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Biodesalination – On harnessing the potential of Nature’s desalination processes

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Abstract

Water scarcity is now one of the major global crises, which has affected many aspects of human health, industrial development and ecosystem stability. To overcome this issue, water desalination has been employed. It is a process to remove salt and other minerals from saline water, and it covers a variety of approaches from traditional distillation to well-established reverse osmosis. Although current water desalination methods can effectively provide fresh water, they are becoming increasingly controversial due to their adverse environmental impacts including high energy intensity and high concentrated brine waste. For millions of years, microorganisms, the masters of adaptation, have survived on earth without neither the excessive use of energy and resources nor compromising their ambient environment. This has encouraged scientists to study the possibility of using biological processes for seawater desalination and the field has been exponentially growing ever since. Here, the term of *biodesalination* is offered to cover all of the techniques which have root in biology for producing fresh water from saline solution. In addition to reviewing and categorizing *biodesalination* processes for the first time, this review also reveals unexplored research areas in *biodesalination* having a potential to be used in water treatment.

Keywords: Biodesalination, Seawater Desalination, Living Organisms, Biomimetic membrane and Bioinspired membrane

Abbreviations:

MSF, Multistage flash distillation; MED, multiple effect distillation; VCD, vapor compression distillation; RO, reverse osmosis; FO, forward osmosis; PMDCs, photosynthetic microbial desalination cells; LCST, lower critical solution temperature; BCPs, block copolymers; MD, membrane distillation; AEM, anion exchange membrane; CEM, cation exchange membrane; MFC, microbial fuel cell; SWRO, seawater reverse osmosis systems; PSBMA, Poly(sulfobetainemethacrylate); MPDSAH, [3-(methacryloylamino)propyl]-dimethyl(3-sulfopropyl) ammonium hydroxide; PSBMA,

Poly(sulfobetaine methacrylate); MEDSAH, [2-(methacryloyloxy)ethyl]dimethyl-(3-sulfopropyl) ammonium hydroxide; MPC, 2-(methacryloyloxy)ethyl phosphorylcholine; MPDSA, [3-(methacryloylamino)propyl]-dimethyl (3-sulfopropyl) ammonium hydroxide inner salt; CBMA, Carboxybetaine methacrylate; SBMA, Sulfobetaine methacrylate; MEDSAH, [2-(methacryloyloxy)ethyl]dimethyl-(3-sulfopropyl)ammonium hydroxide; DMMSA, Sulfobetaine N,N-dimethyl-N-methacryloxyethyl-N-(3-sulfopropyl); DMMSA-BMA, N-methacryloxyethyl-N-(3-sulfopropyl)-co-hydrophobic butyl methacrylate; PVDF, Poly(vinylidene fluoride); PP, Polypropylene; PTFE, Poly(tetrafluoroethylene); PES, Polyethersulfone; PSF, Polysulfone; PVDF, Poly(vinylidene fluoride), SA, sodium acrylate; NIPAM, N-isopropylacrylamide, PSA-NIPAM, sodium acrylate-co-N-isopropylacrylamide and COD, chemical oxygen demand

1. Introduction

Although three quarters of Earth's surface is covered by water, more than 97% of the water is in saline water form that cannot be directly used for either drinking or agriculture. About 0.5% of the remaining 3% is available as freshwater but not evenly distributed throughout the world [17]. As a result, many countries especially those located in Middle East and North of Africa are facing freshwater scarcity [67]. Technological advancements in seawater desalination industry during last decades have provided various solutions to this issue. Today, there are several well-known physicochemical approaches for the seawater desalination e.g. multistage flash distillation (MSF), multiple effect distillation (MED), vapor compression distillation (VCD), reverse osmosis (RO), forward osmosis (FO) and electrodialysis [2]. Among these energy intensive technologies, well-designed and established state-of-the-art RO plants have managed to reduce the energy demands from above 15 kWh.m^{-3} down to about 2.2 kWh.m^{-3} for the desalination of standard seawater (35000 ppm NaCl, with 50% recovery)[78]. However, it is still 1.5 to 2.0 times greater than the minimum theoretical energy requirement based on the thermodynamic principles. Therefore, there is still a continuing quest for improved desalination processes with even lower energy demands. On the other hand, the RO desalination like any other industrial process has adverse environmental impact which needs to be considered, assessed and mitigated. These environmental impacts consist of impingement and entrainment during water intake, which results in a new source of mortality to the marine environment and disposal of the concentrated brine with salt concentration twice as much as the feed water [43]. In addition, the emission of air pollutants and greenhouse gases from the thermoelectric energy used by RO desalination plants is another adverse impact which exacerbates climate change. A current RO desalination plants with an average energy consumption of $3\text{-}4 \text{ kWh/m}^3$ produces 1.4 to 1.8 kg CO_2 per tones of produced water [18]. Even when the desalination energy drops to the theoretical minimum level, the carbon footprint is still substantial (see Table 1). Other issues with RO process are membrane fouling and deterioration, which result in

