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Desalination and water recycling by air gap membrane distillation

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Abstract

Membrane distillation (MD) is an emerging technology for desalination. Membrane distillation differs from other membrane technologies in that the driving force for desalination is the difference in vapour pressure of water across the membrane, rather than total pressure. The membranes for MD are hydrophobic, which allows water vapour (but not liquid water) to pass. The vapour pressure gradient is created by heating the source water, thereby elevating its vapour pressure. The major energy requirement is for low-grade thermal energy. It is expected that the total costs for drinking water with membrane distillation will be lower than \$0.50/m^3, even as low as \$0.26/m^3, depending on the source of the thermal energy required for the evaporation of water through the membrane.

Keywords: Desalination; Water recycling; Air gap membrane distillation; Water costs

1. Introduction

Of all the earth's water, 97% is salt water, only 1% is fresh water available for humans to drink and 2% is frozen. During the last 50 years, the earth's water demand has increased by a factor of four to 4,000 km^3/y. This is about 33% of the available water from rainfall. An increasing number of areas on our planet suffer from water scarcity, and many more will suffer in the near future because of the rapid depletion of groundwater and surface water. There are several ways to tackle this problem [1]:

- Retain (storage, conservation and rain harvesting)
- Reduce demand and spillage
- Recycle and reuse water
- Reinvent technology, improving efficiency
- Low-cost desalination of brackish and seawater

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Today, thousands of desalination plants are operating worldwide, collectively converting about 27.5 million m^3 of seawater and brackish water into fresh-water per day. This is just 3% of the world's consumption of drinking and sanitation water. At the expected decrease in desalination costs (typical production costs for large-scale units are now well below \$1.00/m^3), desalination capacity is projected to grow. Desalination costs for small-scale units range from \$1.00 to \$3.00/m^3 [1]. The desalination market 2005–2015 will generate expenditure in the region of \$95 billion, of which around \$48 billion will be derived from new capacity (\$30 billion of cap-ex and \$18 billion of op-ex) [2]. Additional capacity of 31–32 million m^3/d is expected to be commissioned during this period. This represents a 113–116% increase in the total active installed capacity over this period [1,2].

2. Existing desalination technologies

Current commercially available desalination technologies can be subdivided in thermal processes, which evaporate water to separate it from the salt that remains in the brine, and membrane processes, which make use of a membrane as a separating agent.

The thermal processes are multiple-stage flash evaporation (MSF), multiple-effect distillation (MED) and vapour compression (VC). The membrane processes are reverse osmosis (RO) and electrodialysis (ED). MSF and RO are of equal importance and together these two technologies account for 87% of the world-wide desalination capacity. The membrane processes, particularly reverse osmosis, will continue to take market share from thermal desalination, with 59% of the total new built capacity being membrane based. This reflects the growth of the market outside the Gulf region (the traditional heartland of the MSF thermal process), as well as increased use of RO technology to supplement MSF in the Gulf [2].

Membrane distillation (MD) is an emerging technology, which can be used for desalination, but also for recycling. Membrane distillation differs from other membrane technologies in that the driving force for desalination is the difference in vapour pressure of water across the membrane, rather than total pressure. The membranes for MD are hydrophobic, which allows water vapour (but not liquid water) to pass. The vapour pressure gradient is created by heating the source water, thereby elevating its vapour pressure. The major energy requirement is for low-grade thermal energy.

3. Membrane distillation configurations

A variety of methods have been employed to impose the vapour pressure difference across the hydrophobic membranes [3]. In every case, the raw water to be desalted directly contacts the hot side of the membrane.

Different MD configurations used in practice are depicted in Fig. 1. In all MD processes an aqueous feed solution is brought into contact with a micro-porous and hydrophobic membrane. The solution does not penetrate into the membrane pores, which is called membrane wetting, due to the hydrophobic nature of the membrane. The function of the membrane is to support a liquid–vapour interface; the membrane does not alter the vapour–liquid equilibrium. The driving force for mass transport across the membrane is a difference in partial vapour pressure of the moving species. The way in which this partial vapour pressure difference is created differs for every MD configuration, as will be explained with the help of Fig. 1.

