Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2021.27821

238 (2021) 28–37
October

Membrane degasification for desalination industries: a literature review

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Received 18 April 2021; Accepted 9 September 2021

ABSTRACT

Degasification refers to the process of removing dissolved gases from the liquid, mainly water or any other aqueous solution. Dissolved gases like oxygen and carbon dioxide present in the boilers and distillation systems feed require efficient removal as they can lead to severe corrosion, premature failures of equipment, excessive maintenance, and shutdowns. The current conventional treatment techniques for gas removal from feeds are costly and not environmentally friendly. In this regard, there is a strong need for research on degasification and identify the strategies that can be applied in improving the effectiveness of degasification. This paper focuses on membrane degasification for desalination industries in comparison with conventional methods used for degasification, their merits and demerits, and future development of the degasification process.

Keywords: Membrane degasification; Membrane contactor; Dissolved gas; Desalination

1. Introduction

Dissolved gases like nitrogen, oxygen and carbon dioxide exist naturally in water [1]. Their existence in water therefore should be carefully controlled and monitored to prevent them from affecting water treatment processes [2]. For instance, dissolved oxygen (DO) in water reacts with metals forming an oxide layer on the surface of the metal. This then results in corrosion of the metal parts of the water treatment plant which necessitates degasification processes to avoid corrosion problems [3]. Therefore, the removal of such dissolved gases in water treatment industries/desalination industries is very critical and of great importance. The current methods for DO removal are either mechanical or chemical treatments. Hydrazine is used as a chemical for converting DO into water. Hydrazine has limitations such as slow reaction rate, undesired side product and intrinsic harmful properties (toxicity). On the other hand, mechanical processes have inherent drawbacks such as complexity, high cost and intensive energy requirement [4–7]. New membrane-based technologies known as membrane contactors (MCs) can offer far more reliable options for the removal of dissolved gases from feedwaters [6,8–11]. Membrane degasification is a liquid–gas separation process using a membrane as a separation barrier. In this process, the membrane is hydrophobic and allows the only gaseous component to diffuse through the membrane. Over the last decade, membranes for degasification have developed significantly; today the technology is commercially available as an MC for degasification of boiler feed [12–21]. The advantages of recent MCs compared with other gas removal processes are higher efficiency,

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chemically free, more compact, smaller footprint, and lighter weight which reduces capital investment [22]. Furthermore, it is flexible and easy to scale up [23].

2. Dissolved gases in seawater

The major gases present in the atmosphere are mainly nitrogen (N_2), oxygen (O_2), and argon (Ar) which cover 99.96% of dry air, excluding vapor. The amount of gas in water depends on many factors including pressure, temperature, source of water and concentration of atmospheric gas which is in contact with the water source. The main dissolved gases in seawater are nitrogen (N_2), oxygen (O_2), argon (Ar), carbon dioxide (CO_2), neon (Ne), helium (He), methane (CH_4), krypton (kr), carbon monoxide (CO), nitrous oxide (N_2O), and xenon (Xe) [24,25]. The percentage of dissolved gases in seawater is different from the fresh water due to a change in salt content ratio in seawater compared with freshwater. The percentage of dissolved gases in seawater is based on the gasses dissolved in seawater at equilibrium with the atmospheric concentration of gases. The percentage of gases in the atmosphere and seawater is shown in Table 1. Table 1 indicates that DO and argon gas percentages in seawater are higher than those in the atmospheric due to gas's solubility characteristic, temperature and salt content of seawater [24–28].

3. Analysis of dissolved oxygen

DO can be easily quantitatively analyzed by titration method, gas chromatography and DO meter. The concentrations are expressed either in milligram per liter (mg/L) or as in percentage. The sample collected for the tests are more sensitive to biological activity and environmental contamination, so it is customary to treat the sample with certain chemical reagents or to take necessary actions before analysis [29,30].

