]$$

The rejection of saline solutions is adversely influenced by high pressure difference across the membrane (ΔP), a larger pore size (d) and a small surface tension of the solute (γ); the $\cos \varphi$ (the wetting angle of the membrane material) should preferentially be highly negative. Previously tested membranes have nominal pore sizes between 0.1 and $1 \mu\text{m}$ (Lawson, Alklaibi).

Fane et al found high specific fluxes already in 1987: $J_s = 3.4 \cdot 10^{-7} \text{ kg}/(\text{m}^2 \cdot \text{s} \cdot \text{Pa})$. Coatings are used to prevent the (partial) penetration of pores by water, which would be detrimental to the high flux.

Silicon-fluoropolymers coated fibers were for instance investigated by Sirkar and Li (2003). The highest fluxes with these proprietary membranes were $70 \text{ kg}/\text{m}^2 \cdot \text{h}$ in the vacuum mode and $54 \text{ kg}/(\text{m}^2 \cdot \text{h})$ in the direct contact membrane distillation (DCMD) mode, both obtained with $85 \text{ }^\circ\text{C}$ brine temperature, a temperature difference across the membrane of almost $70 \text{ }^\circ\text{C}$ and 1% NaCl brine. These values indicate a specific flux of approx. $3 \cdot 10^{-7} \text{ kg}/(\text{m}^2 \cdot \text{s} \cdot \text{Pa})$, which is measured at high liquid velocities and is completely attributed to a transfer resistance at the membrane side of the process. The coating of the membranes clearly creates an extra resistance to the transport of water through the membrane. Incorporating the transfer resistance at the condensing side and/or lower velocity speeds would reduce this high specific flux value. The work presented here will show high values, close to the values presented by Fane et al in 1987, despite the extra resistance to temperature transport in the Memstill® module.

In selecting a suitable membrane for MD attention should be given to the shape of the pores in the membrane. Franken (1988) showed that pore systems containing 'sharp edges' have an advantage over more straight, cylindrical shaped pores, because such sharp edged pores are not easily wetted. Also the number of pores per m² membrane above the (calculated) Laplace diameter is relevant in selecting the optimal membrane, because such large pores are easily wetted. The large pores can be considered as pinholes, as they will bleed some of the feed water to the distillate, resulting in an adverse product water quality.

Of high importance to the economical feasibility of MD is the energy-efficiency of MD. This is defined by the ratio between energy transfer by water vapor transport (latent heat Q_l) and the total heat transfer (latent heat Q_l and conduction Q_c), and should as be close as possible to 100%.

$$\eta = \frac{Q_l}{Q_l + Q_c} \quad [10]$$

A low value for η will result in a much higher energy input to produce 1 m³ of water and hence jeopardize the feasibility. High values of η will require almost full blocking of sensible heat transfer by the membrane, as previously stated.

Results

Three pilots have been performed until end of 2008. Pilot number 1 was situated in the street of Johor near Singapore, using polluted seawater. Pilot number 2 was located in Rotterdam, using (brackish) seawater near the discharge point of River Rhine, and pilot number 3 used brackish water in the harbour of Rotterdam, polluted with surface active agents and high concentrations of sludge and silt. The feed for the third pilot was only filtered at a relatively large pore size of 80 μ m, whereas more elaborated pretreatment schemes (additional fine filtering and in pilot #1 also carbon adsorption) were used in the other pilots.

The feed and product water of pilot #2 have been analysed on a number of parameters. The typical results are presented in table 1. The table shows the excellent performance of the MD pilot. The electric conductivity has decreased by a factor 46,000, and the sodium and chloride content has decreased by at least a factor 70,000. The product meets international standards for distillate water. It shows that the number of 'pinholes' in the pilot modules are virtually absent. Also the internal water tightness of the module proved to be sufficiently secured. The separation performance of this MD pilot therefore outperforms other techniques such as RO and MSF.

Table 1 Separation performance of pilot #2 (date 01-09-2006); production capacity 1 m³/h.