3.1. Direct contact MD

In direct contact MD (Fig. 1a), the feed solution is in direct contact with the membrane on both sides. The temperature of the feed solution is higher than that of the permeate solution to

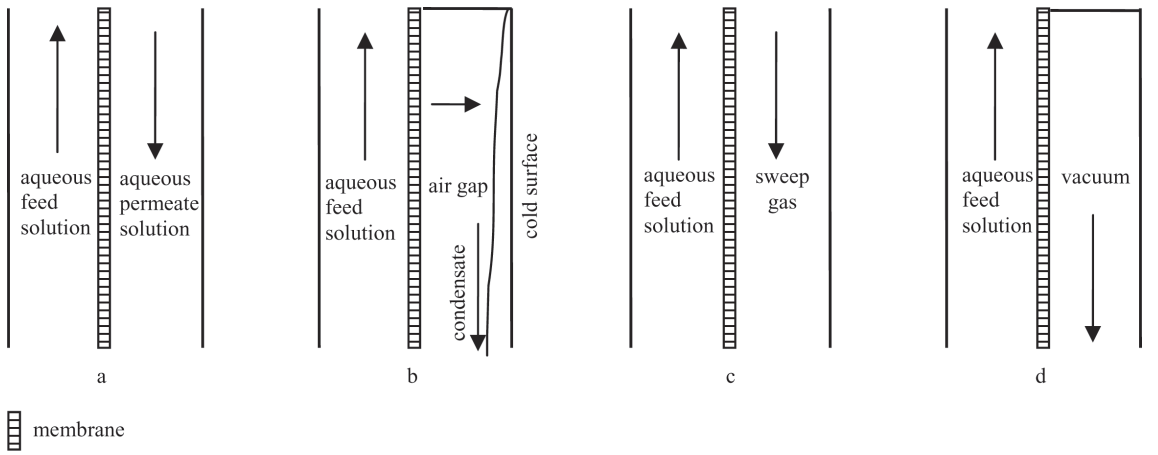

Fig. 1. MD configurations, (a) direct contact MD, (b) air gap MD, (c) sweep gas MD, and (d) vacuum MD [3].

create a driving force for vapour transport across the membrane. If the purpose of the process is to desalinate seawater, the permeate solution is fresh water. However, if the purpose of the process is to concentrate the feed solution, the permeate solution can be a strong salt solution to increase the driving force. The latter process is also called osmotic distillation. In osmotic distillation the temperature difference can be reduced to a large extent. Because the membrane is the only barrier between both solutions the obtained water vapour fluxes in direct contact MD are relatively high. Unfortunately this is also true for the energy flux by heat conduction, so that heat losses in direct contact MD are also relatively high.

3.2. Air gap MD

In air gap MD (Fig. 1b) only the feed solution is in direct contact with the membrane. The permeate is condensed on a cold surface. There is an air gap situated between the membrane and the cold surface to reduce energy loss by heat conduction through the membrane. The main drawback of the air gap is that it is also an additional resistance to mass transfer. Air gap MD is suitable for all direct contact MD applications. However, it is also suitable to separate other volatile substances, e.g. alcohols from an aqueous solution [4,5]. This is not possible in direct contact MD, because those substances are likely to wet the membrane at permeate side due to lower surface tension and/or smaller contact angle with the membrane. Since in air gap MD the permeate is not in direct contact with the membrane, there is no danger of membrane wetting at the permeate side in this case.