extensive use of membrane cleaning chemicals and membrane replacement. Chemical cleaning (including both pre-treatment and cleaning process during operation) and membrane replacement account for about 7 and 5% of the overall life cycle cost of an seawater reverse osmosis systems (SWRO) desalination plant respectively [22]. When compared with the artificial desalination process, the fascinating biological systems perform highly efficient separations in living organisms without compromising their surrounding environments. This provided enough motivation for biologists and engineers to conduct a significant amount of research to identify robust new methods of water desalination with lower energy consumption, reduced cost and minimized adverse environmental impacts. As presented in Table 1, the innovative *biodesalination* approaches such as the photosynthetic microbial desalination cells (PMDCs) can not only desalinate saline water but also are able to generate electricity without compromising ambient environment [37]. The new interdisciplinary field of *biodesalination* has resulted in several research papers introducing new potential desalinating approaches and also addressing different aspects of this field. Despite the large number of publications, there is no comprehensive overview to categorize and define the nature inspired novel desalination process. Amezaga *et al.* [3] and Minas *et al.* [62] referred to *biodesalination* as the production of fresh water using bacteria whereas *biodesalination* was described elsewhere as removing sulfate from highly polluted mine water in a bioreactor [106]. Biologically based fresh water production has also been addressed under different terms such as photosynthetic microbial desalination [37] or microbial desalination cell technology [9]. The large amount of publications and unclear definitions indicate the “*biodesalination*” has now reached the stage where a comprehensive review is required to cover the latest progress, understand knowledge gaps and most importantly define the term of biodesalination. This review paper is therefore to serve these purposes. Here, we propose the term *biodesalination* as utilizing living organisms or biological elements directly or indirectly, mimicking their structures and mechanisms, borrowing concepts or inspiration from their desalination mechanisms for the production

of sustainable fresh water. As presented in Figure 1, *biodesalination* processes can be classified into three main subfields of bioinspiration, biomimetics and direct use of living organisms for desalination.

Table 1. Energy requirements, environmental impact and energy gain potential for different desalination techniques [18, 37, 61, 84]

Desalination technology	Energy requirement (kWh.m ⁻³)	CO ₂ emissions (kg CO ₂ .m ⁻³)	Energy gain potential (kWh.m ³)
Thermal evaporation ^a			
MSF (Multi-Stage Flash)	72.94 - 88.33	20.4-25.3	~0
MED (Multi-Effect Distillation)	43.17 - 66.11	11.8-17.6	~0
VC (Vapor Compression)	7.5 - 13.00	-	~0
Membrane separation			
RO (Reverse Osmosis)	4.00-8.00	1.4-2.79	~0
Biodesalination			
PMDCs (Photosynthetic microbial desalination cells)	~0 ^b	~0 ^b	4.21

^a the energy requirement for MSF, MED and VC is combined energy demand (i.e. thermal and electrical energy) of non-cogeneration units which use natural gas.

^b the zero values are associated with the desalination process governed by microorganisms and the required energy and CO₂ emission for capital cost and operation are not considered here.

Figure 1. Classification of biodesalination

In biomimicry for desalination, the biological systems or their elements are considered as prototypes to introduce potential advanced innovative desalination technologies which desalinate seawater in an efficient and sustainable manner. Biomimetics introduces an engineering solution by focusing on the basic science and exploring the fundamental principles of biological systems [110]. However, it poses difficulty in technological implementation. Bioinspiration comes into the course when a bridge between basic science and applied engineering is necessary for technological implementation [101]. Therefore, bioinspired desalination processes are based on a concept borrowed from a biological system, function, process or an element, which are not, quite often, resemble to the biological prototypes. Usually a concept development process is required to assemble the borrowed idea into a

desalination process. Comparing with bioinspiration and biomimicry for desalination, the idea of direct use of living organisms such as aquatic plants, algae, zooplanktons and bacteria for seawater desalination has recently been studied as an attractive option for seawater desalination. The review first discusses the mechanisms by which Mother Nature desalinates saline water, followed by the bioinspired desalination processes based on borrowing concepts from biological systems. The hot topic of biomimetic membranes for desalination will be discussed as an emerging sustainable approach in *biodesalination*. Our final focus will be on the feasibility of using of living organisms for seawater desalination.

2 Desalination mechanism in nature

Sodium and chloride are the most abundant dissolved ions in saline water and lands [17], which play important physiological roles in most living organisms. However, high sodium concentration within cells could lead to damage of the proteins and interfere the metabolism [23]. Therefore, living organisms have employed different strategies to balance sodium concentration [85], which included 1) blockage of the Na^+ from entering into the cytosol (salt resistance) and 2) reduction of the Na^+ concentration in the cytosol (salt tolerance) via an intricate transport systems through cellular membrane [86].

2.1 Transport system in the cell membrane of living organisms

As shown in Figure 2, there are a variety of transporters in the plasma membrane of living organisms. Although these transporters may be classified based on different characters, they mainly contain three groups: pumps, carriers, and channels with many subgroups in each separated group [89]. The main function of these transporters is to allow the passage of water and nutrients, and to maintain the chemical balance within the cell [6]. The most important character of these transportes is selectivity feature with different affinities: certain carrier has the capability to recognize specific ions or molecules within a complex solution and facilitate their transport. For example potassium pumps can recognize

(K^+) from (Na^+) even though both of them have similar physicochemical properties [97]. This is one of the most basic and optimized prototypes in *biodesalination*, and this concept has been applied for membrane preparation: Aquaporin as a water channel has been applied for biomimetic membrane preparation. This aspect will be discussed in Section 4 [1].

Figure 2. Schematic representation of govern carriers presented in plant cell membranes. AQ.

Aquaporin, NSCC: Non selective cation chanell.

2.2 *Bacteria's mechanisms for desalination*

For the unicellular creatures like bacteria, the osmosis pressure need to be controlled when living in saline water condition. Therefore, the most involved natural mechanisms in halophilic bacteria are osmoregulation. For the bacteria adapted in salt water conditions, K^+ gradient is maintained by a combination of Na^+/H^+ antiporter and K^+ uniporters [24]. A more flexible strategy is found in moderately halophilic bacteria that existing in a wide range of salinities. This strategy relies on the

accumulation of high concentrations of organic compatible solutes. Compatible solutes are small compounds which are highly soluble in water and do not interfere with the cellular metabolism [36]. In response to an osmotic stress, bacteria mainly accumulates organic compounds like sugars, polyols, amino acids and amino acid derivatives [65]. For example, glycine betaine transporter accumulates the osmoprotectant glycine betaine in *Methanohalophilus portucalensis*. In addition, some moderate halophiles, e.g. *Halobacillus halophilus*, use a unique hybrid strategy to adapt with changes in salinities of their environment. The hypothesis of this hybrid strategy is based on the fact that *H. halophilus* amasses compatible solutes as well as chloride in the cytoplasm. Halophilic bacteria physiologically uses Na^+ for electrochemical gradient generation and to improve the membrane stability. Na^+/H^+ antiporters have highly pH dependent activities and functions in the regulation of the internal pH. It also uses the H^+ gradient to expel Na^+ from the cell. The generalized transport reaction catalyzed by Na^+/H^+ antiporter is: $\text{Na}^+ (\text{in}) + 2\text{H}^+ (\text{out}) \rightleftharpoons \text{Na}^+ (\text{out}) + 2\text{H}^+ (\text{in})$ [68].