		Feed	Distillate
Na	(mg/l)	7900	0.110
Cl	(mg/l)	13900	0.130
SO ₄	(mg/l)	1280	< 0.020
Ca	(mg/l)	300	0.039
Mg	(mg/l)	960	0.017
HCO ₃	(mg/l)	150	3.5
Silica	(mg/l)		0.275
SSolids	(mg/l)	9.2	< 1.0
pH		8.03	6.63
EGV	(μ S/cm)	34500	0.75

The three pilots were based on extensive bench scale testing work, using modules with the same length-width dimensions as in pilot #3, but having a small number of membranes, and using NaCl solutions at sea level concentration in clean tap water (0.60 M NaCl). The objective of the bench scale testing was to find the optimal configuration of membranes, spacers and condensation materials. Also ample time was spent on finding a way to guarantee the production of water tight modules. The bench scale work also provided data on the effects of the module configuration, the channel velocity and degassing of the feed water. Some data are provided in figure 3 as an example. Bench scale modules are numbered from M1 onward.

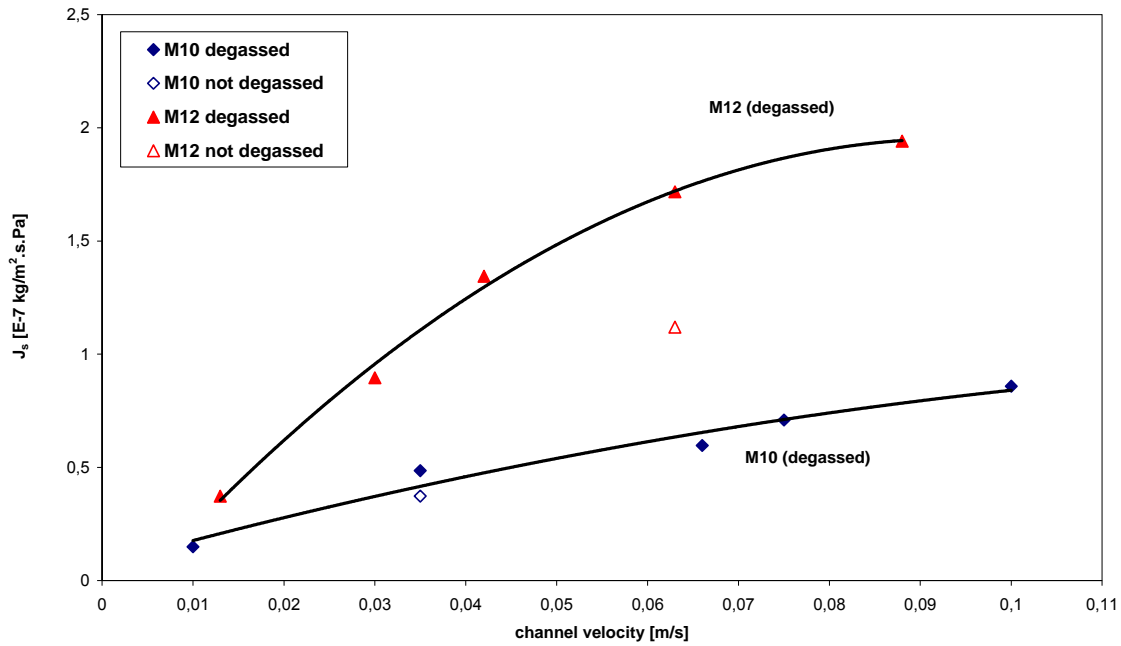


Figure 3 Specific flux vs. liquid velocity in the membrane channel in bench scale modules M10 en M12, showing the effect of different spacers and degassing of the feed.

Effects of degassing of the feed flow are shown in more detail in table 2. Degassing is performed with a separate membrane module in the feed flow and applying a low pressure (approx 0.15 bar absolute). The effect of degassing showed to be highly variable and is occasionally even negative, but in most cases a strong positive effect is found. The effect of degassing is largest at the lowest (average) temperature levels in the module because degassing will have a strong effect on the partial water vapor pressure in the membrane and hence the flux. This is clearly demonstrated in module M28.

Table 2: effect of degassing the MD feed

Module	Specific flux J_s ($\cdot 10^{-7}$ kg/(m ² .s.Pa))		T_{av}	Effect of degassing on specific flux
	Not degassed	Degassed		
M10	0.373	0.485	40 °C	+ 30%
M12	1.12	1.72	40 °C	+ 54%
M19	0.70	1.05	55 °C	+ 50%
M21	0.74	0.70	55 °C	- 5%
M22 (1 st run)	0.98	0.99	55 °C	+ 1%
M22	0.55	0.60	70 °C	+ 9%
M22 (2 nd run)	0.94	0.98	55 °C	+ 4%
M28	0.71	1.94	40 °C	+173%
M28	0.50	0.85	70 °C	+ 70%
M30	0.92	1.00	55 °C	+ 9%
M31	1.60	3.10	40 °C	+ 94%