3.3. Sweep gas MD

In sweep gas MD (Fig. 1c), which is also called membrane air stripping, the vapour at the permeate side of the membrane is removed by a sweep gas and subsequently externally condensed. Like air gap MD, it can also be used for removing volatile substances other than water [6–8]. An advantage of using a sweep gas is that the resistance to mass transfer of the air gap is reduced substantially. However, drawback is the dilution of the vapour by the sweep gas, which leads to higher demands on the condenser capacity. Or, if a relatively small flow of sweep gas is used, heat transferred across the membrane causes a temperature increase of the sweep gas. This forms a problem, because it leads to higher vapour pressures at the permeate side and thus a lower driving force. Lately, Rivier et al. suggested a thermostatic sweep gas MD

module in which cooling water that flows along the adjacent wall cools the sweep gas within the module [9]. This configuration is a combination of (b) and (c) in Fig. 1.

3.4. Vacuum MD

Instead of using sweep gas the vapour can also be removed by evacuation (Fig. 1d) and subsequently external condensation. Vacuum MD can be used for the separation of various aqueous mixtures with volatile compounds [10–12], and recently it has also been proposed as a means for seawater desalination [13,14].

3.5. Memstill® concept

The Memstill® concept was developed by TNO, a scientific institution in the Netherlands, for desalination of seawater by air gap membrane distillation carried out in a counter current flow configuration. A schematic presentation of the technique is given in Fig. 2. Cold seawater flows through a condenser with non-permeable well-wettable walls via a heat exchanger into the membrane evaporator in counter current mode. The wall of the evaporator consists of a microporous hydrophobic membrane through which water vapour can diffuse and by which liquid water (with dissolved salts) is retained. The condenser and the membrane can either be tubular, as shown in Fig. 1, or as flat sheets with spacers between the sheets.

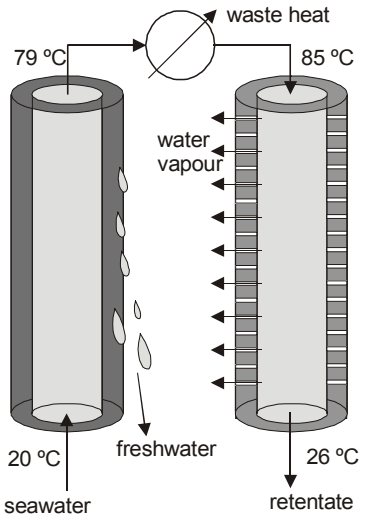

Fig. 2. Principle of Memstill® process.

The Memstill® concept can best be classified as air gap MD, Fig. 1b, in which the cold surface is cooled by the feed flow which is preheated by the condensing water vapour. As said before, a drawback of this configuration is the resistance to mass transfer of the air gap. Reduction of air gap width or air gap pressure or both will lead to a reduction of this resistance to mass transfer. Reduction of the air gap pressure does not mean that the process becomes a vacuum MD process, since the objective remains to condense the product water inside the module at the highest possible temperature.

4. Driving force: temperature difference and temperature level

In the Memstill® process the applied driving force is the temperature difference between the hot and the cold water flow. The direct driving force that makes the water molecules diffuse across membrane and air gap, is the water vapour pressure difference between the hot water surface at the inner membrane radius and the condense layer surface. At both surfaces local vapour–liquid equilibrium is assumed [3]. As can be seen in Fig. 3 the saturated water vapour pressure increases

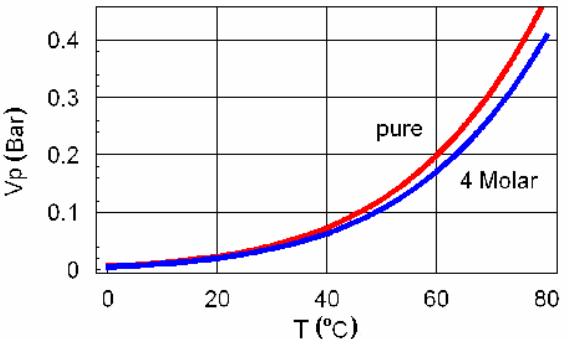

Fig. 3. Saturation pressure of pure water and concentrated brine as a function of temperature.

exponentially with temperature [14]. This means that a given temperature difference results in a larger flux with increasing hot water temperature. For example a temperature difference of 10°C results for hot water temperatures of 80°C and 40°C in vapour pressure differences of respectively 162 and 31 mbar, which differs a factor of five. So, the production of water vapour is much more efficient at higher temperatures.