3.1. Iodometric method

In this method, DO is treated with manganese sulfate ($MnSO_4$) together with an alkaline iodide/azide mixture. Manganese instantly reacts with DO and forms insoluble manganese dioxide (MnO_2). Azide is added to the water sample along with manganese sulfate to avoid any experimental error due to nitrite ions in the water sample, which will oxidize the iodide ion (I^-) to molecular iodine (I_2) in acidic conditions [29,30]. The iodide ion (I^-) in the water sample oxidizes to iodine (I_2) in acidic conditions quantitatively. The amount of liberated iodine is then titrated against standard thiosulfate ($S_2O_3^{2-}$) using starch as an indicator. The amount of oxygen in the water sample is determined from the titer value (four molecules of thiosulfate react with one molecule of oxygen). The stoichiometric chemical equations are shown below.

$$Mn^{2+} + 2OH^- + \frac{1}{2}O_2 \longrightarrow MnO_2(s) + H_2O \quad (1)$$

$$MnO_2 + 2I^- + 4H^+ \longrightarrow Mn^{2+} + I_2 + 2H_2O \quad (2)$$

$$NaN_3 + H^+ \longrightarrow HN_3 + Na^+ \quad (3)$$

$$HN_3 + NO_2^- + H^+ \longrightarrow N_2 + N_2O + H_2O \quad (4)$$

$$I_2 + 2S_2O_3^{2-} \longrightarrow S_4O_6^{2-} + 2I^- \quad (5)$$

$$2S_2O_3^{2-} + 2H^+ + \frac{1}{2}O_2 \longrightarrow S_4O_6^{2-} + 3H_2O \quad (6)$$

3.2. Oxygen electrode method

The modern method for DO measurement is the use of a DO meter, which uses either electrochemical or optical sensors/electrodes [31–33]. These methods are rapid, accurate and inexpensive for measuring DO concentration in different sources of water. The two types of oxygen sensing technologies available in the market are optical sensor

Table 1
Percentage of gases in atmosphere and seawater

Gas	Chemical symbol	Percentage in atmosphere	Percentage in seawater
Nitrogen	N_2	78.08	62.6
Oxygen	O_2	20.95	34.3
Argon	Ar	0.934	1.6
Carbon dioxide	CO_2	0.033	1.4
Neon	Ne	0.0018	0.00097
Helium	He	0.00052	0.00023
Methane	CH_4	0.00020	0.00038
Krypton	Kr	0.00011	0.00038
Carbon monoxide	CO	0.000015	0.000017
Nitrous oxide	N_2O	0.000050	0.0015
Xenon	Xe	0.0000087	0.000054

technology and electrochemical sensor technology. Optical sensing technology measures the emitted light from the luminescent dye after the interaction of oxygen and emitted light from the luminescent dye. The cross-section of an optical dissolved oxygen sensor is shown in Fig. 1.

Electrochemical sensor technology measures the DO by diffusion and reduction principle. The DO present in the water sample diffuse across the membrane and then gets reduced on the surface of the cathode. The reduction process produces an electrical current that is directly related to oxygen concentration. The cross-section of the electrochemical oxygen sensor is shown in Fig. 2.

The reduction and oxidation reaction involved in the electrochemical oxygen sensor are as follows:

Anode: silver (Ag)

Cathode: gold/platinum (Au/Pt)

Electrolyte: aqueous potassium chloride solution (KCl)

Anodic oxidation reaction:

$$4Ag \longrightarrow 4Ag^{+} + 4e^{-} \quad (7)$$

$$4Ag^{+} + 4KCl \longrightarrow 4AgCl + 4K^{+} \quad (8)$$

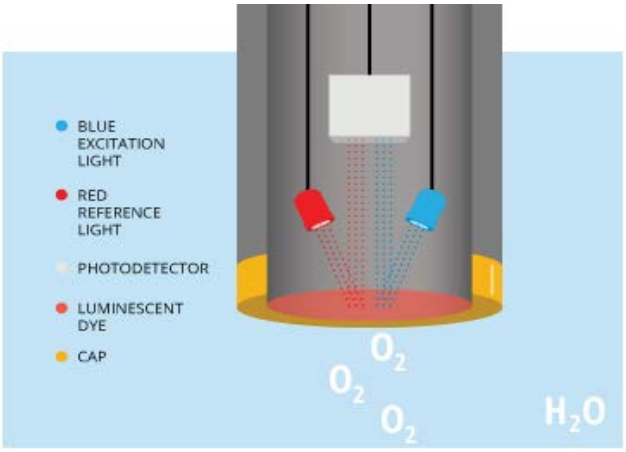

Fig. 1. Cross-section of an optical dissolved oxygen sensor.