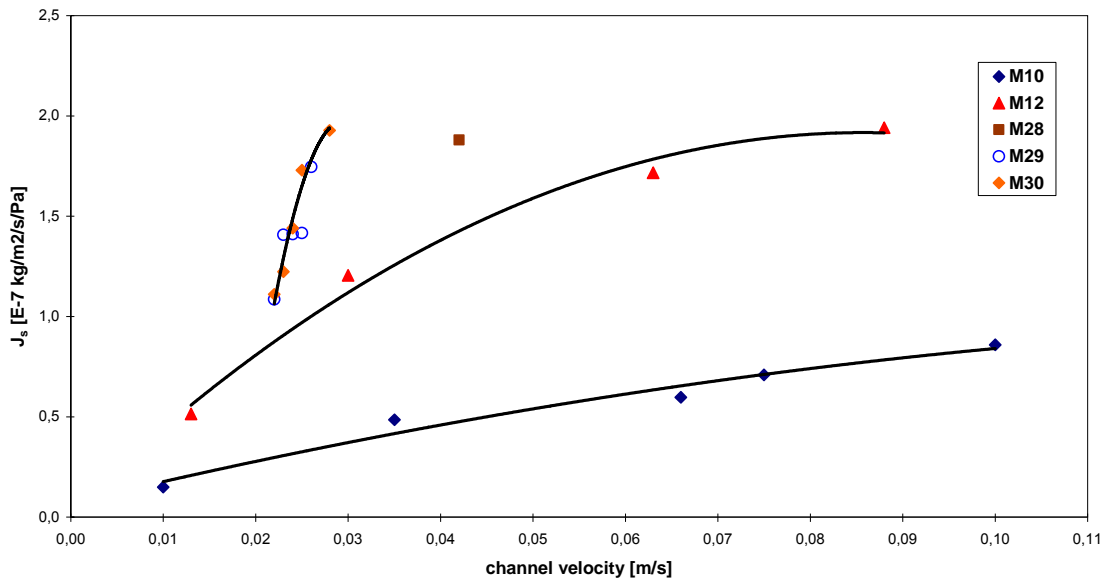


Fig 4: Specific flux in bench scale tests, at 40 °C and after degassing of the feed flow