2.3 *Plant's mechanisms for desalination*

Extremely high salinity is a serious problem for plants as it limits water absorption, denaturates enzymes, and degrades soil structure. Therefore, plants rely on sophisticated mechanisms to resist saline conditions [85]. These mechanisms are generally classified into salt-secretion (e.g. *Avicennia* sp.), salt-accumulating (*Atriplex* sp.), and salt excluding [21, 54]. The salt-secretion mechanism is based on exporting salt via excreting organs in plant leaves (Figure 3A) [40, 63]. Salt accumulation is observed to accure on the leaves of some halophytes (Figure 3B) that desalinate soil by compartmentalization of Na^+ and Cl^- in their above ground tissues. Salt-excluding mechanism is based on the blockage of salt [34] using specific features such as antiporters. Micro green algae *Dunaliella salina* is a typical example of salt-excluding plants because it can survive in as high as 3M NaCl [91] due to its extreme salt blocking ability. This ability is due to the presence of a Na^+/H^+ antiporter on the vacuole membrane (tonoplast) and H^+/ATPase pumps on the plasma membrane.

Figure 3. Salt tolerance mechanisms in plants: crystal formation on the leaf of *Avicennia germinans* (courtesy of Dr. Ulf Mehlig, <http://www.ulf-mehlig.de/>) (A) and *Atriplex* leaf (courtesy of Dr. Amir Abas Minaiefar) (B)

2.4 *Animal's mechanisms for desalination*

Sea fishes (e.g. *Heterodontus portusjacksoni*), sea birds (e.g. *Larus marinus*) and some reptiles (e.g. *Amblyrhynchus cristatus*) use kidney and salt glands to desalinate saline water [83]. There are many secretory tubules in salt glands with diameter and length that vary depending on the salt uptake of the specie [19]. Salt gland allows reptiles and sea birds to efficiently secrete out salinity. These animals use a complex set of reactions which includes chloride ions forming an electrical gradient, allowing sodium to pass through the tight junctions of secretory cells into the tubular lumen along with a negligible amount of water [26].

2.5 *Human's mechanisms for desalination*

In human, the levels of salts concentrations in blood are controlled by kidneys through the proximal tubule in the nephron [23]. Desalination and blood filtration process comprise three main stages of (a) glomerular filtration (ultrafiltration process) to retain large proteins (b) selective or tubular reabsorption

of valuable components such as nutrients and (c) tubular secretion of undesirable or excessive substances. In addition to kidneys, sweat glands also help the body to remove the unnecessary salts including sodium ions [56].

3 Bioinspiration for desalination

3.1 Hydrogel-driven forward osmosis process

Osmosis is a naturally occurring process to transport water across a semi-permeable membrane from a lower solute concentration region to the higher side. In living organisms, osmosis plays a vital role in maintaining cells' normal cytosolic osmolarity and regulating cell volume via shrinking or swelling in hypertonic or hypotonic solutions, respectively. When a mammal cell is placed in contact with a hypotonic solution, the cell swells and absolute pressure increases until the cell membrane ruptures. Plants also take advantage of osmosis to absorb soil water through their root hairs. However, watering plants with saline water wilts them in a plasmolysis process as the soil around the roots could become hypertonic. Principles of direct osmosis in biological membrane phenomenon have been utilized in downstream processing, i.e. Forward Osmosis (FO), to desalinate seawater. As presented in Figure 4, in a typical FO desalination process, a draw (osmotic) agent is used to induce osmotic pressure gradient across a membrane to drive water molecules pass through the membrane from seawater (low osmotic pressure region) to the draw agent (high osmotic pressure region). In the next stage, the draw agents are recovered from the desalinated water to continue the FO desalination cycle [80]. As presented in Figure 5a, red-onion cells undergo a shrinking and swelling cycle when exposed to saline and distilled water. In an inspired approach, *Höpfner et. al* [28] presented that ionic hydrogel particles made from poly(acrylic acid) with three dimensional cross-linked network are able to desalinate saline water by undergoing swelling and shrinkage cycles. During the swelling stage, the ionic groups fixed on the polymer strands of the network shield off the ions in the seawater and allow only water molecules to pass through. On the other hand, in the shrinkage stage a hydraulic pressure is used to

squeeze out the absorbed water (Figure 5b). However, the swelling/shrinkage approach is still in its infancy as its energy demand is significantly higher than that of reverse osmosis. According to Li *et al.* [45], at room temperature a pressure of 3MP can only recover 3.2% water from a PSA-NIPAM swollen hydrogel with 80% water content. To overcome the hydrogel dewatering energy demanding issue, thermo-responsive polymer hydrogel is introduced [45, 46, 77, 78, 80]. The thermo-responsive polymer hydrogel particles are placed on a semi-permeable membrane to provide osmotic pressure and only draw water molecules from a saline solution. As presented in Figure 5b, the hydrogel network experiences a phase change at its lower critical solution temperature (LCST) and releases the absorbed water. Razmjou *et al.* [78] recently studied the feasibility of using bifunctional polymer hydrogel layers as draw agents for continuous production of fresh water using solar energy. Their thermodynamic analysis showed that the minimum required energy for the recovery of 1 g swollen hydrogel is in the range of 1.12–3.57 kWh/m³, which shows that such a draw agent has a great potential to be used in the FO desalination process. They also proposed (Figure 6) a conceptual design of semibatch hydrogel-driven FO process using solar energy for the continuous production of pure water, which is a significant step forward toward the commercial implementation of hydrogel driven FO system for seawater desalination.