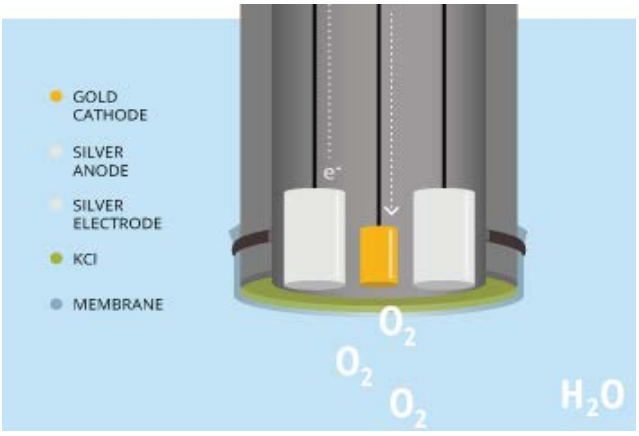

Fig. 2. Cross-section of electrochemical oxygen sensor.

Cathodic reduction reaction:

Au/Pt cathodic electrode is inert, and it does not participate in the reaction but passes electrons.

$$O_2 + 4e^{-} + 2H_2O \longrightarrow 4OH^{-} \quad (9)$$

$$4OH^{-} + 4K^{+} \longrightarrow 4KOH \quad (10)$$

Overall reaction:

$$O_2 + 2H_2O + 4KCl + 4Ag \longrightarrow 4AgCl + 4KOH \quad (11)$$

3.3. Gas chromatographic method

Gas chromatography (GC) is an advanced qualitative and quantitative analysis technique for DO measurement [34,35]. The analysis of gas molecules/compounds is made by GC using vaporization of analyte without decomposition. The different gas molecules/compounds are separated in the column and then detected quantitatively and quantitatively by the detector. The retention time indicates the quality of the gas molecules/compounds and the area under the peaks indicates the quantity. The schematic representation of GC is shown in Fig. 3.

4. Effect of dissolved gases in seawater desalination industries

The two main technologies used for seawater desalination are membrane and thermal desalination technologies [36]. Membrane desalination comprises microfiltration, ultrafiltration, nanofiltration, reverse osmosis and electrodialysis; thermal desalination includes multi-stage flash distillation and multiple-effect distillation. Dissolved gases in seawater feed may damage the boiler/or steel pipe by developing metal oxide on the surface. The main reason to develop a metal oxide is due to a chemical reaction between boiler/or steel pipe with DO in feed, salinity level, and temperature. The chemical reaction takes place at the interface between the metal surface and seawater through an electrochemical process. The overall corrosion process is a combination of anodic and cathodic reactions. For example, corrosion of iron (Fe) is:

Oxidation of iron:

$$2Fe \longrightarrow Fe^{4+} + 4e^{-} \quad (12)$$

Reduction of oxygen:

$$O_2 + 4e^{-} + 2H_2O \longrightarrow 4OH^{-} \quad (13)$$

Overall reaction is:

$$2Fe + O_2 + 2H_2O \longrightarrow 2Fe(OH)_2 \quad (14)$$

Further oxidation of ferrous hydroxide ($Fe(OH)_2$) results in ferric hydroxide ($Fe(OH)_3$) which then turns to rust

($Fe_2O_3 \cdot xH_2O$). The metal oxide developed on the surface of the boiler/or pipelines will result in reduced performance efficiency, premature failures of equipment, excessive maintenance, and shutdowns [37,38].

5. Methods of degasification

There is a number of methods available for the removal of dissolved gases from the liquid. The selection of methods for dissolved gases removal in a different application depends on the nature of the degasifying feed, targeted gas lower limit and quantity of liquid [6,39].

5.1. Pressure reduction method

The pressure reduction method is also called vacuum degasification which is used to remove dissolved gas from the liquid by reducing the partial pressure of the liquid. Sonication and stirring of the degasifying liquid under vacuum usually enhance the efficiency. Vacuum degasification is widely used in water treatment, laboratory testing, and soil purification. The two types of vacuum degasification systems commercially available are tank systems and spray systems as shown in Fig. 4 [5,6,40]. The degasification efficiency is higher in the spray system compared with the tank system due to the more interfacial surface area between gas and liquid.