5. Heat transport

In MD processes mass transport and energy transport are coupled. Water evaporates from the hot water flow, before it diffuses across membrane and air gap. When reaching the other side of the air gap, the water vapour condenses on the cold surface. So, heat of evaporation must be supplied from the hot water bulk to the hot water-membrane interface, and heat of evaporation must be withdrawn from the condense layer by the cold feed flow. This leads, qualitatively, to a temperature profile as is shown in Fig. 4.

Since a larger energy flux increases the temperature difference between the hot water bulk and the evaporating surface ($T_h - T_1$) and between the condensing surface and the cold water bulk ($T_3 - T_c$), it leads to a smaller net driving temperature difference between the evaporating and condensing water surfaces ($T_1 - T_3$). This phenomenon is often called temperature polarisation in membrane science and it reduces the water vapour flux. Thus, next to temperature level and temperature difference, the water vapour flux is also influenced by the horizontal energy flux across the module, which consists next to latent energy of evaporation that is coupled to the water vapour flux also of heat conduction.

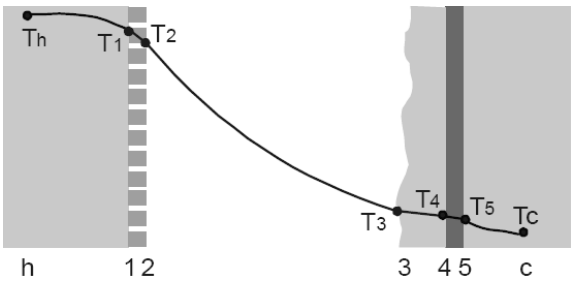

Fig. 4. Horizontal temperature profile in a Memstill® module. h = hot water flow, 1–2 = micro-porous hydrophobic membrane, 2–3 = air gap, 3–4 = condensed fresh water product layer, 4–5 cooling plate, c = cold water flow.

This energy flux formed by latent heat of evaporation is inherent to the process and cannot be reduced relatively to the vapour flux. The only way to minimise its negative influence is to create good conditions for heat transfer in the hot and cold water channel and have a well conducting cooling plate with enough capacity. Martínez-Díez et al. showed that in a flat sheet direct contact MD module the use of coarse channel spacers increases the flux by 30–40% in comparison with an empty channel [15], while they did not encounter any increase in pressure drop worth mentioning. The spacers increased the heat transfer coefficient from bulk to membrane surface from 1200–1900 W/m^{2}K (depending on flow velocity) for the empty channel to 3000–10,000 W/m^{2}K for the coarse spacer filled channel. For tubular and hollow fibre membranes the heat transfer coefficient with laminar flow is 2900 W/m^{2}K for an inner diameter of 1 mm which increases to 9700 W/m^{2}K for a 0.3 mm inner diameter fibre [16]. Turbulent flow with a Reynolds number of 5000 gives a heat transfer coefficient of 19,000 W/m^{2}K for the 1 mm inner diameter tube. The performance of tubes and hollow fibres is thus comparable to that of flat membranes with spacer filled channels in this respect.

