

RECENT ADVANCES IN MEMBRANE SCIENCE AND TECHNOLOGY IN SEAWATER DESALINATION – WITH TECHNOLOGY DEVELOPMENT IN THE MIDDLE EAST AND SINGAPORE

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Summary

The development of seawater desalination membranes is continuously progressing and has demonstrated highly remarkable achievement in performance. Indeed, there appears to be no limit to the performance of these membranes in sight. The first historic discovery of Loeb-Sourirajan asymmetric reverse osmosis (RO) membrane enabled seawater desalination on an industrial scale. Later, the membrane performance was improved by the development of thin-film-composite (TFC) membranes by Cadotte. The fluxes of currently available commercial membranes are an order of magnitude higher than the RO membranes of 1960s with salt rejection as high as 99.8 %. Further attempts are being made to increase the flux and to improve membrane stability by incorporating nanoparticles and by modifying the membrane surface. Moreover, fabrication of membranes that can reject boron, arsenic, and organic compounds of low molecular weights to higher degrees than currently available membranes is required to produce less hazardous drinking water. Membranes that can withstand ultra-high pressure for seawater desalination and membranes that are operable at ultra-low pressures for brackish water desalination were achieved for drinking water production. Typically, membrane distillation, forward osmosis, and carbon nanotube membranes are considered potentially applicable for seawater desalination. The current status and future direction of membrane desalination in the Middle East and Singapore have been discussed as case studies.

1. Introduction

It is reported that 96.5% of the earth's water is located in seas and oceans, 1.7% in the ice caps, 0.8% is considered to be fresh water with the rest being brackish water. Since water shortage has been a problem for many communities and humans have been

searching for the solution for a long time, desalination, turning salty water into fresh water, is not necessarily a new concept. Today the shortage of drinking water is a serious world-wide concern due to population growth and the increased demand for drinking water that exceeds readily available water resources. Over 1 billion people are without clean drinking water and approximately 2.3 billion people (41% of the world population) are living in water scarce regions (Greenlee et al, 2009). As a result, people have started to search for solutions with water reuse and seawater desalination as the keys for the sustainable growth of human activities. Water reuse is basically for the production of water for uses such as irrigation, power plant cooling water, industrial process water and ground water recharge. It has also been accepted as a method for the production of drinking water in Singapore (Newater). Alternatively, desalination has become a primary source of the drinking water production. Current desalination technology consists of two methods. One is a thermal desalination method that has been developed over the past 60 years, while the other is membrane processes that have been developed over the past 40 years.

Desalination is a general term for methods to remove salt from salty water to produce fresh water. Notably, the definition of fresh water depends on the country. For example, the US Environmental Protection Agency (EPA) has non-enforceable standards of 250 mg/L chloride and 500 mg/L total dissolved salts (TDS) for fresh water (EPA, 2002). The World Health Organization (WHO) and the Gulf Drinking Water standards recommended a drinking water standard of 1000 mg/L TDS (Fritzmam et al, 2007). In comparison to the government standards, most desalination facilities are designed to achieve a TDS of 500 mg/L or less (Greenlee et al, 2009). When the desalinated water is used for other purposes, e.g. crop irrigation, the TDS concentration may be higher. The feed water salinity for desalination facilities ranges from 1000 mg/L to 60,000 mg/L. Most of the seawater resources contain 30,000 to 45,000 mg/L TDS, while the brackish water within a range of 1,000 to 10,000 mg/L is treated by Reverse Osmosis (RO) (Greenlee et al, 2009). As mentioned above, desalination processes fall into the following two categories, i.e. thermal processes and membrane processes. The thermal process has been used for many years since the prehistoric era but the operation of the large scale distillation plant for drinking water production began in 1950s (Greenlee et al, 2009). In the beginning a process called multi-effect distillation (MED) was used but later a process called multi-stage flash (MSF) distillation was developed. The Middle East as a whole holds about 50% of the world desalination capacity and primarily uses MSF technology. However, facilities based on the membrane process have rapidly been installed since the 1960s and now surpass the thermal process in new plant installations. Outside of the Middle East, new RO plant installations have been continuing steadily. In 2001, 51% of the new installations were based on RO process, while in 2003, RO process accounted for 75% of the new production capacity.