5.2. Inert gas purge method

In this method, inert gas is purged under vigorous stirring for a long time to remove reactive gases such as oxygen and carbon dioxide. The solubility of inert gas in most of the solvents is less due to the chemical inertness nature, and the commonly used inert gases for the purging process are nitrogen, argon, and helium. Purging is not an effective way of degassing solvent; however, it is used in roughly degassing organic solvents. The schematic representation of the gas purge degasification system is shown in Fig. 5 [5,6,41].

5.3. Chemical method

In the chemical method, special chemical additives are used to remove dissolved gases. Oxygen is removed from the water as iron chips, sulfate as sulfur dioxide (SO_2), carbon dioxide (CO_2) as sodium carbonate (Na_2CO_3), calcium oxide (CaO), and calcium carbonate ($CaCO_3$) using hydrazine (N_2H_4), sodium sulfite (Na_2SO_3), sodium thiosulfate (Na_2S_2O, $5H_2O$), sodium sulfate (Na_2SO_4), ferrous sulfate ($FeSO_4$), ammonia (NH_3), etc. The chemical reactions involved in oxygen removal using hydrazine and sodium sulfite are shown below:

$$N_2H_4 + O_2 \longrightarrow 2H_2O + N_2 \quad (15)$$

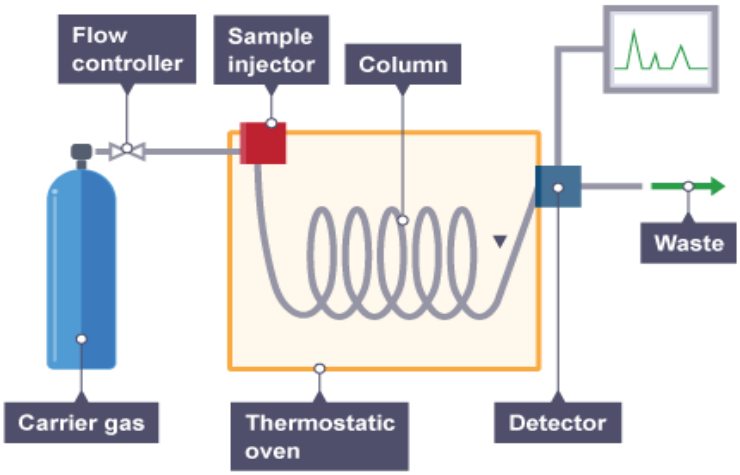

Fig. 3. The schematic representation of gas chromatography.

Tank degasification system Spray degasification system

Fig. 4. Schematic representation of tank and spray degasification system.

$$2Na_2SO_3 + O_2 \longrightarrow 2Na_2SO_4 \quad (16)$$

5.4. Mechanical degasification method

Mechanical degasification technology is one of the widely used degasification methods to remove dissolved gases like oxygen, carbon dioxide and ammonia. The method uses long packed tower connected with a vacuum pump and oxygen/gas purge system as shown in Fig. 6 [42]. The mechanical degasification system has superior degasification capability, eco-friendly and cost-effective compared with the chemical method. The main drawback of mechanical degasification technology is the requirement of a large area and higher capital investment.

5.5. Membrane degasification method

Membrane-based degasification technology is an emerging gas separation technology in desalination, thermal power plant, petrochemical and chemical industries. The membrane is a selective barrier in the degasification

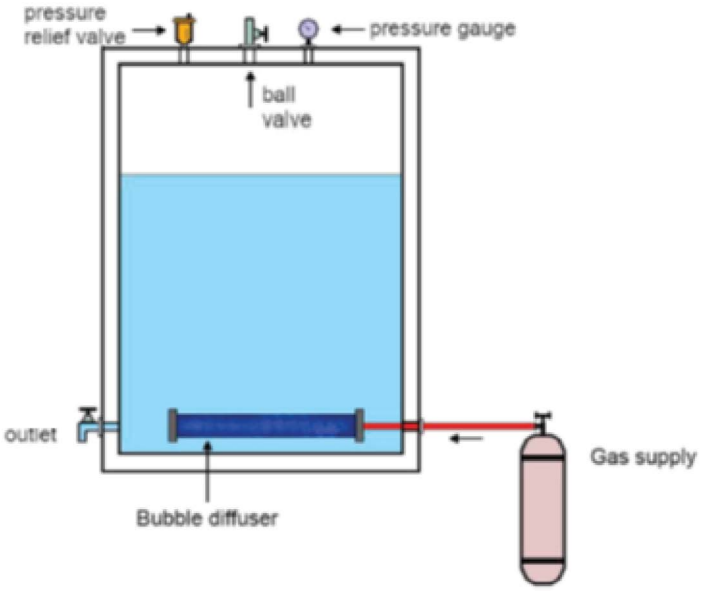

Fig. 5. Schematic representation of gas purge degasification system.