Energy transport by heat conduction that forms the other part of the energy flux is especially important in direct contact MD. Fane et al. measured energy losses by heat conduction of 50–20% for a hot water temperature of 40–70°C [17], Ohta et al. measured at temperatures between 33–60°C energy losses as high as 90–70% [18], and Martínez-Díez et al. measured energy loses of 65–

48% for hot water temperatures between 21 and 48°C [19]. The negative influence of conductive energy loss is twofold. Firstly, the energy is lost for evaporation of water. Secondly, it leads to extra temperature polarisation because of the increased energy flux. This is why the use of an air gap to reduce energy loss can be so advantageous. Several studies show theoretically possible efficiencies of air gap MD between 80 and 95% [20–22]. But although the flux calculations have been confirmed by measurements, this is not the case for the energy efficiency calculations. Shoji Kubota et al. carried out measurements concerning energy loss in an air gap MD unit [23]. They measured energy losses of 75–50% at temperatures between 30 and 50°C, which are for a large part due to heat loss to the environment.

Description of the energy transport in a MD system is straightforward. Assuming steady state leads to a constant heat flow across all heat transfer resistances between the bulk at feed and permeate side. For the Memstill® process these resistances are: the hot water boundary layer, the membrane, the air gap, the product layer, the cooling plate, and the cold water boundary layer (Fig. 4). Gryta et al. showed that the heat transfer relations for heat exchangers are useful to describe heat transfer in the boundary layers of an MD process, although the correct relation should be carefully selected, especially for flat membranes [24,25]. The energy transport across membrane and air gap is usually described with a summation of latent heat of evaporation (water vapour flux times evaporation enthalpy) and heat conduction [3]. However, Gryta and Tomaszewska suggest that the change in sensible heat of the water vapour should also be taken into account, because of the temperature difference across the membrane [24]. The latter term is for the Memstill® process not very important (less than 1% of the latent heat transport).

Energy transfer equations are used to calculate all intermediate temperatures. Especially the temperatures at the hot water and product water surface (T_1 and T_3) are of interest to calculate the water vapour flux. The other intermediate temperatures can be used for optimisation considerations. E.g. a large temperature difference across the cooling plate ($T_4 - T_5$) suggests that the plate should be larger or thinner or that a better conducting material should be used. The energy equations cannot be solved apart from the mass transfer equations, since they contain the water vapour flux.

6. Mass transport

The mass transfer across the air gap needs no discussion because it is conveniently described by diffusion through stagnant air [26]. The driving force for mass transfer across the membrane is the water vapour pressure difference between feed and permeate side. Apart from a pressure difference as driving force, the temperature difference across the membrane also induces mass transfer, called the Soret effect [26]. However, Banat and Simandl showed that the thermal diffusion contribution towards mass transfer is positive but negligible in MD [27]. The recognised transport mechanisms for mass transfer across the membrane are usually molecular diffusion and Knudsen diffusion and, sometimes, viscous flow. Molecular diffusion has a partial pressure difference as driving force and non-identical molecules that are in the way form the resistance to mass transfer. The driving force for Knudsen diffusion is also a partial pressure difference, but in this case molecules bounce into the membrane matrix, which forms the resistance to mass transfer. Knudsen diffusion is thus important for small pores and/or low pressures. Finally, viscous flow has a total pressure difference as driving force, and the membrane matrix forms the resistance against it.

Many studies concerning direct contact MD [15–17,28] and also air gap MD [5] use a theory in which an overall mass transfer coefficient C, fitted to the experimental data, describes mass transfer across the membrane in direct contact MD

and across membrane and air gap together in air gap MD. C is defined according to Eq. (1), in which J is the water vapour flux in kg/m^{2}s and $\Delta p_{w,\text{surface}}$ is the water vapour pressure difference between evaporating and condensing surface.

