

Pressure retarded osmosis: From the vision of Sidney Loeb to the first prototype installation – Review

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ABSTRACT

The energy released from the mixing of freshwater with saltwater is a source of renewable energy that can be harvested using pressure retarded osmosis (PRO). In PRO, water from a low salinity solution permeates through a membrane into a pressurized, high salinity solution; power is obtained by depressurizing the permeate through a hydroturbine. The combination of increased interest in renewable and sustainable sources of power production and recent progress in membrane science has led to a spike in PRO interest in the last decade. This interest culminated in the first prototype installation of PRO which opened in Norway in late 2009. Although many investigators would suggest there is still lack of theoretical and experimental investigations to ensure the success of scaled-up PRO, the Norway installation has evoked several specialized and main-stream press news articles. Whether the installation and the press it has received will also boost competitive commercialization of membranes and modules for PRO applications remains to be seen. This state-of-the-art review paper tells the unusual journey of PRO, from the pioneering days in the middle of the 20th century to the first experimental installation.

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1. Introduction

Continuous dependence on fossil fuel combustion is accelerating changes in our climate toward dangerous long-term effects [1,2]. New renewable and sustainable sources of energy production must be explored to reduce reliance on fossil fuels use [3]. Currently, the most developed renewable energy sources include solar, wind, biomass, geothermal, and hydro [4]. Another potential source of renewable and sustainable energy is the salination of water, or the energy released

from the mixing of freshwater with saltwater [5]. One process of capturing the energy released from the mixing of freshwater with saltwater is called pressure retarded osmosis (PRO). In PRO, water from a low salinity solution permeates through a membrane into a pressurized, high salinity solution; power is obtained by depressurizing the permeate through a hydroturbine.

PRO systems can be classified based on their configuration: open-loop or closed-loop. Open-loop systems are solar-driven processes where renewable energy is produced from the mixing of relatively freshwater with saltwater. A variety of natural waters can be utilized, with the vast majority of studies investigating the mixing of river water with seawater in estuary systems [6]. Fig. 1a shows how the energy spent by the sun to evaporate water from the sea is recovered during the

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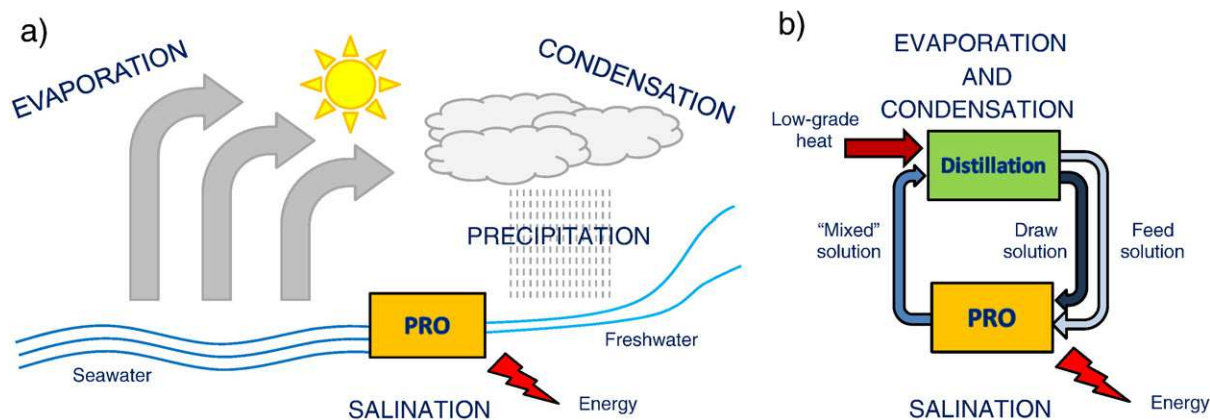


Fig. 1. (a) Schematic diagram of an open-loop water salination cycle using PRO to recover solar energy. This system is referred to as an open-loop system because the freshwater is “lost” to the sea. (b) Schematic diagram of a closed-loop PRO system to recover heat energy. This system is often referred to as an osmotic heat engine.