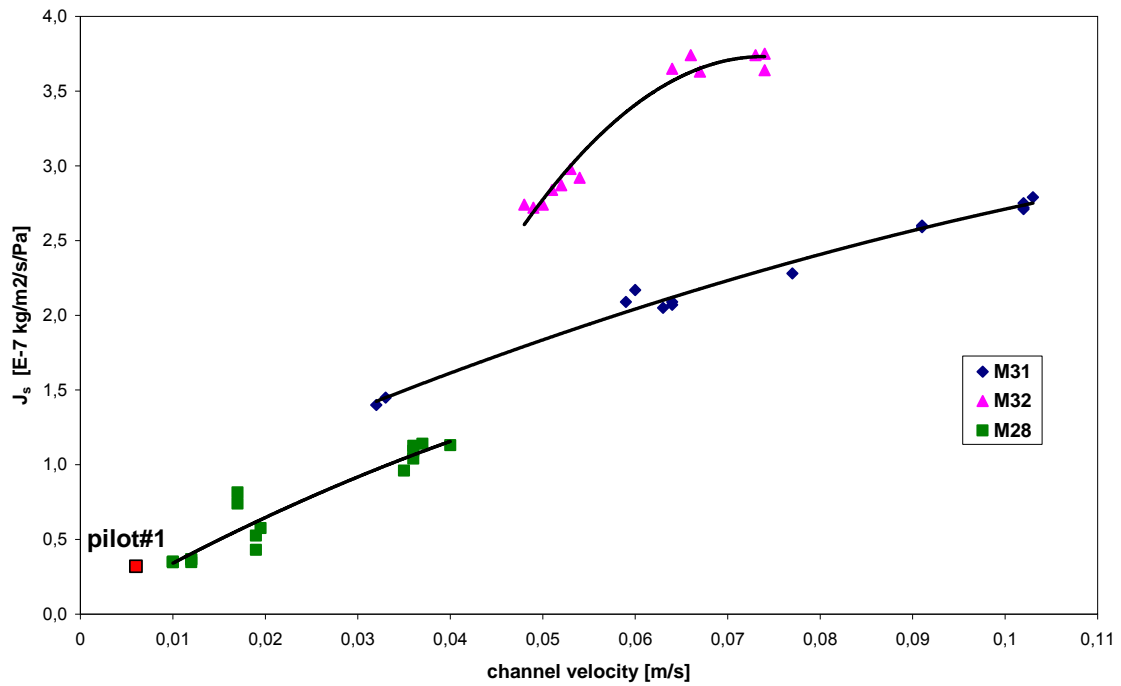


Fig 5: Specific flux vs. velocity membrane channel at 55 °C; selected bench scale modules after degassing of the feed (M28 uses a spacer that is different to M31 and M32). Pilot #1 is shown as reference. This pilot has the same configuration as M28

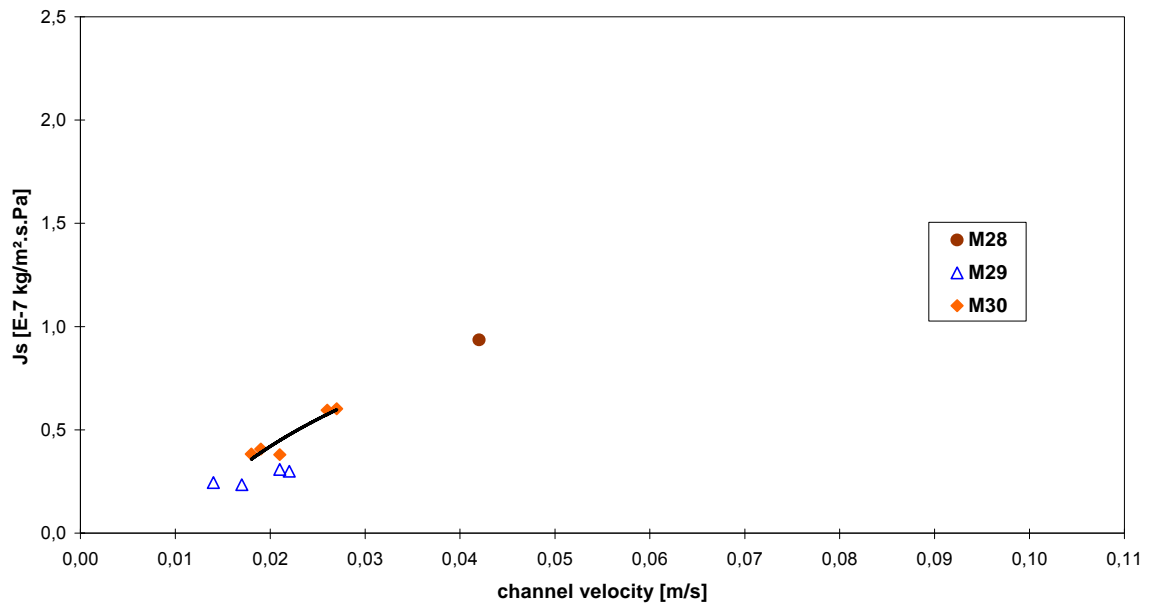


Figure 6: Specific flux as function of channel velocity, for various bench scale modules at 70 °C, after degassing of the feed.

Over 30 bench scale modules have been build and tested over extended periods of time. Module M32 showed best performances of the tested configurations: a specific flux between 2.7 and 3.8 10^{-7} kg/(m².s.Pa) has been obtained at superficial channel velocities between 0.05 and 0.075 m/s. The pressure drop in the liquid channel was still modest (in total 0.3 to 0.5 bar) and the average temperature level in these tests are close to what is needed in commercial installations. These fluxes equal a latent heat transport of between approx 3.3 kJ/(m².s) and 4.6 kJ/(m².s). Other modules (M28, M31, M33) showed approx. 40% lower specific fluxes, which are close to performances previously reported in literature, with modules not using the Memstill® concept (see for instance Aklaibi etal (1997), Guijt (2002)). This means that the overall performance of the Memstill® concept, including its additional heat resistances in the product channel and condenser material, is similar to concepts previously studied. This is attributed to both the 'milking effect' of the product (keeping the product water film very thin) and the improved condenser material with an extra low heat resistance.

The energy efficiency in most bench scale modules and the first two pilot modules varied between 50 and 75%, depending on actual process conditions and temperatures used. However, a thicker membrane (150% of the usual thickness) resulted in a better energy performance. Results indicated an energy efficiency between 70 and 90% in the third pilot. See table 3.