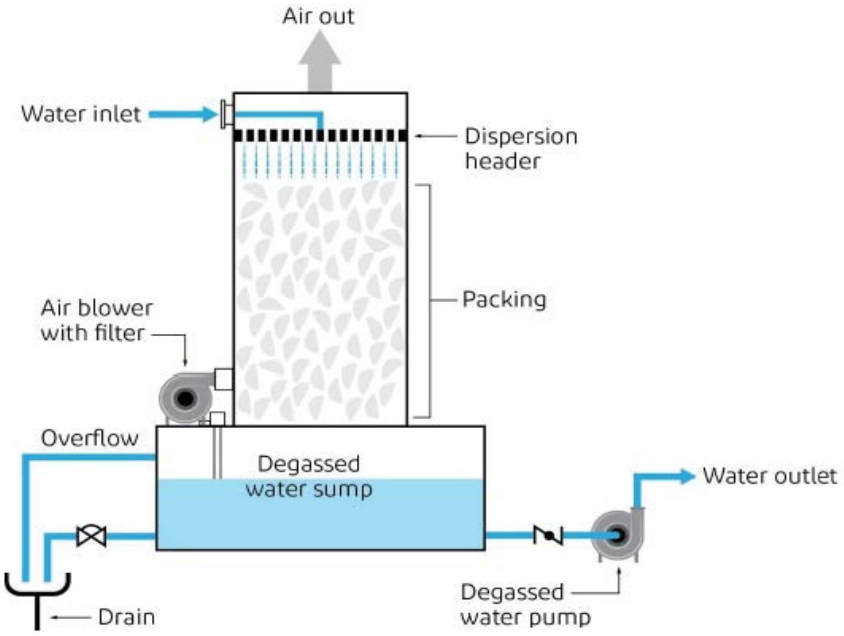

Fig. 6. Schematic representation of mechanical degasification method.

process which transports selective gaseous molecules. In desalination technologies, hydrophobic membranes are used as a selective barrier to remove dissolved oxygen and carbon dioxide as a pre-treatment technology to avoid corrosion problems. The principle of the membrane degasification process is shown in Fig. 7. The membrane degasification technology has more advantages than the aforementioned conventional methods which are: higher degasification efficiency, chemically free, compact, smaller footprint and lighter weight which reduce capital investment. Furthermore, it is flexible and easy to scale up [23,42].

6. Literature review

MCs have been applied in various applications over the past decade and have been established as an effective approach to reducing/eliminating gases from water. The elimination of dissolved gases in boiler loops is one of the widest-ranging field applications of the technology, which it can successfully accomplish, and provide significant advantages for eradicating or decreasing chemicals in the water. The application of MCs is still at its infancy stage. The available literature review studies show that more research and development are required for the application of MCs for degasification application in desalination industries.

Different methods have been applied in degasification by a number of researchers. Butler et al. [5] conducted a study in which four common techniques for the removal of DO from water were examined. One of the techniques includes; boiling water at 1 atm and boiling water under reduced pressure. In addition, the authors investigated the techniques of nitrogen purging, which seems to be both a fast and effective approach to scrub deionized water of dissolved oxygen and the sonication process conducted under reduced pressure. Winkler method was used for the analysis of residual oxygen in nitrogen purging after treatment for 20–40 min at a flow rate of 25 mL/s. The findings reveal that nitrogen purging was an effective method of eliminating oxygen from water. On the other hand, the results showed that boiling water at 1 atm is the least effective [5]. The schematic representation of the operating units used in the experiment is shown in Fig. 8.

Bessarabov et al. [43] demonstrated the use of non-porous asymmetric poly(vinyltrimethylsilane) membranes and composite membranes for gas separation. The schematic representation of the two-channel degasification MC is shown in Fig. 9. The conventional two-channel membrane experimental results showed that the overall mass-transfer coefficient depends on the liquid flow rate and the liquid-film resistance. The main benefit gained from the two-channel membrane is easy control of deoxygenation percentage in the treated water [43].