One of the main challenges in FO desalination process is flux reduction due to internal concentration polarization (ICP) which is the accumulation of the retained salts and other constituents adjacent to the both membrane sides. Since hydrogels particles are uniform networks, the concentration polarization on permeate side is negligible. Although forward osmosis technology has been widely studied for the desalination of brackish and seawater, their application in industrial scale is limited due to the lack of highly efficient and stable FO membranes, as well as the difficulty in draw solution recycle [111]. A better draw solution with clear commercial potential and lower ICP tendency is worthy of further

investigation. The biological materials with their unique separation properties may serve as an inspiration for the development of novel FO desalination processes.

Figure 4. A typical FO desalination process

Figure 5. Shrinking/swelling process of A) red-onion cells and B) thermo-responsive Fe_2O_3 nanocomposite ionic hydrogel based on the procedure introduced by Razmjou *et al.* ^[77]. The nanocomposite is able to desalinate saline water by undergoing swelling and shrinkage cycles.

Figure 6. A conceptual design of semibatch hydrogel-driven FO process using solar energy for the continuous production of pure water. Reprinted with permission from[78]. Copyright 2014 American Chemical Society

3.2 *Super-hydrophobic modification for membrane distillation*

Desalination by membrane distillation (MD) has recently gained significantly more attention as it could desalinate seawater at lower temperature (40-90°C) than the boiling point of water. In MD, a temperature gradient provides a vapor pressure difference across the porous hydrophobic membrane to transport water molecules from a high temperature feed (seawater) to a low temperature permeate side (desalinated water). Commercial implementation of MD is limited by fouling, pore wetting phenomenon, conduction heat loss as well as temperature and concentration polarization at the membrane interface [76]. Hierarchical structures with multilevel roughness of super-hydrophobic

surfaces in nature such as lotus and rice leaves (Figure 7 a) have inspired researchers to construct artificial super-hydrophobic surfaces for various applications [11, 16, 20, 42, 51, 52, 98]. Razmjou *et al.* proposed that the aforementioned obstacles of MD commercialization can be minimized by obtaining super-hydrophobic membrane surfaces with water contact angle greater than 150° [76]. It has been demonstrated that the fluorosilanization of TiO_2 nanocomposite PVDF membranes can lead to a nature inspired hierarchical nanostructure with a low surface free energy (Figure 7b), which is essential in designing super-hydrophobic surfaces. Meng *et al.* studied the effect of using templating agents on creating hierarchical structures and super-hydrophobic fluorosilanized TiO_2 nanocomposite PVDF membranes [60]. They found that porous, multi-level microstructures can be tuned by using different templating agents which ensured improved surface hydrophobicity when compared with membrane prepared without template agent [60]. Inspired by the micro/nanoscale hierarchical structures of lotus leaf and silver ragwort leaf, Wang *et al.* created a bioinspired superhydrophobic electrospun fibrous surface for desalination via direct contact membrane distillation (Figure 7c-e) [48]. Their results showed that the bioinspired membrane achieved a high water flux of around $105 \text{ L m}^{-2} \text{ h}^{-1}$ with 20 g L^{-1} NaCl concentration, which is around 5 fold higher than those of typical commercial PVDF MD membranes. Superhydrophobicity can also be imparted to PVDF membrane surfaces by other techniques like electro-spinning [50] and CF_4 plasma [105] surface modification technologies. Liao *et al.* prepared a superhydrophobic membrane with hierarchical surface structure by electro-spinning followed by surface modification. The resultant membrane achieved a high and stable water flux of $31.6 \text{ L m}^{-2} \text{ h}^{-1}$ using a 3.5 wt % NaCl as the feed solution while temperature difference across membrane was 40°C [50]. Yang *et al.* showed that the CF_4 plasma-assisted surface superhydrophobic modification could improve MD performance: the water flux was enhanced by about 30% from around 17 to $24.5 \text{ L m}^{-2} \text{ h}^{-1}$ [105].

Although membrane superhydrophobic modifications for MD based desalination has recently gained significant attention lately, the long-term performance of superhydrophobic MD process has not been fully understood. One of the major obstacles for commercial implementation of superhydrophobic MD process is the instability of the superhydrophobic layer. External perturbation and stimuli such as pressure or vibration can destabilize the layer and damage superhydrophobicity by filling the spaces between asperities. This may lead to an irreversible conversion of the superhydrophobic layer into a hydrophobic or even hydrophilic layer; a transition phenomenon from Cassie-Baxter state into the Wenzel state. These scale-dependent mechanisms by which the destabilization occurs are the condensation and accumulation of nano droplets, capillary waves and surface inhomogeneity. In order to preserve the stability of the surface, it is suggested that the surface should be designed such that it can resist these scale dependent instabilities. Therefore, the design of topographical structure of membranes with high or optimized geometries could be considered as a promising strategy to improve the stability of the modified surface. This will further guarantee the long-term stable performance of the MD desalination. This critical progress should be accompanied by the latest development of micron/nano-fabrication techniques which can be greatly inspired by the properties of surfaces found in natural systems. Those systems that exhibit a huge diversity of complex surfaces at both structural and functional levels, which can be considered as a prototype of superhydrophobic modifications of membranes for MD desalination process.

The practical application of superhydrophobic MD systems for real seawater as a feed has not also been fully investigated to date. Seawater is a complex environment, which contains a variety of organic, inorganic and biological moieties. The interaction of these components with the membrane surface needs to be investigated, particularly with regards to the biological interactions which could lead to the biological fouling (biofilm formation) and water flux reduction.