The heat requirement in the third pilot was on average 520 MJ per produced m³ of water. The bench scale tests required higher heat inputs, between 850 and 2400 MJ per m³ water. The low heat requirement in the pilot is attributed to the better blocking of sensible heat transport (thicker membrane) and the relatively small temperature difference between the main liquid channels. Unwanted energy losses to the surroundings will also be lower in larger installations.

Table 3: Energy efficiency of some bench scale modules and the third pilot, at low channel velocities

Module	Membrane thickness	Channel velocity (m.s ⁻¹)	T _{top} (°C)	Energy efficiency (%)	Flux (g.m ⁻² .s ⁻¹)	Specific flux J _s (*10 ⁻⁷ kg.m ⁻² .s ⁻¹ .Pa ⁻¹)
M28	100%	0,035	74,3	69,5	0.98	1,21
M31	100%	0,027	51,2	67,8	0.68	1,55
M32	100%	0,034	77,0	70,8	1.05	1,05
	100%	0,023	69,0	55,9	0.54	1,10
3 rd pilot	150%	0,022	77,9	89,4	0.78	1,44

It proved to be difficult to conclude on the performance of the pilots. A large data set was lost after an unauthorized reset of the data logger, other results were difficult to compare to bench scale results due to frequent and often unwanted changes in process conditions.

A substantial decline in specific flux has not been observed in pilot #1 and #3. However, a substantial reduction in specific flux over the 4 month testing period has been noted with pilot #2. This may be attributed to a failure in the de-aeration unit, which was then bypassed, but this conclusion is uncertain. During pilot #3 a more or less stable specific flux was obtained during the 4 month testing period, although the specific flux in one of the modules seemed to start at a higher value during the first few days. The specific flux in pilot #3 varied between typically 1.1 and 1.44 10^{-7} kg/(m².s.Pa), depending on the superficial channel velocity (between 0.011 and 0.022 m/s).

No serious fouling problems by biological growth or scaling have been found during the pilot tests. During the third pilot test frequent stand-stills and over 30 restarts were necessary due to technical failures in equipment outside the MD module. Silt and sludge accumulated in the modules during the frequent stand-stills of the installation, leading to an increased pressure drop in the module.

The channel velocity had to be reduced due to this accumulation of silt and sludge over time. Also, various internal and external leakages occurred during the testing of pilot # 3, which is attributed to the new, mechanical construction technique for the module and the little experience with this technique at that time. These leakages could be easily repaired by shutting off the leaking liquid channel; however, this resulted in a lower total production capacity of the module.

Evaluation

Energy efficiencies between 70 and 90% have been found for relative thick membranes. This indicates an heat requirement of approx. 0.1 to 0.4 times the heat of evaporation plus the sensible heat added at the hot side of the module (up to approx. 5 K), under assumption that the heat of condensation is fully recovered in the MD module. Heat requirements of approx. 300-400 MJ/m³ product water are now calculated from the pilot tests results for full scale installations. This is similar to energy requirements of large scale MSF and MED processes (200-400 MJ/m³). In recent thermal evaluations of the MD process we showed the feasibility of much lower energy consumption, i.e. 36 MJ/m³, which is important when heat has to be specially generated for the process. Further work on improvements of the membrane and the MD process are envisaged, with this value as goal. The improved energy efficiency is related to the reduced heat losses by conduction in the membrane. The heat losses are reduced by a factor two, simply by using a membrane of approx. twice the normal thickness. The product water rate in the tests remained however more or less constant, because the main resistance to mass transport is still located outside the membrane. i.e. the transport of heat from the condensing surface to the (mixed) feed water stream.

Relatively high energy performances will be found for high top temperatures. The losses by heat conduction are linearly related to the temperature difference between the evaporating surface and condensing surface on the membrane, whereas the driving force for the process, ΔP_m , will increase exponentially with temperature. The selected materials in the Memstill®-modules will limit for the time being the operating temperature to 80-85 °C. This temperature allows good performance of the MD process (ΔP_m up to approx. 0.1 bar), allows the use of low cost waste heat and avoids the need for pressurized systems. Applying waste heat in MD has the advantage that no additional fuels are needed, hence no additional CO₂ emissions are to be expected. Besides improved energy efficiency, we are also focusing on new applications of MD (e.g. wastewater treatment and full water-salt separations) and higher operating temperatures (by selecting new materials).