Shao et al. [44] conducted pilot-scale experiments using hollow fiber membranes for dissolved gas removal application. The investigation variable parameters of feed flow rate, temperature and vacuum. Results of the study showed that the oxygen removal efficiency and mass transport coefficient decreased with time due to fouling which can be improved by the air backwash cleaning method.

Membrana-Charlotte [20] evaluated Liqui-Cel™ MC technology for dissolved gas removal from water in many hydrocarbon processes. The Liqui-Cel® MCs are made by microporous hollow fiber membranes of hydrophobic polymers. Compared with the vacuum tower degasification technique, Liqui-Cel® MCs showed higher efficiency, no chemical usage and a smaller footprint. The Liqui-Cel® MCs technology is commercially available for the removal of dissolved oxygen and carbon dioxide from the liquid feed.

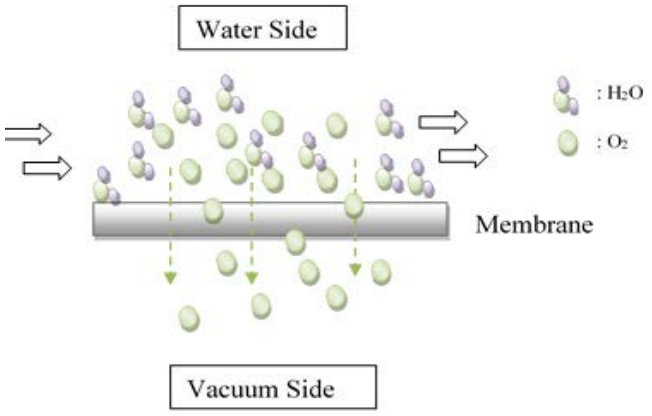

Fig. 7. Principle of membrane degasification.

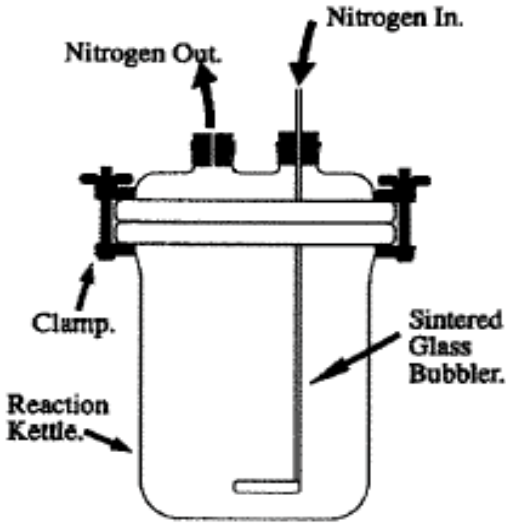

Fig. 8. Schematic representation of degasification apparatus.

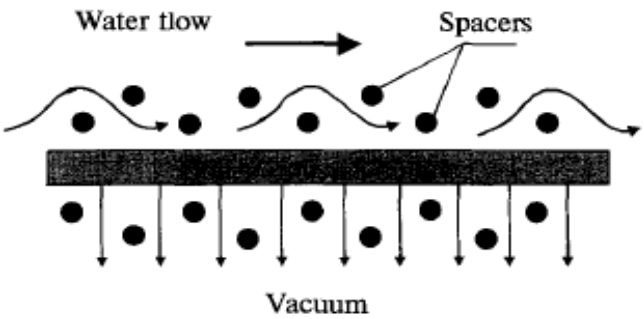

Fig. 9. Schematic diagram of the deoxygenation of water flowing in a two-channel membrane contactor.

Li et al. [15] explored the preparation and properties of hydrophobic poly(vinylidene fluoride)-SiO_2 mixed matrix membranes for DO removal from water. The findings reveal that the addition of SiO_2 nanoparticles affected the membrane morphology, physicochemical property and porosity. The membranes with 2.5 wt.% SiO_2 nanoparticle concentration showed high oxygen permeation flux [15].

Li et al. [45] studied the deoxygenation performance of polydimethylsiloxane (PDMS) mixed-matrix membranes for dissolved oxygen removal from water. The results showed that an increase in tetraethoxysilane (TEOS) content in the PDMS membrane improved the oxygen removal efficiency. The optimal concentration of the PDMS-TEOS was 10:5 (PDMS:TEOS). The deoxygenation performance indicated superior performances.