mixing of freshwater with seawater in an estuary. This energy can be quantified by imagining that every stream and river in the world ends at its mouth by a waterfall 225 m high, or the height of a seawater column that develops 22.4 atm of pressure [7]. It is estimated that the global energy production from the mixing of freshwater and saltwater in estuaries would be on the order of 2000 TWh per year [6], while the estimated global energy production from all renewable sources is approaching 10,000 TWh per year [4]. In other locations, such as terminal lakes, open-loop mixing of river water or seawater with hypersaline water is possible [8–10]. In the Great Salt Lake, the feasibility of harvesting the chemical potential difference between fresh river waters and the hypersaline water of the Great Salt Lake was investigated for power generation [8,9]. In the Dead Sea, harvesting the chemical potential difference between Gulf of Aqaba seawater and the hypersaline water of the Dead Sea was also investigated [10].

Closed-loop PRO systems, on the other hand, are designed to convert low-grade heat into mechanical work (Fig. 1b) [11]. Although this configuration does not represent renewable energy, when low-grade heat is available, these systems may be a viable source of sustainable energy [12]. This concept was first illustrated by Loeb in 1975 [11] and was recently revitalized by a publication of McGinnis et al. [12].

PRO can be seen as the inverse process of reverse osmosis (RO). Whereas RO uses hydraulic pressure (i.e., energy) to oppose, and exceed, the osmotic pressure of an aqueous feed solution (e.g., seawater) to produce purified water (i.e., fresh water) [13], PRO uses the osmotic pressure of seawater to salinate fresh water and induce hydraulic pressure (i.e., energy). Because of its similarities with RO, initial efforts to develop PRO relied on membranes and membrane modules originally designed for RO. This enabled the collection of early experimental results without the need for specifically tailored apparatus, but also resulted in power outputs far below expected outputs [14–17]. However, it was the use of pressure exchangers that were originally developed for RO applications that enabled substantial PRO design improvements [18]. Furthermore, it was the availability of Hydration Technology Innovations (HTI) (Scottsdale, AZ) commercial membranes for forward osmosis (FO) (another emerging osmotic process) that enabled more realistic testing of the PRO process.

This paper presents a state-of-the-art review of PRO including its unusual history of development that begins in the 1950s, remains quiet for 20 years, and then timidly expands for another 20 years until receiving significant attention over the past decade. The review follows a timeline from the early days to the most recent developments of this technology with special consideration given to the role of Sidney Loeb and his substantial contributions during four decades of research and publication on PRO. Strengths and limitations of the PRO process are reviewed and discussed in relation to the future of PRO technology.

2. Timeline

2.1. Early studies (1950s)

The concept of harvesting energy generated from the mixing of freshwater and saltwater was first reported in a *Nature* article by Pattle in 1954 [5]. Pattle described that when a volume (V) of a pure solvent mixes with a much larger volume of a solution of osmotic pressure (π), the free energy released is equal to πV . Pattle concluded that it is possible to use osmotic forces and selectively permeable membranes to obtain power by mixing freshwater and saltwater. However, this paper did not spark immediate further research interest as there were no subsequent articles on the subject for 20 years.

2.2. The 1970s

Following the 1973 oil crisis [19], oil prices spiked (shown by the line graph in Fig. 2) and interest in PRO was renewed. Several investigations of the technical and economic feasibility of PRO were published after 1973 [7,14–17,20,21] (represented by the bar graph in Fig. 2). After the first Kyoto meeting of 1997, the call for renewable power generation amplified and PRO research picked up again.

