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Graphene oxide membranes for enhancing water purification in terrestrial and space-born applications: State of the art

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ABSTRACT

We present a compendious review of the literature and state-of-art of the research into graphene oxide (GO) membranes for both terrestrial and space-born water purification applications. The performance of GO membrane is compared to polyamide composite properties broadly used in today's desalination plants as well as to other membranes composed of various polymeric materials. An in-depth comparison is also conducted between GO membranes and the water reclamation system onboard the International Space Station (ISS). Based on empirical data from the literature, GO membranes have the potential to reduce the specific energy consumption in both RO facilities and the ISS by operating at reduced pressures while keeping high ion rejection. Mass reductions can also be achieved by replacing components in the ISS's current water reclamation system with a GO membrane. Additionally, GO membranes can increase the water availability for the crew onboard the ISS, raising the count from its current 6 members to upwards of 60 members while retaining energy and mass savings.

1. Introduction

An effective way to help alleviate the growing problem of global freshwater scarcity is by purifying seawater via reverse osmosis (RO), which is the most common architecture globally. However, while large scale seawater desalination plants have already demonstrated their much needed success, the widespread adoption of these plants is held back due to their high energy costs. One approach to lowering the energy costs is through enhancing a membrane used in RO with better separation properties. To that end, graphene oxide (GO) has been demonstrated across the literature to be an effective material for use in water purification. While some research has shown GO's effectiveness as an adsorbent designed to uptake and remove targeted species [1–6], others have focused on using GO and modified GO as a membrane to filter out impurities [7–12]. These investigations have shown that GO membranes greatly lowered the applied pressures required for water separation from salts [13–18] and organic molecules [19–22], translating into a significant reduction in energy consumption.

GO membranes can also be used in other water purification ventures, including those beyond Earth. The International Space Station (ISS) currently relies on a monthly resupply from Earth in order to continue its mission. Installing an environmental closed-loop life support system (ECLSS) onboard the ISS to recycle water has supplemented this resupply, demonstrating a water recovery rate of 70% from processing astronauts' urine and other wastewaters. However, several unforeseen setbacks have limited the system's full potential. Additionally, the ECLSS system must be upgraded both to further reduce logistical dependence on terrestrial resupply and to test hardware that will be used on future missions to deep space, including Mars.

This article presents the current state of research into the methods used in GO membrane synthesis as well as the experimental conditions and varying techniques used to characterize them in order to better understand how the current theories of the proposed GO water purification mechanisms originated. The potential benefits of replacing commercial RO membranes in desalination plants with GO membranes to help reduce energy consumption and promote wider adoption of the technology is also detailed. Finally, a thorough analysis of applying GO membranes to enhance the water recovery systems onboard the ISS is outlined with a promising outlook.

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2. GO membrane implementation: motivation and needs

2.1. Enhancing terrestrial desalination technologies

2.1.1. RO membranes

Today's desalination of seawater, the composition [23] of which is summarized in Table S1, is practiced across the globe predominately by RO plants with a production capacity that has increased from 38 million m^3/day in 2009 [24] to just over 51 million m^3/day in 2015 [25]. This capacity accounts for 59% of global seawater desalination versus other processes, such as multi-stage flash (MSF) distillation. One reason for this trend is that RO requires much less energy (3–6 kWh) than MSF (10–16 kWh) per 1 m^3 of processed water [26]. However, the required energy for RO is still a high enough barrier at the scales seen in plants today (100,000+ m^3/day) to prevent widespread adoption in areas that need it most. A summary of several plants and the RO membranes they use is presented in Table S2, though the most commonly used membranes are thin film polyamide composites. In order to make RO desalination technology more feasible, its energy costs must be lowered and their effectiveness improved. Out of the many processes associated with RO desalination, the mechanical energy needed to force the seawater across RO membranes accounts for up to 75% of the total power required for the entire process, using 5 to 8 MPa of applied pressure [27–32]. Therefore, reducing the amount of pressure required during the pumping process towards the thermodynamic equilibrium limit will directly and meaningfully lower the energy cost of the entire process.

Different designs exist for decreasing the applied pressure. One such design is a hybrid desalination plant which can be described as using two or more desalination techniques simultaneously, for instance MSF and RO, to purify water [24,28,33]. A typical design first processes the seawater through an MSF array and then allows the resulting permeate to be pumped through an RO setup. Advantages to a hybrid design include using common inlets/outlets into the sea to save on capital costs as well as controlling the temperature of the water entering the RO membranes using waste heat from the MSF process to better maintain the optimal temperature of the feed water year-round. Lately, hybrid desalination plants have expanded in definition to also encompass those plants which take advantage of renewable energy sources, such as solar [24,28,30,34,35] and wind [30], to supplement the energy costs. Another design beyond hybrid systems is the RO deep sea system, which performs RO underwater in offshore locations and then pumps the product water onto land [36]. Advantages to this setup include a greatly reduced energy consumption of 0.7 kWh/m^3, or 77% less than typical RO plants. However, one large drawback to this design is that other components of the plant are also immersed underwater along with the purification components, decreasing overall system lifetime. Additionally, the RO deep sea system restricts certain RO membranes from being used due to other factors, such as no pretreatment of the water being performed.