$$J = C\Delta p_{w,\text{ surface}} \tag{1}$$

C is typically $3\cdot10^{-10}$–$22\cdot10^{-10}$ (m^3/m^{2}s.Pa) or 0.1–0.8 (kg/m^{2}h.mbar) for direct contact MD [13–15,26] and $3\cdot10^{-10}$–$7\cdot10^{-10}$ (m^3/m^{2}sPa) or 0.1–0.25 (kg/m^{2}h.mbar) for air gap MD [5]. A clear advantage of this method is that by the value of C different MD systems can be easily compared with respect to productivity. However, C is only a constant as long as temperature level and total pressure are constant. With increasing temperature (and constant, atmospheric pressure) the water vapour pressure increases and the fraction of air in the membrane pores and the air gap is decreased. Because the air fraction is the resistance to molecular diffusion, C increases with increasing temperature [29]. Decreasing the total pressure will also decrease the fraction of air and thus increase C. Furthermore, it should be recognised [16,29] that both Knudsen and molecular diffusion contribute to C. Lowering the total pressure makes Knudsen diffusion more important which will also have its influence on C. Since both temperature and pressure describe a large range in the Memstill® process, C cannot assumed to be constant and thus it is not convenient to use this theory.

A more useful theory for the Memstill® case combines the transport mechanisms explicitly and has constant membrane parameters that do not depend on process conditions. Such a theory is the dusty-gas model [30], which has been introduced in the MD-field by Lawson [31]. The dusty-gas model states that the total flux of a component is the sum of its viscous flux (J^V) and its diffusive flux (J^D), Eq. (4). The viscous flux is described with the Hagen–Poiseuille equation applied to a compressible gas, Eq. (3).

$$\frac{J^D/M_w}{K_0 v_{M,w}} + \frac{p_a J^D/M_w - p_w J_a^D/M_a}{K_1 P D_{wa}} = \frac{-1}{RT}\nabla p_w \tag{2}$$

$$J^V = \frac{-M_w p_w B_0}{RT\mu}\nabla P \tag{3}$$

$$J = J^V + J^D \tag{4}$$

The diffusive flux is a combination of Knudsen diffusion and molecular diffusion, the first and the second term on the left-hand side of Eq. (2) respectively.

What is important here is that K_0, K_1, and B_0 are constant parameters that represent the membrane mass transport properties for Knudsen diffusion, molecular diffusion, and viscous flow. All other variables in above equations are physical properties and process conditions. In several studies, which use sometimes only one or two of the transport mechanisms, the membrane mass transport properties are calculated from membrane pore size, porosity and an estimated tortuosity assuming that the pore structure of the membrane can be described with parallel cylindrical pores [4,19,24,30,32]. This approach gives a good idea of the order of magnitude of these constants, but is not very accurate. Mason and Malinauskas mention deviations of as much as a factor of five [30]. Lawson and Lloyd determine the Knudsen diffusion and viscous flow membrane morphology parameters with single gas permeation experiments at variable pressure, and subsequently fit the molecular diffusion parameter to the direct contact MD experiments [33–35]. A large advantage of measuring all membrane parameters separately is that the model calculations become predictive.

7. Current state of the Memstill® technology

Measurements with hollow fibre membranes with variable air gap width, at a hot water entrance temperature of 65°C show that, for air gaps of around 1.5 mm or smaller, the energy efficiency of the process decrease with more than 20%. This is due to thermal conduction taking place across product water, which forms water bridges between the membrane and the condenser wall. For large air gaps of 3 mm energy efficiencies of typically 85–90% were obtained [36].

The highest fluxes are obtained at the lowest possible pressures in the air gap. This lowest pressure is equal to the saturated water vapour pressure of the hot water entering the module. At this pressure, the air gap width is of negligible influence on the water vapour flux.

In order to minimize both concentration polarisation and temperature polarisation, research is carried out applying different spacers in the membrane channel. Supersaturation profiles of barium sulphate are shown to be dependent on the inlet temperature of the membrane channel, and at certain conditions a maximum value of the barium sulphate concentration lies in the bulk of the channel rather than at the membrane surface [37].

