Reverse Osmosis: A History and Explanation of the Technology and How It Became So Important for Desalination

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ABSTRACT

From cellulose acetate (CA) hollow fiber membranes to advanced thin-film composite membranes in a spiral-wound configuration, developments in reverse osmosis (RO) membrane technology have allowed the desalination industry to tackle an even broader range of applications than ever before. This paper focuses on the advancements in membrane chemistry that have allowed RO membranes to offer extremely high salt rejection, operate at lower pressures, and handle a wide range of operating conditions. In addition, this paper discusses the history of RO element design, including an explanation of the industry-standard spiral-wound element design and why this design allows for RO to be a critical technology for an ever-increasing amount of desalination applications. Current RO desalination applications will also be discussed so that the reader can better understand the practical relevance of both RO membrane chemistry and element progression. Finally, this paper touches on future advancements in RO membrane technology and where the RO desalination industry is headed. Overall, an explanation and history of RO membrane chemistry and element design along with a discussion of how this technology is applied as a result will give the reader a fundamental understanding of RO as a desalination technology.

INTRODUCTION

Reverse osmosis (RO) is a membrane-based separation technology used to remove dissolved solids from a liquid. While RO can be used in a variety of non-water applications, the majority of RO applications are water-based, which will be the focus of this paper. The breadth of water applications that RO can be used in helped the technology become the multi-billion-dollar market it is today. In fact, the global RO market is expected to reach USD 8.87 billion by 2021 with a compound annual growth rate (CAGR) of greater than 9% (Reverse Osmosis Membrane Market, 2017). As evidenced by its large market size, RO has become a critical separation technology in modern water desalination applications.

Since RO is such a widely used technology in desalination applications, it is important to understand the history and development of modern RO elements. This paper discusses the history and development of RO membrane chemistry and RO membrane elements. In addition, this paper discusses RO desalination applications and the future of RO in desalination. Overall, the reader will be provided an excellent background of the technology and the desalination applications it is used in.

MEMBRANE CHEMISTRY HISTORY

As a naturally-occurring phenomenon, osmosis is the natural migration of pure water from a region of lower solute concentration across a semi-permeable membrane to a region of higher solute concentration. RO essentially "reverses" this natural migration by using applied pressure to force pure water to flow from a region of higher solute concentration across a semi-permeable membrane to a region of lower solute concentration. As such, RO can be used to concentrate salts and produce water with a lower salt concentration.



Figure 1: RO uses applied pressure to force pure water from a region of higher solute concentration across a semi-permeable membrane to a region of lower solute concentration.

One of the key things needed to apply RO for practical use is a semi-permeable membrane. The use of a semi-permeable membrane was first demonstrated by Jean-Antoine Nollet, a French physicist, in 1748 (History of Reverse Osmosis Filtration, n.d.). Nollet utilized a pig's bladder as a membrane to demonstrate the natural process of osmosis and showed that solvent could selectively pass through the bladder-membrane. This idea of using a membrane to selectively pass and reject certain species did not further progress until the mid-twentieth century when it became relevant again.

THE FIRST RO MEMBRANE – In the aftermath of World War II, it was evident that water shortages were going to become increasingly problematic in the United States. As such, Senator and then President John F. Kennedy began advocating the idea of large-scale desalination (Cooley, 2006). Encouraged by this public interest in desalination, researchers at the University of California, Los Angeles (UCLA) began developing the first RO membrane. In 1959, researchers Sidney Loeb and Srinivasa Sourirajan created the first asymmetric RO membrane out of cellulose acetate (Loeb, 1981). The Loeb-Sourirajan membrane selectively rejected sodium chloride and total dissolved solids (TDS) under pressure while allowing water to pass through, which is exactly what was needed to make RO membranes successful in desalination.

Following the successful development of the Loeb-Sourirajan membrane, Loeb and Joseph McCutchan, a professor at UCLA, were given the opportunity to pilot the technology at a city water plant in Coalinga, California. To prepare for this opportunity, UCLA did further development work on the original Loeb-Sourirajan membrane to improve its strength, performance, and salt rejection characteristics (McCutchan, 1970). On June 4, 1965, Coalinga began their pilot testing of an RO membrane to produce additional water for their inhabitants. After three-and-a-half years of successful piloting, it was evident that RO membranes were a great answer to President Kennedy's request for large-scale desalination.