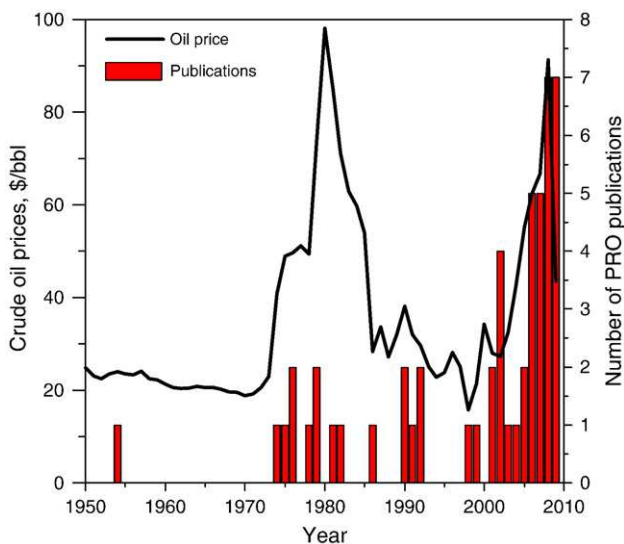


Fig. 2. Increased interest in PRO for power generation with increasing crude oil prices. Interest in PRO is indicated by the number of PRO peer-reviewed publications collected utilizing the ISI Web of Knowledge database. Annual crude oil prices are inflation-adjusted to the year 2010 and collected from Ref. [22].

Further, the more recent increase in PRO publications also coincided with the crude oil price spike of 2008. Both of these have led to the publication of over 20 PRO papers during the first decade of the new millennium.

In 1974, Norman [7] proposed the first diagram of an osmotic salination energy converter. In this diagram (Fig. 3), freshwater (with higher water chemical potential) permeates through a semipermeable membrane into a pressurized seawater chamber (with lower water chemical potential). The water that spills over the top of the column turns a waterwheel and powers a generator. The waterwheel configuration enables pressurization to occur simply due to the water column. This visually explicit diagram effectively shows the conversion of water chemical potential into hydrostatic potential. One year later, Loeb and Norman proposed the term “pressure retarded osmosis (PRO)” to be used for water salination with an osmotically driven membrane process [21].

The first experimental PRO results were published in 1976 by Loeb et al. [14]. In their investigation, hollow-fiber seawater RO membranes enclosed in a “minipermeator” (Permasep B-10) were tested. Pressurized brine flowed on the shell side of the bundle of hollow-fiber membranes and freshwater flowed through the bore. This study was closely followed by further experimental investigations of Loeb and Mehta [15–17]. These studies successfully proved the PRO concept but also revealed power outputs (from 1.56 to 3.27 W/m² using hypersaline draw solutions) that were far below the expected outputs based on the osmotic pressure difference across the membrane. Internal concentration polarization was introduced and was thought to have a strong adverse effect on the water permeation rate, and therefore, on the overall economics of power generation by PRO [16]; however, a comprehensive report on internal concentration polarization in osmotically driven membrane processes was not published until later [23]. Also, during this time Loeb and Mehta [15] proposed a model to predict flux and pressure in PRO. They theorized that two different water permeability constants exist, one driven by the hydrostatic pressure, and the other driven by the osmotic pressure. This theory was later further investigated by Seppälä et al. [24].

Another approach to PRO, the closed-loop osmotic heat engine (Fig. 1b), was patented by Loeb in 1975 [11]. The osmotic heat engine is a means of converting heat energy into mechanical work using engineered osmosis. In an osmotic heat engine, the working fluid

(i.e., water) permeates through a semipermeable membrane into a pressurized concentrated draw solution (e.g., an NaCl solution); the draw solution volume increases and flow is induced through a turbine to produce power. The draw solution is then separated from the working fluid by heating using a thermal process. Thus, the osmotic heat engine uses osmotic pressure to convert heat energy into mechanical work.

2.3. The 1980s

In the 1980s, the PRO research community expanded and papers from four different research groups were published [23,25–27]. In 1981, a theoretical and experimental study on the production of useful energy by PRO was published [25]. Results from the experiments showed that a power per unit membrane area (i.e., a power density) of 1.6 W/m² could be achieved. The authors believed that such power densities could justify the construction of a cost-competitive osmotic power plant. Also in 1981, Lee et al. [23] developed a model (that has since become the reference PRO performance model) that used results from FO and RO experiments to predict PRO performance. In this model, projected water flux and power density were evaluated with consideration of internal concentration polarization. The general equation Lee et al. used to describe water transport in PRO was:

$$J_w = A(\Delta\pi - \Delta P) \tag{1}$$

where J_w is the water flux, A is the water permeability coefficient of the membrane, $\Delta\pi$ is the osmotic pressure differential, and ΔP is the hydraulic pressure differential. In PRO, the power density is equal to the product of the water flux and the hydraulic pressure differential across the membrane:

$$W = J_w \Delta P = A(\Delta\pi - \Delta P)\Delta P \tag{2}$$

J_w and W as a function of ΔP are illustrated in Fig. 4, adapted from Lee et al. [23]. By differentiating Eq. (2) with respect to ΔP , it can be shown that W reaches a maximum (W_{max}) when $\Delta P = \Delta\pi/2$. Substituting this value for ΔP in Eq. (2) yields:

$$W_{max} = A \frac{(\Delta\pi)^2}{4} \tag{3}$$

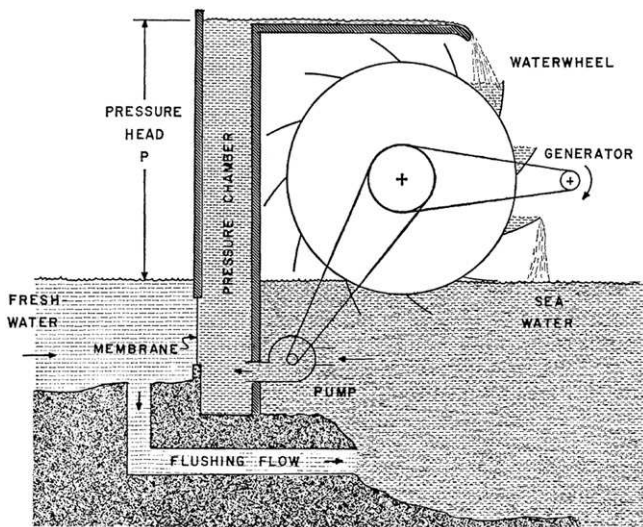


Fig. 3. Schematic diagram of an osmotic salination energy converter as proposed by Norman [7]. Freshwater permeates through a semipermeable membrane into a pressurized seawater chamber; the water that spills over the top of the column turns a waterwheel and powers a generator.

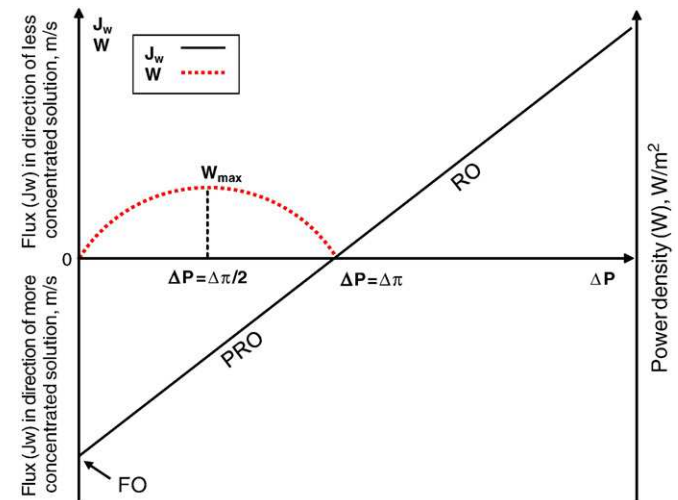


Fig. 4. Flux (J_w) and power density (W) for PRO as a function of applied pressure (ΔP). Magnitude and direction of flux at the FO point and in the RO region are also shown. Figure adapted from Ref. [23].

The PRO zone (where $\Delta P < \Delta \pi$) and W_{max} are shown in Fig. 4. Also in Fig. 4, the FO point (where $\Delta P = 0$), the flux reversal point (where $\Delta P = \Delta \pi$), and the RO zone (where $\Delta P > \Delta \pi$) are indicated.