Biofouling is an issue when process water, waste water or surface water is being used as feed. In one laboratory plant ($5 \cdot 0.1$ m^2 membrane area) with water from a pond as feed, a flux decline, especially in the segment with the highest temperature (57°C), was observed after 800 h due to biofouling, but no evidence of breakthrough of micro-organisms was found with the membranes used. After 2200 h, the direction of the flow was reversed, which restored the original flux rate almost completely. Another lab installation ($2 \cdot 0.1$ m^2 membrane area) was in operation elsewhere during 6000 h with surface water as feed, after pre-treatment by coagulation/flocculation. In spite of severe biofouling of the membranes and partial clogging of some of the inlet channels (no prefilter was used), also here no evidence of bacterial breakthrough was observed.

Presently, pilot tests are being carried out with several modules up to a membrane area of 60 m^2. These pilot plants will be scaled up to 600 m^2 in the near future and pilot plants, with capacities ranging from 50 to 250 m^3/d, will be installed in The Netherlands and in Singapore, where process (waste) water and salt water will be used as feed.

8. Potential of the Memstill® technology

The energy consumption of the Memstill® is low (average 73.75 MJ/m^3) compared to MSF (147.5 MJ/m^3) and MED, because in the Memstill® technology, low grade heat, with a temperature of 50–100°C, or solar energy can be used to heat up the feed. Since Memstill® operates in counter-current mode efficient use of the energy supplied is obtained. Also in comparison with RO, the energy consumption is low. RO requires electrical energy for the high pressure pumps and MD only uses heat.

The Memstill® installation consists of prefab modules, which results in easy construction and minimal site work. Since the components are mainly made of polymeric materials, which are commercially available, the construction is light-weight and, therefore, requires a light foundation and is easy to (re-)locate. Due to its properties, a Memstill® unit can easily be placed on ships and drilling platforms in the off-shore industry for the production of drinking water from seawater. Because of the polymeric construction, corrosion plays only a limited role.

In MD, the driving force is a difference in vapour pressure on both sides of the membrane and this has the advantage that MD is less susceptible to fouling, compared to RO, where fouling and compaction of the fouling layer and the membrane are major drawbacks of this technology. The only pre-treatment for MD is removal of solids out of the feed stream. Therefore, less chemicals or pre-treatment are required for MD than for RO, MSF of MED.

Memstill® can both be applied in small-scale

as in large-scale applications and is suitable for a number of applications, such as production of drinking or ultrapure water from surface water, brackish water or seawater, concentration of brines and treatment of waste streams.

9. Cost comparison

In Fig. 5, the present and expected costs for several desalination processes are shown. From this figure, it can be seen that the RO costs have decreased dramatically. The main reasons for this are the lower module costs, the development of energy-efficient RO membranes and energy recovery systems, which recover about 50% of the energy required for RO.

The process characteristics for the Memstill® process are:

- Specific flux: $J_s = 1.5 \cdot 10^{-10}$ m³/m².s.Pa
- Heat energy: 80–240 MJ/m³
- Production: 25–50 m³/d.module
- Recovery: 50%

Since the costs for the thermal processes are not expected to decrease to a level lower than about \$1.00/m³, the Memstill® process will only be compared to RO. In Table 1, the energy and investment costs of Memstill® MD and RO are compared for a seawater desalination capacity of 105,000 m³/d with different process conditions. The heat supply to the MD process can be fuel fired, generated by cogeneration of heat and electricity or by a waste heat source. For RO, there are two configurations calculated: a minimal and a standard one.

Only in the case of a waste heat source, the energy costs of MD are at the same level or lower than those of RO. The hardware costs for MD are much lower than those for RO, mainly because for MD no high-pressure pumps are required. On the other hand, the costs for MD modules are higher because of the separate membrane and condenser compartments, but the total fixed costs for Memstill® are \$0.16–0.17/m³, compared to \$0.25–0.35/m³ for RO. Also, the cost for cleaning of the membranes is higher for RO than for MD. The lowest total costs of the water produced are obtained with a Memstill® installation with a cheap waste heat source of \$0.10/GJ.