After the Coalinga piloting success, more and more desalination plants installed cellulose acetate RO membrane systems in the 1960s and 1970s. In addition to municipal desalination, power plants began using cellulose acetate RO systems to desalinate water for their industrial processes. Over time, it became evident that RO could be used in even more applications if only the membrane could tolerate a wider range of operating conditions. While the cellulose acetate membrane was a great start, its narrow design tolerances ultimately limited its practical applications. Cellulose acetate membranes are only stable operating in pH ranges of 4 to 7, and can only operate in temperatures up to 35°C. In addition, cellulose acetate RO membranes operate at relatively high pressures in the 350-450 psi range. To increase the applicability of RO in desalination, a next generation membrane was needed that had wider operating tolerances and a lower operating pressure.

THE NEXT GENERATION RO MEMBRANE – Following the success of initial membrane chemistries, researchers began focusing on the next generation of RO membrane that would be able to tackle an even wider range of operating conditions. One of these researchers, John Cadotte of North Star Technologies (later part of FILMTEC), developed the first interfacial composite polyamide membrane in 1972 (Kucera, 2015). This new membrane featured both

better rejection and flux at lower operating pressures than the previously-developed cellulose acetate membranes.

Over the next few years, Cadotte worked on additional membranes based off this initial success with a composite polyamide membrane. By the late 1970s, Cadotte had developed a fully aromatic interfacial composite membrane, which was ultimately patented in 1978 as FT30 membrane by FILMTEC (now part of DOW). This FT30 membrane, which featured a polyamide chemistry and a thin-film composite design (Figure 2), ultimately became the industry-standard chemistry for RO membranes that is still used today.

With its higher rejection, higher flux, and higher strength, this polyamide RO chemistry is utilized as a standard brackish water chemistry by most RO membrane manufacturers today. Its thin-film composite structure differs from cellulose acetate membrane structure as shown in Figure 2 below. While cellulose acetate membranes have a support layer and a cellulose acetate layer, thin-film composite membranes feature a support layer, usually made of polyester, a substrate layer, usually made of polysulfone, and a polyamide active layer. Most modern nanofiltration (NF) and RO flat sheet membranes utilize this thin-film membrane design, which has allowed membrane manufacturers to develop new chemistries to meet industry demands. However, it is worth noting that cellulose acetate RO membranes still have a fit in niche applications today due to their higher chlorine tolerance compared to thin-film composite membranes.



Figure 2: Comparison of cellulose acetate (2.1) and thin-film composite membrane (2.2).

MODERN RO MEMBRANES – Due to the success of the thin-film composite polyamide patented by Cadotte, membrane manufacturers began exploring additional RO membrane chemistries to meet the demands of an even broader range of applications. Now that RO desalination systems could produce low-TDS water, end users began pushing for energy-usage reduction, fouling reduction, and even higher rejection. As such, membrane manufacturers began exploring low-energy, low-fouling, and high-rejection membranes.

To reduce energy usage, membrane manufacturers began working on membranes that could operate at lower pressures than standard brackish water RO membranes. In 1995, Hydranautics introduced the first low-energy polyamide RO membrane (Kucera, 2015). This membrane

featured higher fluxes than standard brackish water RO membranes at the time, which allowed the membrane to produce the same amount of permeate but at a lower pressure. Unfortunately, to operate at a higher flux, the membrane had to sacrifice rejection. This is the basis for modern low-energy membranes, which are offered by most RO membrane companies. Over the years, further work has been done to improve rejection while maintaining a high flux, and a wide range of membranes with varying rejections and operating pressures are available on the market today.

Another characteristic that is very important to RO system operation is fouling. Throughout normal operation, RO membranes in desalination processes accumulate foulants, including scale, silt, organic matter, and more. Since scale could be mostly handled with antiscalants and silt could largely be removed in pretreatment, membrane manufacturers began focusing on making RO membranes that were resistant to organic fouling. In 1994, TriSep Corporation (now part of MICRODYN-NADIR) introduced the first low-fouling thin-film composite membrane chemistry (Kucera, 2015). This membrane featured a more neutral surface charge than standard polyamide RO membranes to resist fouling, making its low-fouling characteristics inherent to the membrane itself. Over the years, several other membrane companies also developed low-fouling membranes to handle the growing industry's request to resist organic fouling.