Lee et al. [23] also studied concentration polarization, a phenomenon that can severely reduce the effective osmotic pressure difference across the membrane due to the accumulation or depletion of solutes near an interface. As a result of water crossing the membrane, the solute is concentrated on the feed side of the membrane surface and diluted on the permeate side of the membrane surface. Because the membranes used in PRO are typically asymmetric (comprised of a thin dense layer on top of a porous support layer), concentration polarization occurs externally on the dense layer side and internally in the support layer side. Both internal and external concentration polarization reduce the effective osmotic pressure difference across the membrane; however, internal concentration polarization was expected to be more severe (recently proven by Achilli et al. [28]). Disregarding external concentration polarization, Lee et al. improved their model to include the effect of internal concentration polarization on water flux in PRO applications:

$$J_w = A \left[\pi_D \frac{1 - \frac{C_f}{C_D} \exp(J_w K)}{1 + \frac{B}{J_w} [\exp(J_w K) - 1]} - \Delta P \right] \quad (4)$$

where π_D is the osmotic pressure of the draw solution (e.g., seawater), C_f is the salt concentration of the feed solution (e.g., freshwater), C_D is the salt concentration of the draw solution, and B is the salt permeability coefficient of the membrane. The solute resistivity for diffusion within the porous support layer (K) is defined by:

$$K = \frac{t\tau}{D\varepsilon} \quad (5)$$

where t , τ , and ε are the thickness, tortuosity, and porosity of the support layer, respectively, and D is the diffusion coefficient of the solute in the draw solution. The term $t\tau/\varepsilon$ is often referred to as the membrane structural parameter (S). K can be used to determine the influence of internal concentration polarization on water flux.

Substituting Eq. (4) into Eq. (2), the power density becomes:

$$W = J_w \Delta P = A \left[\pi_D \frac{1 - \frac{C_f}{C_D} \exp(J_w K)}{1 + \frac{B}{J_w} [\exp(J_w K) - 1]} - \Delta P \right] \Delta P \quad (6)$$

After Eq. (4) is solved numerically to determine J_w , Eq. (6) can be solved algebraically to determine W as a function of ΔP .

In the same publication, Lee et al. [23] validated their model under FO and RO conditions but not under PRO conditions because of difficulties they had in building a laboratory-scale PRO membrane module. They concluded that internal concentration polarization markedly lowers water flux under PRO conditions and, after performing a simple economic analysis, that membranes with significantly improved performance would be necessary for PRO to become economically feasible for power generation. The work from Lee et al. inspired several investigations – albeit over two decades later – on concentration polarization phenomena in osmotically driven membrane processes by McCutcheon and Elimelech [29,30]. Also two decades later, their model was validated under PRO conditions in a study by Achilli et al. [28].

Returning to the 1980s, Mehta continued to test the then state-of-the-art RO membranes for PRO applications [26]. Results showed low water flux and power densities that were hindered by severe internal concentration polarization. In accordance with Lee et al.'s [23] findings, Mehta concluded that membranes developed for RO applications are not suitable for PRO applications, and if the objective of economical energy generation using salinity gradient systems is to be realized, it is absolutely necessary to develop and optimize the performance of semipermeable PRO membranes. The lack of mem-

branes unique to PRO was also acknowledged in another investigation [27].

2.4. The 1990s

In 1990, Loeb et al. [31] conducted a comparative study on the mechanical efficiency of several theoretical PRO plant configurations. A continuous-flow terrestrial PRO facility, a continuous-flow underground PRO facility, and an alternating-flow terrestrial PRO facility were evaluated to determine the most efficient way to keep the PRO saltwater circuit under pressure. It was found that the alternating-flow terrestrial PRO plant had higher efficiency but required the use of two pressure vessels in addition to the usual PRO equipment. The requirement for pressurization at a cost that wouldn't outweigh the value of the energy generated was a problem that would not be solved until the next decade when pressure exchangers would be developed [18].

In the same year, Reali et al. [32] computed the salt concentration profiles in the porous support layer of anisotropic membranes under PRO conditions using numerical techniques. This study highlighted the role of membrane characteristics (water permeability coefficient (A), salt permeation coefficient (B), thickness of the porous support layer (t), and effective salt diffusivity (D)) on the water and salt permeation through the membrane. The salt concentration profiles in the porous support layer of anisotropic membranes were also determined analytically in a later study [33]. The decade ended with Loeb [10] revitalizing the interest in PRO by asking if PRO energy production at the Dead Sea would be a challenge or a chimera. Loeb's paper concluded that, depending on system configurations, electrical energy could be produced at a cost ranging from 0.058 to 0.07 \$/kWh – costs comparable to the average retail electricity price in the United States at that time (0.067 \$/kWh) [34].