Although membrane distillation carries a number of drawbacks, it is believed that MD holds a strong promise for future desalination installations. Possible drawbacks of MD include:

1. relatively high energy consumption in the present state-of-the-art (although the energy source, i.e. waste heat, is usually low grade),
2. sensitive to surfactants, which may cause wetting of the membrane,
3. separate treatment may be needed to remove volatile contaminants from the product water.

The process needs no expensive hardware. The membrane modules itself and possibly the infrastructure to connect the MD process to the source of waste heat and the source of water are the most expensive parts. The process may be downsized and may still be economically feasible at a scale of 100 m³/day or even less. For instance, it may be used to produce high purity water in one single step from brackish water or even sea water.

The research has resulted in a configuration that outperforms previous work on MD, both in the fluxes obtained and, most of all, the scale of the process. The pilots used modules up to 300 m² membrane area and showed that serious problems with internal or external water leakages can be avoided or solved. Also no important reduction in process performance due to settling of solids in the module, frequent temperature variations, etc. have been noted. The modules are based on polymeric materials and have shown low costs for large scale production in economic evaluations. Capital cost for the installation are estimated to be 165 euro per m³/day capacity, including filtering at 40 µm and de-aeration as pretreatment steps (Hanemaaijer et al, 2006b).



Fig 7: photo of the module for pilot #1

Conclusions

A high selectivity of MD has been proven in pilot tests, resulting in a high quality distillate. Salt separation factors well above 10,000 have been obtained with a commercial available membrane product. An (near) optimal module configuration have been obtained, partly by trial and error research in bench scale tests. A very good flux has been found in combination with a) low pressure drops along the flow channels and b) low energy losses by internal conduction of heat. A thicker membrane may reduce the internal energy losses, resulting in better energy efficiencies, but it will also result in lower fluxes. However, the flux reduction by using a thicker membrane will be marginal when other flux determining factors in the process are still dominant. The effect of degassing of the influent on the process performance is non-conclusive, although almost all tests show a positive effect of degassing on the flux, as expected. The module has been scaled-up to a size where low cost applications become realistic. The present module contains up to 300 m² membranes. No scaling or biofouling problems have been found during pilot testing periods (between 4 and 14 months). The specific flux remained more or less stable over time, though it varied with the liquid velocity in the channels of the module. The modules have a simple construction process, with no corrosion problems expected during their use in warm seawater. Due to low production costs, low cost water production may become possible. The promise of low cost water production has been shown in various cost evaluations. Costs for large scale applications are typically between Euro 0.30 and 0.50 per m³. The use of low cost (waste) heat or an application in cogeneration, relatively high top temperatures (>350 K) and the use of simple water pretreatment (e.g. sieving at approx. 40 μm) are here prerequisites.

Symbols and abbreviations

A = surface (m²)
d = pore size (m)
 ΔH_v = heat of evaporation (J/kg)
J = water flux (kg/s/m²)
J_s = specific flux (kg/s/m²/Pa)
K_h = thermal conductivity of membrane (W/m/K)
K_m = specific mass transfer of membrane (kg/m²/s/Pa)
k_m = heat conductivity (W/m²/K)
L = length of the membrane channels
M = mass of water produced (kg)
m = mass flow of water per area of surface (kg/s/m²)
MD = membrane distillation
MED = multieffect distillation
MSF = multistage flashing
n = number of membranes in membrane module
RO = reverse osmosis
t = time (s)
 ΔP = water vapour pressure difference across membrane (Pa)
W = width of channel for main liquid flow
q = local heat transfer (W/m²)
Q = heat transfer (W)
T = temperature (Kelvin)
 ΔT_m = temperature difference across membrane (Kelvin)
 δ = membrane thickness (m)
 ϵ = porosity of the membrane (-)
 θ = thermal polarization (-), as defined in relation [4]
 η = energy efficiency of MD (-), as defined in relation [10]
 γ = surface tension of the solute (Pa.m)
 ϕ = wetting angle of the membrane material

Subscripts

1 = low temperature side of membrane module
2 = high temperature side of membrane module
av = arithmetic average (at retentate side)
case = external case of the MD module
top = high temperature side of module, the entering liquid
c = conduction
c = condensation (in Q_c)
ext = external surface
env = environmental conditions
h = hot stream or retentate stream
hm = hot stream, at membrane surface
l = latent heat
ln = logarithmic mean
m = at (or across) the membrane surface
p = liquid product stream
pm = liquid product stream, at membrane surface
s = specific
x = length parameter