Martić et al. [46] developed a hollow fiber degasification system for the removal of dissolved gases via nitrogen vacuum mode. The nitrogen is used as a carrier gas in the degasification process and this process is highly efficient to remove DO from water at appropriate operating conations. The study proved that the cost to remove oxygen from water using membrane process (1.58 EUR/m^3) was less compared with the thermal process (as 2.99 EUR/m^3).

Mao et al. [47] explored the deoxygenation performance of the newly fabricated tri-bore hollow fiber polyvinylidene fluoride membranes. The tri-bore hollow fiber membranes showed excellent mechanical strength, high porosity and high degasification performance [47].

In recent years, the DuPont Water Solutions industry developed membrane-based Ligasep™ degasification modules with higher surface area and degasification efficiency. The modules were hydrophobic hollow fiber membranes. During the degasification process, the feed liquid flows outside the hollow fibers and a sweep gas or vacuum is applied inside the fibers. The schematic representation of the Ligasep™ degasification modules is shown in Fig. 10 [42].

Based on the aforementioned introduction and literature, it is clear that the trend toward using membrane contactors in the gas removal process in various applications including water production is growing. MC is obviously an effective method for the removal of dissolved gases in the desalination process. However, the technology is still at the infancy stage which requires more research studies to identify the strategies that can be applied to improve its effectiveness.

7. Merits and demerits of MC's

The use of the MC module in comparison to more conventional energy-intensive methods renders several advantages such as the following:

- larger surface area per unit volume,
- faster mass transfer rates,
- more independent control of phase flow rates,
- avoidance of dispersion between phases,
- possibility of a modular construction.

These advantages, without complications of flooding and loading, make the membrane modules a more attractive choice over the traditional physical and chemical methods.

The technology also does not involve the use of chemicals which makes it safer and eliminates the risk of chemical contamination in the product water. New development in the technology involves the arrangement of devices in series in order to enhance gas removal efficiency up to 99.9%. Therefore, the use of membrane degasification technology is recommended to remove dissolved oxygen in the desalination process. However, the membrane degasification technology is still at pilot-scale level due to the unavailability of suitable membranes for commercial application in desalination industries.

8. Cost comparison of MCs and conventional degasification methods

The MCs are a simple process to separate the dissolved gases from the feed water in the desalination industries

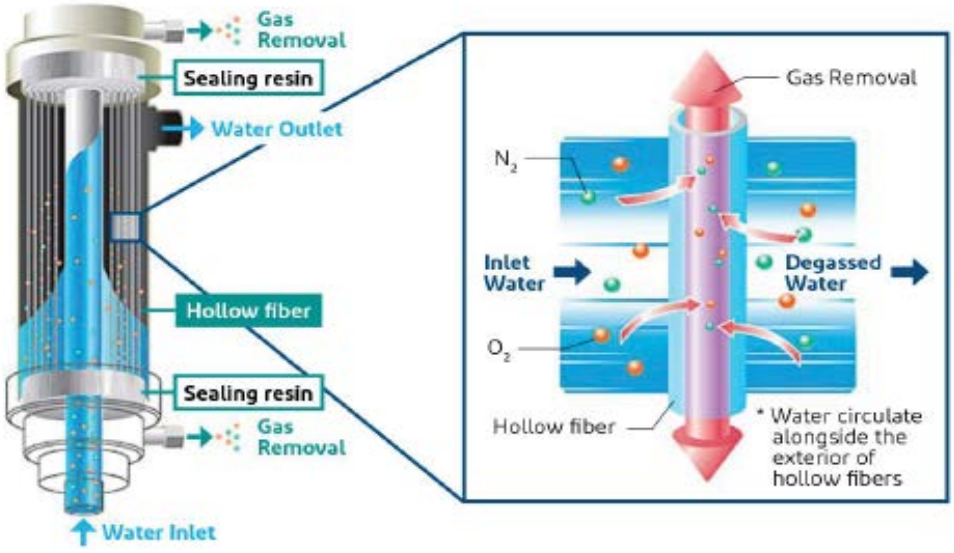

Fig. 10. Schematic representation of Ligasep™ degasification modules.

compared with conventional methods. Conventional degasification methods consume a lot of chemicals that are unsafe to the environment and add additional costs to desalination industries. On the other hand, the thermal degasification process requires higher energy to increase feed temperature compared to MC's [46]. Table 2 shows the cost of treating one cubic meter (m^3) of feed water. Thermal degasification process cost is almost two-folds higher than that in membrane degasification process [46]. This is due to additional chemical cost and energy requirements in the conventional degasification process. The theoretical chemical cost comparison data of forced draft tower and Liqui-Cel® MCs was conducted by 3 M Separation and Purification Sciences Division is shown in Table 3 [48].