RO, nanofiltration (NF) and electro-dialysis (ED) are the typical membrane processes available for desalination. RO and NF are called pressure-driven membrane processes since the transmembrane pressure difference is the driving force for the mass transport, while for ED the electrical potential difference is the driving force for the mass (ions) transport. ED is an older membrane desalination process than RO and NF. NF is a relatively new membrane separation process developed in 1980s. While NF can not desalinate seawater to produce drinking water in one step, it can be used successfully to

treat the mildly brackish water (Greenlee et al, 2009). It is hence desirable to use NF in combination with RO for seawater desalination. The most important feature of NF is its capacity to remove divalent ions such as calcium and magnesium that contribute to hardness of water. However, RO membrane can remove monovalent ions such as sodium and chloride and hence has become the mainstream of membrane desalination technology. Salt rejection as high as 99.7 and 99.8%, can be achieved by RO (Hydranautics, 2007; Greenlee et al, 2009; Reverberi and Gorenflo, 2007). RO membrane technology is also applicable for both seawater and brackish water desalination.

As mentioned earlier, the first countries that used the desalination process on a large scale for drinking water production were in the Middle East. Seawater desalination plants began to be developed in the 1950s, and the first industrial desalination plant was opened in Kuwait in 1960s. The first successful RO plant used brackish water as feed in the late 1960s (Amjad, 1993). In the following decades, membrane permeability was much improved and RO membranes were then applied for seawater desalination (Van der Bruggen and Vandecasteele, 2002). From the early 1960s to the end of the 1990s, the membrane productivity (flux) and salt rejection have much improved as shown in Figure 1. Table 1 also shows similar advancement made for the Dow RO membranes.

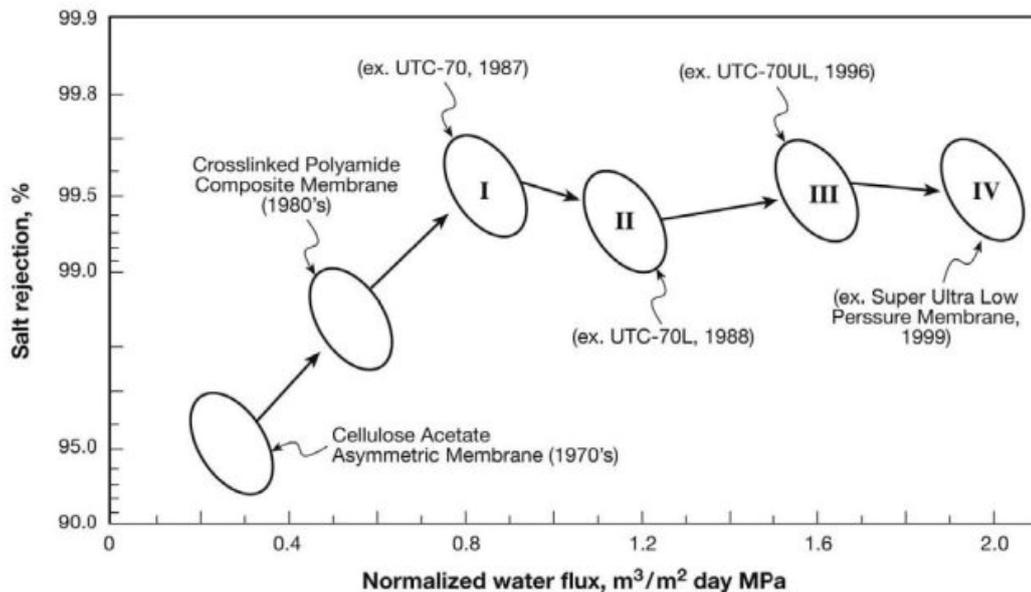


Figure 1. Progress in membrane performance during the past forty years. Reverse osmosis conditions: feed NaCl concentration, 1500-3000 ppm; operating pressure, 0.5-3.0 MPa; temperature, 25°C; pH, 6.5. I, II, III and IV are fully aromatic polyamide TFC membranes (adapted from Kurihara and Fusaoka, 1999)

Year	Production capacity (gpd)	Salt rejection (%)
Brackish water		
1990	8,000	98
1998	10,000	99.2

2007	11,000	99.8
Seawater		
1990	4,000	99.4
1998	5,500	99.5
2007	7,500-8,000	99.8

Table 1. Advances in Dow RO membrane

From 1996 to 2007 the rejection of typical seawater desalination membrane increased from 99.6 to 99.8% and the flux increases from 43 to 69 L/m² day bar (Mickols et al, 2005). As of 2009, over 15,000 desalination plants were in operation world-wide, and approximately 50% of those are RO plants. A new recent trend is to construct large seawater desalination plants with a production capacity of 100,000 m³/day or more (Greenlee et al, 2009).