As shown by initial membrane improvements focusing on these characteristics, improving salt rejection and producing low-TDS water is critical in RO desalination applications. Throughout RO membrane history and to this day, membrane companies focus on creating higher rejecting membranes by slightly altering membrane chemistry and/or membrane casting conditions. As a result, modern seawater RO membranes, which are used to treat high salinity feeds (TDS of 35,000+ ppm), often have rejections rated at 99.8%, and even as high as 99.85% (Kucera, 2015). Even modern brackish water RO membranes, which are used to treat feeds of ~10,000 ppm TDS or less, consistently feature rejections of 99.5% and some even up to 99.7%. These high rejections are perfect for many applications where low TDS is critical, such as high purity water applications.

Overall, many types of modern RO membranes are available to handle a wide variety of operating conditions. Whether the application requires lower energy usage, fouling resistance, or high rejection, modern RO membranes are available to take on the challenge (see Table 1 below for a summary).

RO Membrane Type	Characteristic
Brackish water	For feeds with <10,000 ppm TDS
Low energy	Higher fluxing, operates at lower pressures
Low fouling	Resists organic fouling
High rejection	Standard is 99.5%, up to 99.7%
Seawater	For feeds with 35,000-50,000 ppm TDS
High rejection	Standard is 99.7-99.8%, up to 99.85%

Table 1: Overview of modern RO membranes.
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MEMBRANE ELEMENT HISTORY

Over time, RO element design has evolved to meet the operating conditions of an increasing amount of desalination applications. As with membrane chemistry, initial element designs worked, but still had plenty of room for improvement.

INITIAL RO ELEMENT DESIGNS – The original Loeb-Sourirajan membrane was utilized in a flat sheet format in a plate-and-frame design. The membrane itself had little mechanical integrity, so it was mounted on plates that were placed in a unit for operation (McCutchan, 1970). When it became time to pilot at Coalinga, it was evident that this plate-and-frame design was both cost-prohibitive and riddled with design flaws. For instance, the membranes were difficult to seal in place, there were dead flow areas, and the equipment was difficult to handle. To make sure the Coalinga pilot would be successful, UCLA researchers began investigating an alternative membrane format: a tubular design.

Tubular modules consist of membrane tubes up to 1 inch in diameter placed within a larger central tube, as shown in Figure 3 below. To make the tubular design successful, UCLA adjusted the formulation of the original Loeb-Sourirajan membrane to increase both mechanical integrity and salt rejection. This allowed the membrane to be cast in tubes for use in modules at Coalinga. While tubular designs were initially promising and are still used in certain applications today, further RO system piloting demonstrated that RO elements need to have as much membrane area as possible to handle as much flow as possible.

As such, capillary and eventually hollow fiber membranes were developed to increase packing density in membrane elements, thus increasing membrane area within the element (Figure 3). Capillary membranes feature a smaller diameter than tubular membranes, and hollow fiber membranes feature the smallest membrane diameter yet. Hollow fiber membranes are still widely used today, but they are not the primary element design for RO membranes. Fiber breakages and non-optimal manufacturing led the RO industry to seek another better membrane element design.





SPIRAL-WOUND ELEMENT DESIGN – Now that RO membranes had clearly established themselves as a mainstay within the desalination market, various membrane companies began to

emerge. These membrane companies began tackling the issue of element design and ultimately created the configuration that remains the most commonly used RO membrane element configuration today: the spiral-wound element. In 1963, Gulf General Atomics, which later became known as Fluid Systems and ultimately a part of Koch Membrane Systems, developed the first spiral-wound module. Gulf General Atomics put additional research effort into this design and developed the first multi-leaf spiral-wound element in 1968 (Kucera, 2015). This is the element design that current membrane manufacturers utilize today.

Spiral-wound elements feature a high membrane packing density while maintaining a small footprint. In addition, spiral-wound element configuration allows for simpler system plumbing, maintenance, footprint, and design than other element configurations do. All of these characteristics of the spiral-wound element design ultimately allowed the RO membrane desalination industry to rapidly expand.