Figure 7. SEM images of A) lotus leaf surfaces shows the hierarchical structure. Reprinted with permission from^[70]. Copyright 2014 American Chemical Society, B) the bio-inspired hierarchical multi-level superhydrophobic PVDF membrane^[76] and C) schematics of a membrane distillation cell with dual-biomimetic nanofibrous membranes with a nanopapillae, nanoporous, and microgrooved surface morphology that originated from mimicking the lotus and silver ragwort leaf. Enlarged image of D) nanopapillae (50–80 nm) and nanopores (about 20 nm) and E) microgrooves along the fiber axis. Reprinted with permission from^[48]. Copyright 2014 ACS

3.3 *Bioinspired antifouling coatings*

Desalination by the pressure driven membrane processes¹ (i.e. reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF) and microfiltration (MF)) is currently one of the main approaches to address the issue of fresh water scarcity. However, one of its main drawback is fouling which is referred to membrane pores blockage during filtration process as a result of sieving and adsorption of matters onto the membrane surface or within its pores [81]. Fouling leads to the reduction of performance and eventually to the replacement of the membrane module. Different types of membrane fouling have been introduced including inorganic fouling or scaling, colloidal fouling, organic fouling, and biofouling [55]. Among them membrane biofouling due to the formation of biofilm has been regarded as the most serious issue. Biofouling refers to the formation of biofilms through the dynamic process of microbial colonization and growth on the membrane surfaces, which occurs through the six sequential stages of transport, deposition, adhesion, exopolymer production, growth, and proliferation [102]. According to *Mansouri et al.*, there are three main strategies commonly practice to control biofouling: (1) continuous or intermittent biocide dosing in the feed stream, (2) process optimization and physical and chemical cleaning using a variety of chemical agents i.e. acids, alkalines+surfactants and enzymes and (3) membrane development or surface modification of existing membranes to enhance its antifouling properties [55]. Among all of the fouling resistance enhancement strategies, bioinspired surface modification approaches have recently gained attention. Shen *et al.* [87] classified these approaches into two main categories. The first category is based on the introduction of an energy barrier for foulant adsorption to the membrane surface using biologically relevant molecules such as zwitterions and glycocalyx. The second category covers approaches where the surface chemistry and structure is modified in a way that its wettability shifts toward superhydrophilicity or superhydrophobicity. The protein adhesion resistance provided by the phospholipid membrane and the

¹ UF and MF are not considered desalination membrane as they cannot remove salts however in the RO desalination plants they are used extensively as a pretreatment stage in the desalination process, which plays a critical role in preventing or minimizing fouling in RO modules.

zwitterionic functional groups on the cell surfaces has inspired researchers to design membranes in a similar way having antifouling properties. An equal number of positively and negatively charged functional groups naturally formed on the outside of lipid layer (known as zwitterions) provide not only biocompatibility with the surrounding tissues but also an overall neutral state and a strong hydration layer, which prevent the adhesion of exterior matters on the membrane lipid layer of cell membrane [44]. As presented in Table 2, surface zwitterionization of membranes via grafting zwitterionic moieties onto/from membrane surfaces has received great attention to create new generation of membranes with antifouling properties. Although these modifications are mostly on UF and MF membranes, they can easily be used for RO and NF [32] membranes as well. Zhou *et al.* improved hydrophilicity, antifouling and inorganic salts separation properties of PVDF membranes via creating a PCBMA coating layer on the membrane surfaces [113]. In their work, the performances of the pristine and modified membranes were investigated through three cycles of BSA separation-cleaning. They observed that with the increase of the zwitterionic polymer on the membrane surface, higher water flux recovery and improved membrane performance stability were observed (see Figure 8a). Wang *et al.* prepared antifouling UF zwitterionic membranes for BSA separation through blending of the zwitterionic moieties of DMMSA–BMA with PES [103]. As presented in Figure 8b, the modified membrane showed a superior BSA separation performance during four cycles of BSA separation-cleaning than pristine membrane.

Figure 8. Filtration performance of (a) pristine PVDF membrane (M0) and PVDF-coated-PCBMA membranes M2–M14 (M2–M14 represents different concentrations of CBMA in the reaction solution: 0.02, 0.06, 0.1, and 0.14 g/ml for M2, M6, M10 and M14, respectively^[113] and (b) pristine PES membrane and PES-blend-DMMSA–BMA membranes during different cycles of BSA separation-cleaning^[103].

Membrane surface glycosylation has also received significant attention. Saccharides on the cell surface are known as glycocalyx. Their main function is the biological recognition site, and their antifouling properties have been discovered lately [27]. Currently, the surface glycosylation mainly contains three methods, namely physical+ chemical and biochemical [29]. Glycosylation can render antifouling properties by creating a hydration layer on the membrane surface through the generation of extended hydroxyl group rich chains [95, 110]. Besides improving antifouling properties, the glycosylation membrane surface modification also improves the water permeation flux. A particular example by Kou *et al.* demonstrated a substantial ten-fold increase in water flux from 420 to 4350 L m⁻² h⁻¹ [38]. This significant water flux enhancement was attributed to the distinctive change of wettability of polypropylene MF membrane surface from hydrophobicity (contact angle: 120°) to hydrophilicity (contact angle: 36°) after the membrane surface modification by plasma-induced graft polymerization of allyl glucoside.