Saudi Arabia is currently the world leader in desalination with approximately 26% of global production capacity, followed by the United States (17%). In Saudi Arabia most of the desalination plants are based on the thermal process (newly constructed plants are different) and the source water is seawater. In contrast, in the United States 69% of the desalination plants are based on RO and only 7% is seawater desalination plants. While only 20% of the total number of the desalination plants world-wide use thermal process, 50% of the total production capacity is based on the thermal processes. Israel has opened the world's largest seawater RO desalination plant with a production capacity of 330,000 m³/day, or 100 million m³/year. The United Arab Emirates (UAE) opened its Fujairah desalination plant in 2005 with a combined MSF and RO production capacity of 454,000 m³/day.

2. Membrane Processes for Desalination: Overview

2.1. Reverse Osmosis History

As mentioned, RO is currently dominant in membrane separation processes for desalination. Interestingly, the early development of RO membrane for seawater desalination began with the following fundamental equation called the Gibbs Adsorption Isotherm (Sourirajan, 1970).

$$\Gamma = -\frac{1}{RT} \frac{\partial \sigma}{\partial \ln a}, \quad (1)$$

where Γ is the surface excess, R is universal gas constant, T is absolute temperature, σ is surface tension and a is activity.

The equation predicts the presence of a very thin pure water layer at the surface of saline water. Table 2 summarizes the thickness of such pure water layers calculated on the basis of Eq. (1). As the table shows the thickness ranges from 2.6 to 5.6 nm, depending on the concentration of sodium chloride.

Table 2. The thickness of pure water layer at the air/sodium chloride solution interface

It was Yuster's group at University of California at Los Angeles (UCLA) who attempted to skim the pure surface water layer mechanically (Yuster et al, 1958). However, the attempt failed. Applying the concept underlying the Gibbs Adsorption Isotherm, Sourirajan further attempted to collect the pure water layer by applying pressure on the saline water that was brought to contact with one side of a membrane. Sourirajan's attempt was soon found to be successful and desalinated water was produced as permeate from the membrane. After initial attempts using cellulose and silicone coated cellulose membranes, a commercial cellulose acetate (CA) membrane was used, which resulted in a high salt rejection. Even though the latter rejection was considered high enough for drinking water production, the flux of water was extremely low with few drops of permeate collected in a day and the membrane was thought to be practically useless. It should be noted that Reid and Breton obtained, quite independently, similar experimental results of seawater desalination by using a CA membrane at the University of Florida (Reid and Breton, 1959). It took another 4 years for Loeb and Sourirajan to develop CA membranes with fluxes of practical usefulness, which opened up the avenue to the novel membrane desalination process, called Reverse Osmosis (RO).

According to Sourirajan, the mechanism of desalination by RO membrane is as follows: When salty water; e.g, sodium chloride solution, is in contact with the surface of a membrane, an interfacial pure water layer is formed at the solution/membrane interface. Assuming analogy between sodium chloride solution-air interface and sodium chloride solution-membrane interface, the interfacial pure water layer, t , is as thick as a fraction of nanometer. In the presence of a pore, the diameter of which is smaller than or equal to $2t$, the pure water layer will flow through the pore under the pressure applied on the sodium chloride solution and appear on the other side of the membrane as permeate (see Figure 2a). If the pore size is larger than $2t$, the sodium chloride solution will flow in the central region of the pore, contaminating the permeate (Figure 2b).