9. New development in membrane degasification

Membrane degasification in desalination industries still requires research and development to establish on a commercial scale. This is mainly due to fouling, scaling and membrane wetting issues in MC's [49–52]. Therefore, researchers are developing nanoparticle incorporated modified hydrophobic polymeric, ceramic membranes and process and module designs [49,52].

9.1. Membrane fouling and scaling

Membrane fouling and scaling is a major issue in the degasification process. Membrane fouling and scaling mainly occur due to foulant and divalent ions in the feed water [49–51]. In seawater desalination industries, fouling and scaling will occur due to deposition of divalent ions (calcium sulfate, calcium carbonate, magnesium sulfate, etc.), surfactants/additives and biological (bacteria and other microorganisms) substances. Recent membrane development research proved that the membranes which are hydrophobic in nature have increasing antifouling behavior and decreasing scaling issues [15,17,43–45,53]. Additionally, hydrophobic inorganic membranes and hydrophobic thin-film composite membranes have higher contact angle (CA) and showed better degasification/gas separation performance [49,52–54].

9.2. Membrane wetting

Membrane wetting in the MCs may lead to feeding water leakage and reduces the degasification performance [3,52,55,56]. The membrane wetting is due to lower CA and lower liquid entry pressure [49]. Membrane wetting occurs due to many factors including liquid surface tension, the bigger pore size of the membrane, fouling or scaling of membrane and applied pressure or vacuum during the operation [52]. The addition of surfactants at low concentrations is found to be an effective way to overcome the wetting problem. On the other hand, the development of smaller pore size hydrophobic membranes with large CA and higher surface tension are more suitable to avoid membrane wetting. This includes hydrophobic membrane material development, structural modification, polymer modification and nanoparticle incorporation.

10. Opportunities for future development

There are still a number of issues limiting the efficiency of membrane degasification technology. The membrane degasification performance mainly depends on the physicochemical properties of the membrane, which includes the selectivity, porosity, contact angle, surface roughness, liquid entry pressure and antifouling behavior. The physicochemical properties of the membranes can be improved by developing incorporated porous nanoparticles, for example, carbon nanotube (CNT), hydrophobic graphene oxide (GO), hydrophobically modified silica nanoparticles, etc. There is also a great opportunity to develop new membranes for selective gas removal. Since membrane degasification systems consume lower energy, integration with renewable energy resources (solar, wind, waste heat) to the membrane degasification process will lower the fossil fuel dependency in the future.

11. Conclusions

Membrane degasification technology is a more suitable process to separate gases from feed water in the desalination industries. The technology is simple and capable to handle larger quantity feed water with a small footprint which will reduce the capital and operational cost. Further improvement of degasification efficiency can be achieved by adopting nanotechnology in the membrane fabrication process and optimizing the membrane module design and process parameters.

Table 2
Cost comparison of thermal and membrane degasification

Method/process	Plant I	Plant II	Plant II
Annual capacity of the plant, m^3/y	87,600	72,871	40,697
Thermal degasification, EUR/m^3	2.70	2.75	2.99
Membrane degasification, EUR/m^3	1.58	1.62	1.86

Table 3
Cost chemical comparison of forced draft tower and Liqui-Cel® MCs

Configuration	Outlet CO_2	Cost of HCl consumption	Cost of NaOH consumption	Total regeneration cost/y
Forced draft tower	8 ppm	97 USD	109 USD	75,292 USD
Liqui-Cel® MCs	1.5 ppm	75 USD	82 USD	57,686 USD

Capacity of plant: 110 m^3/h; ppm: parts per million; HCl: hydrochloric acid; NaOH: sodium hydroxide.

Acknowledgments

The authors are thankful to the Kuwait Institute for Scientific Research (KISR) for funding and supporting the implementation of this research work.

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