Spiral-wound elements are comprised of "leaves" rolled around a central permeate tube. These leaves contain two sheets of flat-sheet membrane with their active sides facing outward and separated by a permeate carrier in-between. The leaves are glued on three sides so that permeate can only go through the membrane, after which it travels down the permeate carrier to the central permeate tube (Figure 4.1 below). Each leaf is then separated by a feed spacer so that feed water can effectively flow through the element. These leaves and feed spacer sheets are then rolled up around the tube and are covered with an outerwrap to maintain the cylindrical shape of the element (Figures 4.2-4.3). The most common outerwrap for RO elements used in desalination is fiberglass, but other outerwraps are available based on the application (e.g. sanitary outerwraps for food and dairy applications).



Figure 4: Spiral-wound element construction: 4.1 shows the internal components, 4.2 shows how these are rolled into an element, and 4.3 shows a final spiral-wound element.

REVERSE OSMOSIS APPLICATIONS

The use of RO originated when the Kennedy administration set a goal to develop a method to desalinate water to combat water shortages in the United States. Today, spiral-wound RO membrane elements are used for various desalination processes and applications around the world including:

- 1. Desalination for potable use
- 2. Industrial water & wastewater reclamation and reuse
 - a. Cooling tower blowdown
 - b. Boiler feed water
- 3. High purity water (for pharmaceuticals and microelectronics)
- 4. Spot-free rinse
- 5. Ingredient water (for food & beverages)

DESALINATION FOR POTABLE USE – Desalination of ground or surface water, including rivers, lakes, wetlands, and oceans, is increasingly being used to provide drinking water due to the shortage of fresh water supplies. According to the World Health Organization, shortage of fresh water supplies is "…estimated to affect one in three people on every continent of the globe, and almost one fifth of the world's population live in areas where water is physically scarce" (World Health Organization, 2011).

Municipal water supplies are often contaminated with a number of chemicals that may be hazardous to human health; such as chlorine (often added to the water supply to kill bacteria), heavy metals (byproducts from industrial processes and consumer waste) and pesticides (runoff from fields where crops are sprayed). RO elements have proven to be a successful and cost-effective technology to remove harmful heavy metals, disinfectant byproducts, organics, hardness, TDS, and color to meet safe drinking water quality requirements.

INDUSTRIAL WATER & WASTEWATER RECLAMATION AND REUSE – Water and wastewater reclamation and reuse in industrial power plants has become a growing trend in the past decade due to rising water demand and scarcity. In fact, water treatment including the use of RO for power generation is estimated at more than 30 percent of all industrial water treatment sales globally (Water Technology, 2017). RO is now used to treat recycled cooling tower blowdown in zero-liquid discharge (ZLD) applications as well as to purify boiler feed water.

<u>Cooling Tower Blowdown</u> Many industrial and power plants use cooling towers as an efficient means to remove large amounts of heat from hot equipment. Cooling these plants requires a considerable amount of water, however: typically, 80% (IDE Technologies, n.d.) or even up to 90% of the plant's total water needs (WaterWorld, n.d.). With increasing water scarcity in many areas, the treatment and reuse of cooling tower blowdown with RO was introduced.

Cool water circulating through a plant absorbs heat from the equipment and becomes warmer. When this now-hot water is circulated back to the cooling tower, some of the water is lost due to evaporation or the misting effect (when water is lost by drifting into the air). As water evaporates from the system, some of the dissolved solids (TDS) are left behind, causing the remaining water to become more concentrated in TDS. Instead of discharging this water and replacing it with new fresh water, RO is used to remove most of the TDS so that the water can be reused throughout the plant for cooling purposes. Many industries are relying on reusing cooling tower blowdown to reduce their demand on local fresh water supplies.

<u>Boiler Feed Water</u> Purifying water for steam boilers is one of the most common RO desalination applications. Steam boilers used in industrial and power plants require high purity feed water to ensure proper operation. Steam boilers used in industrial and power plants require high purity feed water to prevent scale buildup and reduce corrosion rates. The boiler feed quality requirement varies depending on the manufacturer and whether it is a low-, medium- or high-pressure boiler.