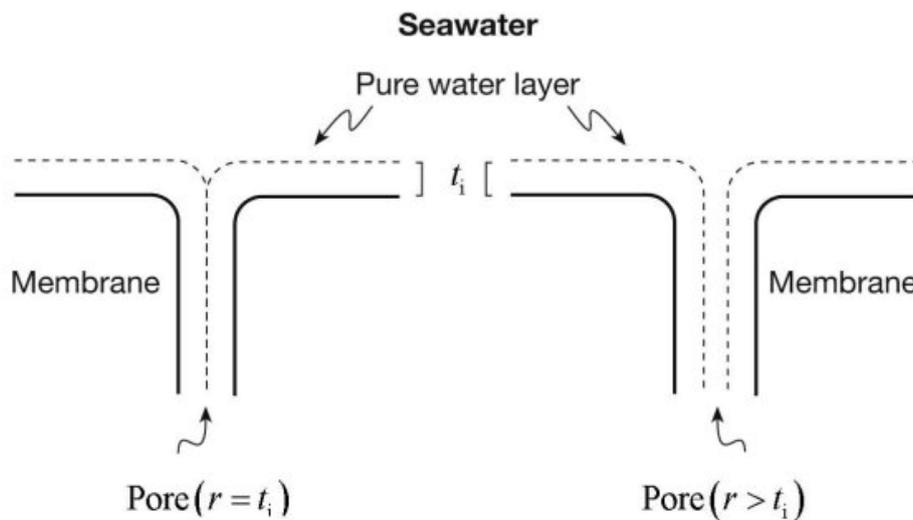


Figure 2. Preferential sorption capillary flow mechanism of RO

Thus, there are twin requirements for the success of RO membrane development. One is the presence of the interfacial pure water layer and the other is the formation of pores, the sizes of which are as small as a fraction of nanometer.

The pore sizes of RO membranes were evaluated theoretically by several researchers. For example, Schultz and Assumma concluded that the size of the pore should be 1.85 nm, based on the SEM images of polymer nodules at the surface of a CA RO membrane (Schultz and Assumma, 1970). Glückauf calculated the salt (sodium chloride) rejection as a function of the pore size based on the Onsager-Samalas equation for the dielectric force (Glückauf, 1965).

Positronium Annihilation Lifetime Spectroscopy (PALS) has become a popular tool recently to characterize the subnanopores. For example, Boussu et al characterized (in-depth) six commercial NF membranes.

By using depth-selective PALS measurements, it was revealed that the skin layer had two different pore sizes: a pore size of 1.25-155 Å as well as a pore size of 3.20-3.90 Å (both depending on the membrane type) (Boussu et al, 2007). Thus, the presence of sub-nanometer pores predicted in the 1960s can now be measured by a sophisticated instrumental technique.

2.2. Asymmetric Structure of the Membrane

Most of membranes that are used in industrial separation processes have an asymmetric structure and so are called asymmetric membranes. Figure 3 shows schematically a typical cross-sectional view of an asymmetric membrane (Matsuura, 1994). As shown in the figure, an asymmetric membrane consists of two layers; i.e. one very thin dense layer at the top of the membrane and a porous sublayer underneath the top dense layer (also called top skin layer).

While the top dense layer governs the permeation properties of the membrane, the porous sub-layer only provides the membrane with mechanical strength. When the material of the top skin layer and the porous sublayer are the same, the membrane is called integrally skinned asymmetric membrane. This type of membrane is made by the dry-wet phase inversion technique. When the polymer for the top skin layer is different from the polymer for the porous sub-layer, the membrane is called composite membrane.

The advantage of the composite membrane over the integrally skinned asymmetric membrane is that the material for the top skin layer and for the porous sublayer can be chosen separately to optimize the overall performance.

This type of membrane is made by coating a thin layer on top of the surface of a porous substrate. Various coating techniques are available but the interfacial in-situ polymerization method has been proven to be commercially most successful.

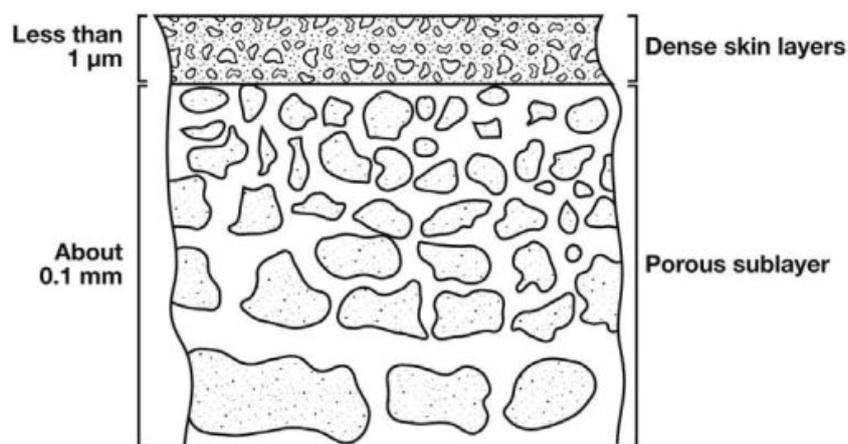


Figure 3. Asymmetric structure of the membrane

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