Table 2. New generation of antifouling membranes which are inspired by zwitterionic functional groups located on the outside lipid layer of cell membranes

Modification	Zwitterionic compounds	Membrane system	Reference
Grafting (High Energy Excitation)	PSBMA	Poly(vinylidene fluoride) (PVDF) MF membrane	[10]
	MPDSAH	Polypropylene (PP) MF membrane	[109]
	PSBMA	Poly(tetrafluoroethylene) (PTFE) MF membrane	[31]
	MEDSAH and MPC	Polyethersulfone (PES) UF membrane	[75]
	MPDSAH	Polysulfone (PSF) UF membrane	[108]
Grafting (Chemical Initiated)	CBMA	Poly(vinylidene fluoride) (PVDF) membrane	[113]
	SBMA	Poly(vinylidene fluoride) (PVDF) membrane	[47]
Click Chemistry	MEDSAH	Polyamide membrane	[107]
	sulfobetaine	PVA-co-PE membranes	[30]
Physical Blending	DMMSA	polyacrylonitrile UF membranes	[90]
	DMMSA-BMA	polyethersulfone (PES) UF membrane	[103]

As mentioned in Section 3.2, the epicuticular wax microstructure of lotus leaves has inspired scientists to create superhydrophobic surfaces with fouling resistance properties. In MD, the superhydrophobic modification can retard fouling through either introducing an air gap between liquid and membrane surface [76] or reducing liquid membrane contact area [53], which minimizes the foulant attachments to the surface. Changing the surface wettability towards superhydrophilicity in order to form a hydration layer on the membrane surface is another antifouling strategy proposed recently [7]. A common approach to obtain superhydrophilic surfaces is by immobilization of nanoparticles featuring hydrophilic groups (e.g. hydroxyl groups). Razmjou *et al.* showed that superhydrophilic modification of PES UF membranes through a low temperature hydrothermal coating of a thin mesoporous TiO₂

nanoparticles layer could substantially improve the antifouling properties of the membranes [79]. In their work, the TiO₂ coating layer reduced the overall roughness of the membrane surface in micro-scale while created hierarchical multilevel roughness in nano-scale (see Figure 9). The reduction in overall micro-scale roughness reduces the adsorption and entrapment of the large macromolecules such as BSA and thus increases the antifouling property of the surface. The hierarchical multilevel roughness in nano-scale shifts the wettability of the surface towards superhydrophility and as a result keeps the small foulants away from the surface, enhancing the fouling resistance property of the membrane surface.

Although bioinspired antifouling membrane surfaces have led to very encouraging results to date, their fully potential has not been fully explored yet. Most of the bioinspired antifouling strategies are applied on the UF and MF membranes while they can also be practiced on the RO membranes, which are more prone to membrane fouling. In addition, the mechanisms by which natural systems resist fouling have not been fully understood yet, thus the consequent bio-inspired antifouling surfaces are still relatively rare. Therefore, the current knowledge gap lies in the clear understanding of the mechanisms and predictive models for the development of any new bio-inspired antifouling surfaces. Future studies should focus into developing antifouling surfaces with fouling resistance against multiple species particularly for seawater desalination. Future studies should also look into creating the bio-inspired intelligent surfaces which can prevent the development of complex biofilm communities. The surfaces should efficiently avoid biofilm formation through altering targeted material surface properties and masking surface topographies. It is an enormous challenge to design such a surface that can prevent or control biofilm formation while withstanding the harsh marine environment. However, this challenge has been successfully tackled by nature. This bio-inspired approach can potentially lead us to understand and develop sustainable surface modification technologies for biofouling control during seawater desalination.

Figure 9. AFM images of (a) control UF PES membranes and (b) TiO₂ nanocomposite UF PES membranes and (c) high resolution FESEM images of UF PES nanocomposite membrane, which shows hierarchical multilevel roughness ^[79].

4 Biomimetics for desalination

Unlike most of polar molecules, water molecules pass through the cell membrane at a very slow diffusion rate [4]. However, plant cells need large amount of water being rapidly transported in many physiological cases such as opening stomata and extension of roots during germination [69]. Therefore their cells have developed an efficient pathway for water transport named aquaporin (Figure 10) [39]. Aquaporins, integral membrane proteins, are specific high rate water channels act as pore in the membrane of biological cells [58]. They have different types and exist in a wide range of cell

organisms including prokaryotic cells [96] algae, plants [5], animals and even humans [8]. Recent studies showed that aquaporin proteins have the ability to completely reject solutes (100% rejection) at water permeability of orders of magnitude higher than the current well-established pressure-driven membrane desalination processes (Table 3) [41].

Table 3. Comparison of reported permeability values of commercial membranes and biomimetic membranes

	Productivity ¹ ($\mu\text{m.s}^{-1}.\text{bar}^{-1}$)	References
FO	0.22	McCutcheon et al. [59]
RO	2	Matsuura et al. [57]
Biomimetic membranes	167	Kumar et al.[41]

¹ Productivity is permeability per unit driving force

The unique structure of aquaporin channels allows selective water permeation at much greater extent than diffusion. Furthermore, water transport through aquaporins has low energy demand and occurs via potential gradient with support of hydraulic and osmotic forces [99]. Therefore they are a useful prototype to mimic the intelligent membranes [88, 100]. Structures of aquaporins and mechanisms of transport through aquaporin channels have been extensively studied in the last decade [1, 92, 93]. Aquaporin 1 subunit is spanned across a cell membrane, which only allows water molecules to pass through the aquaporin down a concentration gradient (Figure 10). Each aquaporin is formed of six transmembrane spanning α -helices which are bundled together. The narrowest part of the hour-glass-shaped cross section of aquaporin proteins functions as a pore of about 0.28 nm. This selective pore acts as a size-exclusion filter rejecting solutes and allowing water molecules to permeate through. Besides size-exclusion, the high selectivity of aquaporin channels is due to the electrostatic repulsion and water dipole reorientation (proton exclusion) [87, 94]. As shown in Figure 10b, a charged arginine

residue close to the filter region provides an electrostatic repulsion which rejects positively charged ions. Water dipole reorientation and proton exclusion are necessary to provide a single file transport of water. The negatively charged residue lined inside aquaporin channels allows hydrogen bonding between water molecules and the protein itself. When a water molecule reaches the center of an aquaporin, it is flipped by the positive charge around two asparagines. This change in orientation allows exclusively water molecules to move across the aquaporin protein making it a highly specific channel. Analogous to biomimetic membranes recently, an attempt has been made by Zhu et al.[114] to use small pore size of zeolitic imidazolate framework (ZIF) membranes which is exactly in between the size of water molecules and hydrated ions for desalination but their system lacks the orientation. A further investigation is necessary to mimick aquaporin membranes.