2.1.2. GO membranes

In comparison to the typical thin film polyamide composite membrane most widely used for RO, GO membranes appear to offer a better alternative for low-cost, low energy desalination. Due to their layered architecture and physically-based sieving mechanism (discussed in further sections), GO membranes have demonstrated ion rejections approaching that of RO membranes. This can be illustrated using the study from Geise et al. (2011) where several polymeric membranes used in seawater RO (SWRO) were investigated for their salt passage (NaCl) and permeance properties [37]. Fig. 1 depicts these SWRO values as well as those from GO membranes in the literature. Several GO membranes yield ion rejection values similar to SWRO; almost half demonstrate 15% passage or less while the majority of the GO membranes yield better permeance. Those membranes which had poor NaCl rejection nevertheless had much higher rejection for other salts. For instance, Wang et al. (2016) and Zhang et al. (2017) also demonstrated 57% [38] and 66% [39] rejection for Na_2SO_4, respectively. Likewise, Goh et al. (2015) observed a rejection for $CaCl_2$ of 92% [13]. These values and more can be found in Table 1.

In addition to their separation performance, GO membranes have also demonstrated excellent anti-fouling and antimicrobial properties (discussed in later sections) while operating at significantly reduced pressures relative to RO membranes. Consequently, the energy demand for water purification can then be lowered; using this pressure reduction, the specific energy of systems using GO membranes can be calculated and compared to those using RO membranes. However, certain assumptions must be considered before quantitative predictions can be made.

First, the specific energy and applied pressures need to be averaged across commercial RO membranes to create a real-world baseline. This calculation yields 3.12 kWh/m^3 of specific energy required and a transmembrane pressure of 6.11 MPa. Next, since the amount of power required for the applied pressure in RO plants is approximately 75% of the total power used, the averaged specific energy is to be scaled accordingly. Hence, 2.34 kWh/m^3 for RO plants is then used to calculate the specific power of GO membranes. Finally, by taking the ratio of pressures used for GO membranes relative to the averaged RO membrane pressures, the specific power of GO membranes, and thus the decrease in pumping energy required, can be obtained. These assumptions culminate as Eq. (1) to generate specific energy for GO membranes as comparison data. Calculation results are listed in Table 1 and additional RO membrane details are listed in Table S2.

$$GO\left(\frac{kWh}{m^3}\right) = 2.34\frac{kWh_{RO}}{m^3}\left(\frac{MPa_{GO}}{6.11\ MPa_{RO}}\right) \tag{1}$$

Although the concentrations of salts used in most investigations are much lower than that in seawater, the main mechanism of species rejection in GO membranes is the physical sieving of species based on their size, which will be elaborated on more in Section 6. Because the size of species is fundamentally constant, ion rejection in GO membranes is largely resistive to concentration variations: if the ion is too big to fit through the membrane, it will be rejected. One study highlights this fact using NaCl concentrations approaching seawater with > 98% ion rejection [49]. Additionally, GO membranes have demonstrated their robustness by being able to withstand pressure levels similar to those used in current RO processes using polymeric membranes, with high ion rejection being achieved at 5.5 MPa of hydrostatic pressure [49] as well as at 5 MPa of flow pressure [48].

Upon examination of GO membranes, energy savings range from 10% to 76% with comparable ion rejection of 98.4% and 99.45%, respectively. Select GO membranes with the highest ion rejection and water fluxes are highlighted in Table 2. Consequently, in conjunction with a typical output for a modern desalination facility of 100,000 m^3/day, an energy savings on the order of 178,000 kWh/day could be realized by using GO membranes. Additionally, exciting implications arise when this reduction is applied to the current global RO production capacity, culminating in a savings of 90.8 GWh/day. This energy savings does not factor in the increased water flux of some GO membranes as well as other factors, including high antimicrobial and antifouling resistance, which would impart additional benefits.

2.2. Enhancing celestial life support systems

Not only can desalination technologies on Earth benefit from the implementation of GO membranes, but so too can systems used in generating freshwater for space exploration. Due to the current limitations associated with spaceflight, missions remain logistically tethered to our home planet. The ISS currently averages resupply of mission critical consumables such as water once per month, though this rate varies [54]. While this resupply is necessary to support ongoing missions, they are expensive to deliver due to the high energy cost in lifting