Acknowledgement

The Memstill® development was supported by the Netherlands E.E.T. program, enabling the development of breakthrough –Technologies (T) towards simultaneous accomplishment of both Ecological (E) and economical (E) goals (contract EETK01001). In this development phase the consortium included the following parties: TNO, Keppel Seghers, Hamers Engineering (resulting in the new company Aquastill exclusively founded for commercialization of Memstill technology), E.ON Benelux, Evides, Heineken International, Gemeente Amsterdam Waterleidingbedrijf, University Twente and Ecological Management Foundation. Special thanks are given to the following persons for their valuable contributions to the project: Hans de Jong, Peter Jan Koele, Bart Nelemans, Eric van Sonsbeek, Chris Dotremont, Hette Hylkema, Ron Biemans and Allerd Stikker.

References

- M.E. Findley, Vaporization through membranes, *Ind. Eng. Chem. Process Des. Dev.*, Vol 6, nr 2, pg 226 (1967).
- E.Drioli, V.Calbrò, Y.Wu, Microporous membranes in membrane distillation; *Pure & appl. Chem*, 58, No 12. 1657 (1986)
- R.W. Schofield, A.G. Fell, C.J.D. Fell, Heat and mass transfer in membrane distillation, *J.Membr. Sci.*, 33, 299-313 (1987)
- A.G. Fane, R.W. Schofield, C.J.D. Fell, The efficient use of energy in membrane distillation, *Desalination*, 64, 231-243 (1987)
- A.C.M. Franken: Membrane distillation, a new approach using composite membranes; Thesis, University of Twente; ISBN 90-9002420-2 (1988)
- S. Bandini, C. Gostoli, G.C Sarti, Role of heat and mass transfer in membrane distillation process, *Desalination*, 81, 91-106 (1991)
- K.W. Lawson, D.R Lloyd, Membrane distillation (review), *J. Membr. Sci.*, 124, 1-25 (1997)
- J.H. Hanemaaijer, J.W. van Heuven, Method for the purification of a liquid by membrane distillation, PCT/NL2000/00366, EPA 00937369.7 (2000)
- C.M. Guyt: Influence of membrane and air gap on the performance of a membrane distillation module; Thesis, University of Twente; ISBN 90-365-1874-1 (2002)
- K.K.Sirkar, Baoan Li, Novel membrane and device for direct contact membrane-based desalination process: phase II, *Desalination and water purification research and development program final report No 96, NTIS* (2003)
- M.N. Chernyshov, G.W. Meindersma, A.B. de Haan, Modelling temperature and salt concentration distribution in membrane distillation feed channel; *Desalination*, 157, 315-324 (2003)
- A.M. Alklaibi, Noam Lior, Membrane-distillation desalination: status and potential, *Desalination*, 171, 111-131 (2004)
- M.N. Chernyshov, G.W. Meindersma, A.B. de Haan, Comparison of spacers for temperature polarization reduction in air gap membrane distillation, *Desalination*, 183, 363-374 (2005)
- Media Analytics Limited, abstract: <http://www.the-infoshop.com/report/gwi47246-desalination.html> (2006)

G.W. Meindersma, C.W. Guijt, A.B de Haan, Desalination and water recycling by air gap membrane distillation, *Desalination*, 187, 291-301 (2006)

J.H. Hanemaaijer, J. van Medevoort, H. de Jong, J.W. Assink, P.J. Koele, A.E. Jansen, Method for the purification of a liquid, WO 2008/054207 A8, PCT/NL2007/050524, EPA 06076956.9 (2006a)

J.H. Hanemaaijer, J. van Medevoort, A.E. Jansen, C. Dotremont, E. van Sonsbeek, Tao Yuan, L. De Ryck, Memstill membrane distillation – a future desalination technology, *Desalination*, 199, 175–176 (2006b)

C. Fritzmann, J. Löwenberg, T. Wintgens, T. Melin, State-of-the-art of reverse osmosis desalination, *Desalination*, 216, 1-76 (2007)

C. Charcosset, A review of membrane processes and renewable energies for desalination, *Desalination*, 245, 214-231 (2009)

Tsung-Ching Chen, Chii-Dong Ho, Ho-Ming Yeh, Theoretical modeling and experimental analysis of direct contact membrane distillation, *J. Membr Sci*, 330, 279-287 (2009)