Figure 10. A) Aquaporin in a cell membrane and the three proposed mechanisms of water transport in a Type I aquaporin channel: B) the electrostatic repulsion due to the presence of positively charged amino acids at the entrance of the aquaporin channel rejects cations, C) rejection of large solutes through size exclusion mechanism and D) reorientation of water molecules, which facilitates single file transport of water molecule and exclusion of protons

In order to use aquaporin channels effectively, they need to be incorporated into a stable bilayer supporting matrix. Synthetic bilayers which are made mostly from amphiphilic block copolymers (BCPs) are most commonly used to create biomimetic bilayer membranes as they have shown superior mechanical stability and better chemical resistance than lipid bilayers [13, 14]. Typically, these artificial membranes are prepared by means of either spanning an aperture [64] or depositing on a solid surface [82]. The artificial bilayer membrane can form across an aperture through two commonly used methods of folding and painting while it can be formed on a solid support through vesicle deposition and monolayer transfer [87]. Although polymer/channel protein hybrid membranes can be an excellent material for the next-generation of desalination technologies, this area is still in its infancy and more work need to be done to improve their mechanical and chemical stability as well as their large scale defect-free production. In a recent study, Kim *et al.* studied the possibility of mass production of aquaporin-Z and managed to achieve a maximum expression of his-tagged AqpZ (12.2 mg/L) using *E. coli* BL21 (DE3) host strain [35]. Although Zhao *et al.* studied the performance of aquaporin-based biomimetic membranes under a more complexed model solution, there is still a need to investigate the biomimetic membranes under real seawater desalination conditions [112]. As mentioned before, aquaporins with different types exist from very primitive living organism such as prokaryotic cells to complex ones like human. However, researchers have focused mostly on the Z type aquaporins for biomimicking membranes [49], while we suggest to use aquaporins from micro green algae *Dunaliella* sp. which is another promising candidate. Because these green microalgae can survive in saline water

with a salinity of up to 3 M NaCl through the absorption of water molecule through their aquaporin channels. Aquaporin channels can also be employed to design intelligent membranes that can reversibly be switched off and on by the addition of HgCl_2 and 2-Mercaptoethanol [92], respectively (Figure 11).

Since the biomimetic membranes have been comprehensively reviewed including the fabrication approaches, important contributions and future challenges [87, 94, 110], here we only focus on the topics which are related to the biodesalination definitions. In general, biomimetic membranes for desalination encounters a few obstacles and challenges which should be the subjects of the future works. These challenges are (a) poor fundamental understating of working principles and interaction mechanisms between functional molecules used and matrix materials, (b) expensive preparation approaches of large quantities of biomimetic membranes, (c) difficulty in scalability of current synthesis approaches, (d) stability of the biomimetic membranes in harsh environmental conditions, (e) lack of durability of the biomimetic hybrid membrane layer in the membrane desalination process with high flux and shear forces, (f) lack of compatibility between biological materials and synthetic materials, and (g) needs for discovering new prototypes which are cost-effective and easier to imitate.

Figure 11. Reversible switching off and on of *Dunaliella bardavil* aquaporins. a) algae cells show plasmolysis in saline condition 5M NaCl, b) plasmolysis was not observed in 5M NaCl in presence of HgCl₂ as the aquaporins was turned off by HgCl₂ and c) the aquaporin channels were reactivated after the addition of 2-mercaptoethanol to the saline solution.

5 Direct use of living organism for desalination

Green industry which does not impose adverse effects on the environment can lead to a sustainable and economically viable future. Exploitation of living organism for desalination can provide a chance for producing affordable fresh water with no or minimum environmental impact.

5.1 Soil-desalination versus water-desalination

Salt-tolerant species are able to survive in saline condition either in land or water. Although some of them are capable of reducing soil salinity [12, 33, 73, 115], they can not reduce the water salinity. Therefore, it is necessary to clarify the difference between soil-desalination and water-desalination by living organism. Some plant species (halophyte), such as *Tamarix* sp. and *salicornia* sp., can grow in saline lands and remove soil salinity. This type of soil salinity reduction phenomenon is known as phytodesalination [73]. Cultivation of these plants in soil may result in a reduction of soil salinity. But in order to apply them for water desalination, they need to be planted in water (hydroponic condition). In this case, there will be two scenarios: (a) the plants absorb saline water and release salt crystals through their leaves without reducing the salinity from water, (b) the cultivated plants in the saline water absorb water but exclude its salts therefore increases the water salinity [15].

5.2 Biodesalination using microbial cells

Prokaryotes have the capability to expel the intracellular salt into the outside media, as they do not have the mechanism to accumulate the salt in organelles/vesicles. Hence, finding a good candidate for biological desalination among prokaryotes including cyanobacteria is challenging because bacteria can not retain salt in the cells. Nevertheless, a novel approach to effectively remove salt from seawater has been developed by modifying microbial fuel cells (MFC) [28]. In MFC, exoelectrogenic bacteria generate electricity by degrading the organic material such as acetate. Therefore, we can generate electrical power along with the removal of organic matter from domestic and industrial wastewater