2.5. The 2000s

In the 2000s, Loeb [8,9] investigated the possibilities of PRO application at the Great Salt Lake and found that, at this location, electrical energy would be produced at a cost of 0.15 \$/kWh. In 2002, Loeb [18] was the first to acknowledge the importance of pressure exchangers, originally developed for RO applications, in enabling cost effective PRO systems and published a paper that described an improved plant schematic incorporating this device. The introduction of pressure exchangers significantly simplifies PRO plant design and eliminates a very large parasitic consumption of power [18]. The same configuration would eventually be utilized by the Norwegian national power company (Statkraft) in their first prototype installation [35].

Several other research groups have also taken part in the recent history of PRO. Statkraft and the Foundation for Scientific and Industrial Research at the Norwegian Institute of Technology (SINTEF) published two papers highlighting the regional and global potential of PRO [6,36]; this work was a prelude to their more notable contribution to the development of PRO, which began near the end of the decade. In the meantime, Seppälä et al. published several papers, ranging from modeling [37,38], to experiments [24,39], and system configurations [40]. Seppälä et al. challenged the assumption that the internal concentration polarization theory (which assumes that when all concentration boundary layers are accounted for, the true response to increasing osmotic pressure is linear [23,41]) explains the apparent non-linear proportionality of water and solute transport versus osmotic and hydrostatic pressure [24]. They proposed and experimentally verified a two-coefficient linear equation to describe water and solute transport through an osmotic membrane. Seppälä et al. concluded that although it is uncertain whether or not dependence on osmotic pressure is linear or non-linear, there is no proof that the apparent non-linearity is caused by concentration polarization phenomena. This theory did not receive further attention and currently, internal concentration polarization is widely accepted as the cause for reduced water flux in osmotically driven membrane processes [42].

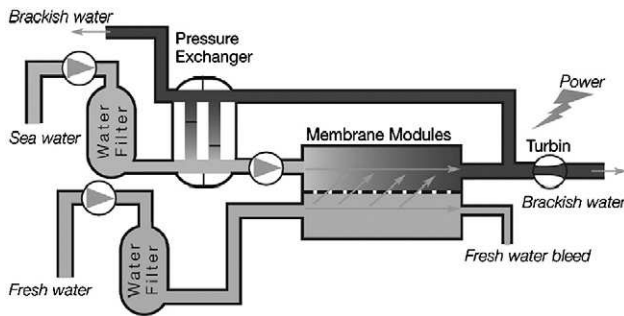


Fig. 5. Schematic diagram of the first prototype PRO installation opened in Norway on November 24, 2009 and designed and operated by Statkraft (Oslo, Norway) [35].

Over 30 years after it was first patented, the osmotic heat engine concept was revitalized in a publication of McGinnis et al. [12]. McGinnis et al. proposed the use of low-grade heat, allowing for the potential recovery of power from waste heat at low cost. McGinnis et al. [12] also proposed the use of a novel thermolytic draw solution based on ammonia-carbon dioxide in order to increase the thermal efficiency of the system. Their model results indicate that, depending on operating conditions, power density may exceed 250 W/m^2 , and also that the thermal efficiency of the engine could be 16% of Carnot efficiency [12].

In the late 2000s, as part of an experimental and theoretical investigation into PRO, Achilli et al. [28] expanded the model developed by Lee et al. [23] by incorporating external concentration polarization in the model. Experimental water flux and power density results were obtained utilizing a custom-made laboratory-scale membrane module with a flat-sheet cellulose triacetate FO membrane (HTI, Scottsdale, AZ). The results closely matched model predictions and power densities exceeding 5 W/m^2 were observed. Power density was found to be substantially reduced due to internal concentration polarization. Sensitivity analyses were performed to evaluate the influence of membrane characteristics on power density. Results indicate that to increase power density by at least one order of magnitude, a combined substantial increase in membrane water permeability and decrease in support layer resistivity must be achieved. If a PRO membrane could be developed to have the permeability of a nanofiltration membrane (yet with low salt permeability) and a support layer with a tenth of the thickness of current membranes (yet with high structural strength), then power densities of 30 W/m^2 could be achieved.