RO may be used to replace ion exchange, including sodium softening, to purify water for low- and medium- pressure boilers. For higher-pressure boilers and steam generators, RO is often used in conjunction with ion exchange and can significantly reduce the frequency of resin regenerations and drastically reduce the amount of chemical and regeneration waste. In some cases, a secondary RO system may replace ion exchange to further purify permeate from the primary RO system. Including RO systems in the boiler feed water makeup circuit has proven to increase production reliability and significantly reduce operating and maintenance costs (Kucera, 2015).

HIGH PURITY WATER – The pharmaceutical and microelectronics industries require highlypurified water for production purposes. In the pharmaceutical industry, water is the most common ingredient among all pharmaceutical drugs, so it must be free of bacteria, organics, and all dissolved solids. RO systems with ultraviolet (UV) sterilizers are often used to treat and disinfect feed water at industrial levels to produce the ultrapure water required for many pharmaceutical production processes.

In the microelectronics industry, high purity water is necessary during manufacturing phases and is used to rinse finished microelectronic components. For example, after processing, capacitors and transistors require ultrapure water from RO systems for cooling. If this water is contaminated with impurities, the products may become stained or corroded before they even leave the production plant. RO technology allows manufactures to create microelectronics of the best quality while reducing operating and maintenance costs of manufacturing equipment (Reverse Osmosis Water Treatment, Purification, Filtration Systems, n.d.).

SPOT FREE RINSE – For many industrial manufacturing plants that build parts for vehicles or aircrafts and commercial businesses like car washes, surface cleaning and rinsing can be an important part of the manufacturing process. Water that has not been filtered properly may lead to scaling and can leave white or grey water spots on freshly cleaned surfaces. These spots are caused by the calcium and magnesium salts (also referred to as hardness) in the source water. RO can remove the minerals that cause scaling and spotting to occur (Pure Aqua, Inc., n.d.).

INGREDIENT WATER – While it is obvious that high quality water is needed for the bottled water industry, it is also a critical ingredient in other food & beverage processes. Purified water is required for both production quality and preventative health measures. RO in conjunction with UV sterilizers can meet these needs by reducing dissolved solids and removing pollutants and microorganisms. Additionally, RO systems are useful in food and beverage manufacturing, processing, and packaging because they can remove many contaminants that may alter the taste or even the odor of the food or beverage.

FUTURE OF REVERSE OSMOSIS IN DESALINATION

Water desalination has become one of the most important methods of alleviating water shortages across the world. While RO has proven very successful thus far, the technology continues to advance. As RO is being used to treat a greater variety of feed waters, including municipal and industrial wastewater effluents as well as other water sources, further improvements are still necessary.

LOW-FOULING MEMBRANES – As RO feed water sources become more complex, there is an ongoing need for membrane elements that have resistance to fouling, scaling, and degradation while simultaneously offering high permeability and solute rejection. Reduced fouling from suspended solids and organics would make membranes even more cost-effective by extending their operational lifetime and lowering their energy requirements. Some low-fouling membranes exist today; research and development surrounding these membranes is focused on modifying the membrane surface. Systems utilizing RO have also increased their pretreatment processes to help remove contaminants before the feed water reaches the membranes.

CHLORINE-RESISTANT MEMBRANES – Membranes with chemical stability are also needed to expand the range of applications suitable for RO desalination. Surface waters and wastewater feed sources often contain biological materials and nutrients that feed bacteria. These materials may severely foul RO membranes and are therefore killed with the use of chlorine prior to RO. However, most polyamide membranes are not tolerant to chlorine and other oxidizers, so they must be removed prior to RO. Exposure to oxidizers may eventually damage the membrane causing the membrane to be susceptible to higher permeate flow and higher salt passage. Developing chlorine (and other oxidizing chemicals and halogens)-resistant membranes is important as RO is being used to treat increasingly challenging feed waters. Improving RO membranes' tolerance to oxidizers would also reduce operating costs by eliminating dechlorination steps prior to the RO system.

ALTERNATIVE MEMBRANE MATERIALS – RO membranes have been enhanced over the years, but most of these developments have been achieved through modifications to the membrane surface itself or improving the element or module design. While manufacturers continue to make modifications to the current, commercially available thin-film composite polyamide RO membranes and elements, researchers continue to look towards additional materials that may offer an attractive alternative to polymeric materials.