with the help of MFC [72]. This novel approach is known as microbial desalination cells (MDCs), which can desalinate water and generate electrical power or hydrogen gas using exoelectrogenic bacteria. MDC is usually constructed by modified MFC unit by adding a desalination compartment in the middle chamber [71]. As presented in Figure 12, the device contains three chambers: 1) an anion exchange membrane (AEM) next to the anode, 2) a cation exchange membrane (CEM) next to the cathode, and 3) a middle chamber for water desalination between the membranes. When electrical current is generated by bacteria on the anode, protons are released into solution. Positively charged ions are prevented from leaving the anode by the AEM and therefore negatively charged ions move from the middle chamber to the anode. In the cathode chamber protons are consumed, therefore positively charged ions move from the middle chamber to the cathode chamber. This loss of ionic species from the middle chamber results in water desalination without any water pressurization or use of draw solutions, and no electrical energy or water pressure is required. It is reported that the MDC can remove 90% salt by a single batch desalination cycle [9]. Qu *et al.* [72] improved the initial design of MDC by adopting a recirculation component (rMDC), allowing the recirculation of solutions between the anode and cathode chambers. This recirculation mitigated pH imbalances inhibiting bacterial metabolism. Although desalination by MFC is an emerging approach, its low fresh water production rate has limited its wider applications. For example the desalination rate of a 200 ml NaCl solution with 10000 mg L⁻¹ NaCl solution is about 6.7 mg h⁻¹ [37] and this value reduces to 2.8 mg h⁻¹ when NaCl concentration increased to 35000 mg L⁻¹ [104].

Currently MDC has been proven to be capable of significant reduction in water salinity, but the salinity removal efficiency is still insufficient to treat the water to drinking water level. Therefore, it can be used for the saline water pretreatment for further downstream RO processing. Reducing the salinity of the treated water would greatly benefit energy consumption in the RO process. It is suggested that the future studies of MDC should be focused on (a) increasing the fresh water

production rate, (b) finding other sources of substrates for bioelectricity generation with higher conductivities (currently mostly synthetic organic materials such as acetate is studied), (c) investigating on the possibility of using MDC for domestic and industrial wastewaters particularly for hazardous wastewaters with high COD values (above 50000 mg/L), (d) finding novel methods to balance charges to increase the extent of desalination (currently increasing the buffering capacity is practiced to keep the balance), (e) engineering practical MDC systems for desalination at larger scales and (f) studying the negative effect of biofilm formation on the cathode and membrane surface.

Figure 12. Schematics represents a three chambers photosynthetic microbial desalination cells (PMDCs) ^[37]

5.3 Genetic engineering for biodesalination

Genetic engineering or genetic manipulation is the artificial process to add a new DNA to an organism that carries a new property, e.g. Na^+ accumulation ability. Genetic engineering has been used to introduce a new generation of plants which can grow in saline conditions. However, as mentioned in section 5.1 living in saline condition differ from saline water desalination. Genetic engineer and

recombinant DNA technology have been offered as an alternative approach to introduce a new class of microorganism with Na^+ accumulating ability [2]. However, it is still challenging to use the recombinant DNA technology for the potential development of a genetically engineered organisms with Na^+ hyper accumulation ability [3]. In addition, manipulation of cell membrane transporter system and further the accumulation of Na^+ in the cell will seriously affect the cell metabolism. Thus, even through microorganism could be forced to uptake Na^+ via genetic engineering, cytoplasmic proteins and enzymes will still be damaged when NaCl concentration exceeds 100 mM [25]. In contrast to prokaryotic cells, eukaryotic cells have higher potential to be genetically engineered. Because they can reduce the Na^+ concentration in the cytosol by accumulating Na^+ in different compartment such as vacuole [66]. Cloning and heterologous expression of salt tolerant genes from *Dunaliella* into *E. coli* and tobacco cells have resulted in higher salt tolerance in these organisms [74]. Therefore, it has been demonstrated that genetic engineering is an effective tool in enhancing salt tolerance in organisms. Metabolic engineering strategies (e.g. enzyme stability, over activity of enzymes) can then be used to further improve salt tolerance in genetically engineered organisms. If these manipulated high tolerant organisms can be used for saline water desalination, they can be used as an effective pretreatment stage before RO units.

6 Conclusions and future directions

The emerging field of *biodesalination* has reached a promising stage. It has now become a distinct field of research, which may shift the fresh water production strategies from conventional thermal or pressure assisted desalination approaches to the *biodesalination* methods. The definition of “*biodesalination*” has been proposed for the first time. It is defined as employing living organisms or biological elements directly or indirectly, mimicking their structures and mechanisms, or adopting concepts from their desalination mechanisms for the production of sustainable fresh water. *Biodesalination* is categorized into three main subfields of bioinspiration, biomimetics and direct use of

living organisms for desalination. Bioinspiration for desalination has already found its way into technology. Bioinspiration alongside the advent of sophisticated high-resolution analytical techniques have provided a great opportunity for surface engineers to design state-of-the-art materials for water separation technologies. Bioinspired approaches for fouling mitigation particularly biofouling is another promising area of research, which can significantly reduce total fresh water production cost. Inspired by the surface architecture of plant leaves has led to the creation of surfaces with superior properties (e.g. extreme wettability including superhydrophobic/hydrophilic surfaces, resistance to harsh environment). This intriguing surface tuning ability has introduced new sustained desalination technologies such as MD, which are both attractive and pertinent. Contrary to the bioinspiration for desalination, the area of biomimicry for desalination is still immature and cannot be considered mainstream. Biomimetic membranes have proven excellent performance (100% salt rejection, at water permeability up orders of magnitude higher than the conventional desalination processes). However, for commercial implementation, the three main challenges of lack of fundamental understanding, scalability and costs need to be addressed. Direct use of living organisms is most sustainable approach for water desalination. The idea is still in its infancy though a few recent reports have shown the potential use of bacteria and blue green algae. To our best knowledge, an efficient aquatic plant being able to reduce salinity of seawater has not been reported. We suggest research in this area should be directed to discover or genetically engineer new species. This species should be able to reduce seawater salinity, has high growth rate and can be easily harvested.

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