Also in the late 2000s, Statkraft and SINTEF publicized more of their building body of work on PRO through several papers ranging from experimental investigations using non-commercial PRO membranes [35,43] to examining desired characteristics for PRO membranes [44]. They also established that to make PRO profitable in the Norwegian energy market, the power density of the membrane should be at least in the range of $4\text{--}6 \text{ W/m}^2$ [43].

And on November 24, 2009, more than 30 years after the first PRO experiment conducted by Sidney Loeb, Statkraft opened the first prototype PRO installation in Norway [45]. The plant configuration (Fig. 5) [35], follows the original plant schematic proposed by Loeb [18]. Water from a low salinity feed solution (e.g., freshwater) permeates through a semipermeable membrane into a pressurized, high salinity brine/draw solution (e.g., seawater); power is obtained by depressurizing the water that crosses the membrane through a hydroturbine. The prototype PRO installation, built by Statkraft, is designed to generate 10 kW of power; Statkraft plans to build a full-scale 25 MW osmotic power plant by 2015.

3. Summary and conclusions

Despite the relatively large quantity of published material on PRO, there are minimal experimental data on power density. Fig. 6 shows a timeline of power density data acquired over the past four decades. It can be seen that there are two general time periods when experimental data were collected – the 1970s and the 2000s. The data were divided into two sets, the set with the darker blue symbols represents the maximum experimental power density achieved with a draw solution of approximately seawater concentration, the set with the lighter orange symbols represents the maximum experimental power density achieved with a draw solution having a concentration higher than seawater. As expected, for experiments conducted in the same general time period, using hypersaline draw solutions leads to higher power density. The hypersaline draw solutions were used to demonstrate the feasibility of PRO in specific locations such as the Dead Sea and the Great Salt Lake [16,17]. Over the whole timeline, it can be seen that the more recent power density values are up to 3 times higher than earlier results, likely due to the improved membranes and membrane modules used in the more recent investigations.

Loeb et al. [14], Loeb and Mehta [15], and Mehta and Loeb [16,17] initially used hydrophobic polymeric membranes developed for RO applications resulting in power outputs far below expected outputs.

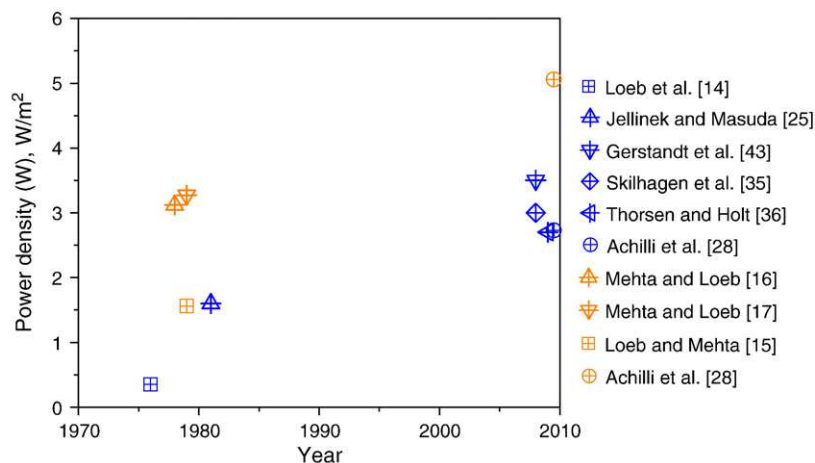


Fig. 6. Comparison of experimental PRO power density results over the past four decades. The darker (blue) symbols represent the maximum experimental power density achieved with a draw solution having approximately seawater concentration and the lighter (orange) symbols represent the maximum experimental power density achieved with draw solutions having concentrations higher than seawater.