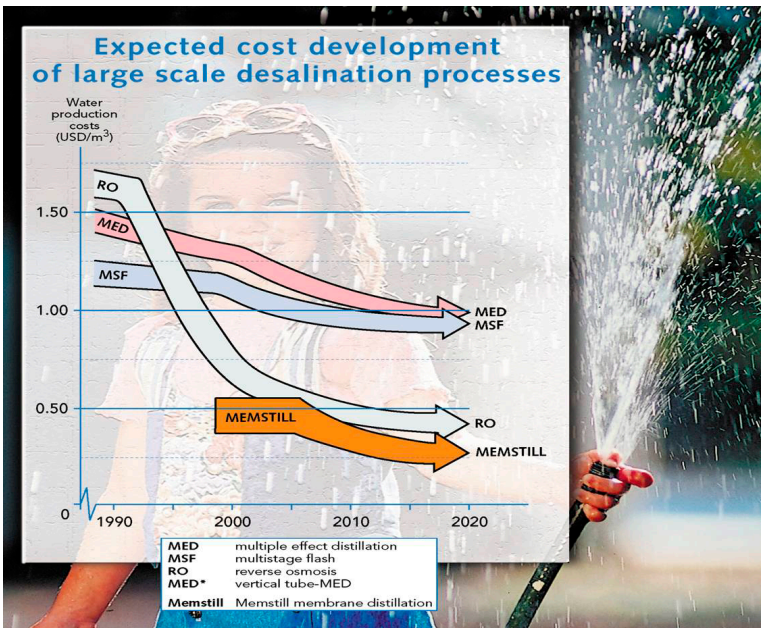

Fig. 5. Expected costs development of large scale desalination processes (from the Memstill® brochure).

Table 1
Water production costs of Memstill® vs. RO (seawater 105,000 m^3/d)

	Memstill®		Memstill®		RO	RO
	Fuel fired	Co-gener.	Waste heat		Min.	Stand.
Energy costs						
Heat, MJ/m^3	231	231	139	139	—	—
(Costs in $/GJ)	(1.30)	(0.50)	(0.50)	(0.10)		
Electricity, kWh/m^3	0.75	0.75	0.75	0.75	2.5	4.5
Heat costs, $/$m^3$	*0.30*	*0.12*	*0.07*	*0.01*	—	—
Electricity costs, $/$m^3$	*0.03*	*0.03*	*0.03*	*0.03*	*0.10*	*0.18*
Fixed costs						
Hardware, excl. membranes, $/$m^3$.d	165		165		750	1024
Module costs, $/$m^3$.d	214		233		35	49
Hardware costs, $/$m^3$	*0.05*	*0.05*	*0.05*	*0.05*	*0.23*	*0.32*
Module costs, $/$m^3$	*0.11*	*0.11*	*0.12*	*0.12*	*0.02*	*0.03*
Auxiliary costs						
O&M, chemicals, filters etc., $/$m^3$	*0.05*	*0.05*	*0.05*	*0.05*	*0.10*	*0.10*
Water costs total, $/$m^3$	***0.54***	***0.35***	***0.31***	***0.26***	***0.45***	***0.63***

10. Conclusions

Air gap membrane distillation is a promising desalination process as it is the most efficient MD process. Because a continuum of evaporative stages in an almost ideal counter-current flow process is occurring in a Memstill® module, a very high recovery of evaporation heat is possible.

Both sweet (surface or waste) water and salt water can be used as feed for the Memstill® module. No breakthrough of micro-organisms was found during a test of 6000 hours with pre-treated surface water as feed.

With waste heat as an energy source, the total water costs with the Memstill® process can be as low as $0.26/$m^3$. The lowest costs of other desalination processes (RO) are at least $0.45/$m^3$.

The Memstill® technology is in operation on pilot plant scale and must still be proven on a larger scale.

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