The use of nanotechnology has allowed for the development of nano-enabled membranes (NEMs), or membranes functionalized with discrete nanoparticles or nanotubes (Buonomenna,

2013). Such non-polymeric RO membranes include inorganic membranes, membranes of a combination of inorganic and polymer, and biomimetic membranes (Kucera, 2015). Some examples include:

- 1. Low-fouling membranes have been developed by coating a polyamide thin-film composite membrane with titanium particles. Studies have shown that along with UV light radiation, this titanium oxide coated membrane has self-cleaning properties as well as increased flux (Madaeni, 2007).
- NanoH₂O Inc. commercialized a membrane in which zeolite nanoparticles were synthesized and embedded within an RO membrane to reduce the overall energy demand in the desalination process. This nanoparticle and polymeric membrane (called thin-film nanocomposite, or TFN) has proven to be more productive (100% more permeability compared to conventional RO membranes while maintaining the same level of salt rejection) and resistant to bacteria (QuantumFlux Reverse Osmosis, 2015).
- 3. Antibacterial and antifouling carbon nanotube (CNT) membranes have been developed. The hollow CNT structure provides frictionless transport of water molecules, minimizing the use of pressure and resulting in a high fluxing membrane. High water flux with selective salt rejection has increased the demand for CNT membranes in desalination. However, the CNT membrane synthesis is still at a premature stage and needs further improvement (Das, 2014).
- 4. Biomimetic membranes incorporate biological elements or ideas from biological systems; taking advantage of the strategies evolved by nature for improving transport efficiency and specificity (Shen, 2013). Biomimetic RO membranes are made of a combination of biologically derived membrane proteins known as aquaporins and bio-inspired synthetic polymers. This allows for the membrane to utilize the aquaporin's selectivity and productivity (Biomimetic Membranes, n.d.).

While successful research has been done on different membrane materials, further development is needed to determine how to mass produce such membranes efficiently and at reasonable costs.

HANDLING HIGHER-TDS FEEDS – With the amount of ZLD applications increasing globally, an economical desalination technology was needed to handle feeds with TDS levels greater than what standard seawater RO membranes could handle (>50,000 ppm TDS). Forward osmosis (FO) is one of the evolving membrane technologies in desalination that has been developed to handle higher-TDS feeds. FO membrane processes differ from standard RO processes in that they use a "draw solution" of a higher salt concentration than the feed solution to force water from the feed into the draw solution. The water is then recovered from the draw solution in a separate process. This process that involve two sets of inlet and outlet flows across the membrane to tackle higher-TDS feeds include counter-flow reverse osmosis (CFRO) and pressure retarded osmosis (PRO). As evidenced by these emerging desalination membrane technologies, membranes will be a mainstay in desalination applications for years to come.

CONCLUSION

As shown throughout the course of this paper, RO membranes have come a long way over the years to meet the challenges of a constantly-evolving desalination market. From humble beginnings as cellulose membranes in the 1960s, modern standard RO membranes feature highly-engineered polyamide membrane chemistries with a thin-film composite design. The developments in membrane chemistry alone have allowed the industry to tackle a much broader range of operating conditions. In addition, membrane modules have evolved from early plate-and-frame, tubular, and hollow-fiber designs to modern spiral-wound elements efficiently packed with membrane area. This spiral-wound module design has allowed system design and operation to become simpler, further expanding the RO desalination market.

These robust membrane and element designs have allowed RO to tackle a broad range of desalination applications, including potable desalination, wastewater reuse, generation of high purity water, and much more. Further membrane developments are being carried out to broaden this range of applications even more. RO membranes themselves are being improved upon to increase rejection, decrease fouling, and tolerate higher concentrations of chlorine. Furthermore, new membrane technologies, such as FO, CFRO, and PRO, are being developed to meet stricter and stricter discharge requirements globally.

As water becomes increasingly scarce and environmental regulations become increasingly strict, it is obvious that desalination applications are here to stay. To meet the needs of these applications, RO membrane technologies will continue to evolve to rise to the challenge.

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