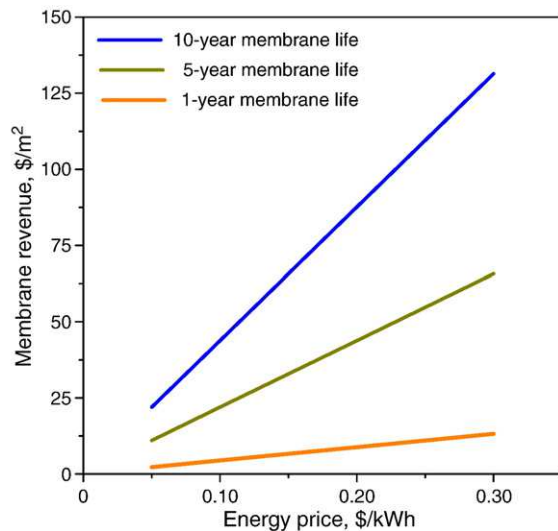


Fig. 7. Sensitivity analysis for membrane revenue as a function of energy price and membrane life. Membrane revenue is calculated as the product between the power density and the energy price. A power density of 5 W/m² is assumed.

More recent, seawater RO membranes have also been found unsuitable for PRO applications due to their hydrophobicity and thick support layer [42,46]. Most recently (in the past few years), cellulose acetate FO membranes [28,43,47] and prototype thin-film composite PRO membranes are being used to improve power density.

Furthermore, it has been established that RO modules are not suitable for PRO applications because of the intrinsic differences between the two processes. PRO modules must be designed to maximize fluid circulation on both sides of the membrane while RO modules only need circulation on the feed side of the membrane [42]. The hollow-fiber modules used in the early experiments [14–17] did not allow for high cross-flow velocity on both sides of the membrane and resulted in poor performance [29]. In recent bench-scale experiments, flat-sheet modules specifically designed for PRO experiments [28,44] and spiral wound modules (Hydrowell®, HTI, Scottsdale, AZ) [47] are being used to improve power density. At Statkraft's prototype facility, modified spiral wound modules are being used. Eventually, multi-dimensional water and solute transport mechanisms in full-scale modules need to be evaluated and derived. This would assist the design of optimized membrane modules for full-scale PRO applications.

To evaluate the current economic feasibility of PRO, estimates of PRO facility revenue per membrane area per year can be determined using:

$$\frac{\text{Revenue}}{\text{Membrane area} \cdot \text{year}} = \text{Power density} \cdot \text{Energy price} \quad (7)$$

Considering an achievable power density of 5 W/m² and a current energy price of 0.10 \$/kWh [48], a facility revenue of 4.4 dollars per square meter of membrane per year (\$/m²y) would result. In order to better understand this, Fig. 7 illustrates the facility revenue generated by each square meter of membrane (\$/m²) as a function of energy price, considering different membrane lifetimes, and based on 5 W/m² of power density. As would be expected, revenues increase with increasing energy price and membrane life. At current energy prices and achievable power densities, for an expected membrane life of 5 years [49], the membrane revenue is 22 \$/m². This value is at the lowest of the range of the current estimated bulk cost of membranes per square meter, 20–40 \$/m² (estimation based on RO-type membranes) [50]. Thus, currently PRO does not appear to be able to produce energy at a competitive cost. In order for PRO to be more competitive, a substantial increase in power density, decrease in

membrane cost, or increase in membrane life (or some combination thereof) must be achieved. Furthermore, a subsidized renewable energy market, such as currently exists in the European Union, may be needed to sustain the continuing development of this technology until membrane and module technology gaps can be filled.

Although many investigators would suggest there still exists a lack of theoretical and experimental investigations to ensure the success of scaled-up PRO, several news articles appearing in specialized and main-stream press during 2009 [51–54] proclaim that PRO is closer than ever to being a viable alternative for renewable energy production. It will be interesting to see if the expedited design and construction of the prototype installation in Norway and the subsequent media interest will inspire competitive commercialization of membranes and modules and enable PRO to be a valid contributor to renewable energy production.

Acknowledgments

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