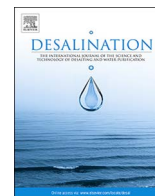




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## Desalination

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## A review on inorganic membranes for desalination and wastewater treatment

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## ABSTRACT

The sustainability of global clean and safe water supply is one of the grand challenges facing the world. Membrane technology based on polymeric membranes is one of the most important and widely recognized technologies for desalination and wastewater treatment. While polymeric membranes are known to be plagued with some bottlenecks, the technical progress and the accompanying knowledge in inorganic membrane development have grown inexorably to solve some of the underlying issues. Aside from the conventionally used ceramic membranes which based on metal oxides, nanostructures such as zeolites, metal organic frameworks and carbon based materials have sparked enormous interest in the preparation of inorganic membranes owing to their tunable nanoscaled structural properties that can render excellent rejection and/or ultrafast water transport. This review provides insights into the physico-chemical properties and fabrication approaches of different classes of inorganic membranes. The transport mechanisms that are associated to their unique structural features are also discussed. Furthermore, the performance evaluation of these inorganic membranes in a wide spectrum of desalination and wastewater treatment applications are also elaborated. Finally, the challenges in the development of inorganic membrane for practical commercial application are identified and the future perspectives are presented.

### 1. Introduction

Industrialization and urbanization are the linchpin to the global economic growth and development that has significantly contributed to the human welfare. Unfortunately, they have also led to increasingly uncontrollable discharge of wastewater and consequently created a series of environmental problems that associate to the climate changes. As water usage is a key operational part in most of the industrial processes, the production of wastewater is inevitable in a wide spectrum of industries [1]. With more preeminent economic growth, wastewater discharges from both industrial and municipal sources have also grown at the similar pace. Water pollution has led to the build-up of environmental contaminants where increasing evidences of pollution and deterioration of water quality have been witnessed in more developing countries. In order to restore the environment and deter the hazards to mankind, various approaches have been targeted to treat the industrial waste [2–5].

In another worsening scenario, continuing population growth, urbanization and industrialization have also resulted in alarming global water demand. In fact, with the water consumption that has been escalating at more than twice the rate of the population in the last

century, water scarcity is among the most chronic issues that challenges the mankind and world in the 21st century. The United Nations predicts that in the coming decade, half of the countries worldwide will confront with water stress if not outright shortages. Desalination and water reuse has been long acknowledged as a feasible mainstay to address this grand challenge by offering safe and clean water in many arid areas, coastal regions or remote locations. Particularly for water-scarce countries such as Middle Eastern and North Africa Countries that have attempted and implemented all other measures to secure fresh water, desalination may serve as the most viable approach to supply fresh water supply hence to allow sustainable development and support population growth in this modern society [6]. Membrane and other nanoporous materials have been considered as the essential technologies to address global water shortage problem [7]. Extensive product line of water and wastewater filtration systems such as conventional pressure driven seawater and brackish water reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF) and microfiltration (MF) as well as osmotically driven forward osmosis (FO) and pressure retarded osmosis (PRO) have been introduced in the market [8–13]. Emerging desalination technologies such as microbial fuel cell and membrane capacitive deionization also found increasing importance in combating water

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issues [14,15]. At the academic level, fundamental, applied and translational research related to desalination technologies are actively blooming as the result of the material sciences and engineering advancement made in this field [16,17].

Undoubtedly, polymeric membranes are currently in the forefront of desalination and wastewater treatment membranes due to their well establishment and excellent long term track records. Today, the industry market based on polymeric membranes for water purification is impressively huge. Contemporary researches have been conducted to address several challenges related to material-based limitations posed by polymeric membranes to resolve the issues with the best practice solutions. One of the critical issues in the use of polymeric membranes in desalination and wastewater treatment deals with the trade-off relationship between permeability and selectivity as well as membrane fouling and scaling. The adverse effects towards sustainable application of polymeric membranes for water purification have triggered great interest in improving the properties of polymeric membranes through different strategies of modification. One of the most impressive and state-of-the-art achievements in this field is the development of mixed matrix membranes (MMMs) and thin film nanocomposites (TFNs) [18]. The innovation that combines the unique features of both polymeric matrix and inorganic fillers has emerged as a promising solution for the underlying problems.

While development of polymeric membrane is progressing in a steady pace, inorganic membranes with exceptional chemical and physical properties, high tunability and reusability have also gained a growing interest. In a broad term, inorganic membranes consist of ceramic membranes and carbon-based membranes. Oxides such as alumina, silica, titania or the mixtures of these components are some of the commonly used commercialized ceramic membranes [19]. From maintenance and economical point of view, inorganic membranes are ideal for in-place chemical cleaning where high temperatures and subsequent chemical cleaning can easily bring about sustainable flux recovery. More importantly, inorganic membranes are less susceptible to the deterioration by bacteria, which are responsible for the bio-fouling degradation of most polymeric membranes [20]. Depending on the materials' nature, inorganic membranes can be fabricated in thin film and multi-layer supporting structures with adequately strong and permeable characteristics or they can also be made in self-supporting structures as standalone sheets and tubes if they are sufficiently permeable [21]. A typical ceramic membrane is made up of a macroporous support layer and a meso- or micro-porous active layer. Historically, due to the unfavourably high manufacturing cost and difficulties in handling, the industrial use of inorganic membranes in water reclamation was uncommon [22]. In fact, at commercial scale, inorganic membranes have only been more acceptably used in the applications that represent great challenges to the conventionally used polymeric membranes such as extremely high temperature as well as harsh and highly contaminated feed environment. Recently, the preparation of low cost ceramic membranes with composition mainly based on affordable clays and organic pore formers, with the cost more similar to that of polymeric membranes, has encouraged the prevalence of ceramic membranes at larger scale [23–26].

Desalination and wastewater treatment using inorganic membranes is an attractive alternative for some challenging water treatment processes such as wastewater containing radioactive substances and highly concentrated organic effluents, oil and grease where the conventional polymeric membranes are either inapplicable or inefficient due to severe fouling and material instability issues [27–30]. Inorganic membranes are also advantageous in brackish water treatment, seawater pretreatment and high temperature desalination, where high rejection above 99% is not a critical requirement [31]. These types of industrial wastewater are normally discharged in large quantity and pose a great treat to the conventional wastewater treatment system due to their complex nature and chemical composition [30]. Various computational and experimental studies have been performed to study the ability of

nanoporous inorganic membranes to remove pollutant ions and/or molecules with different natures and characteristics. The emergence of some new materials particularly metal organic framework (MOF) and carbon-based nanostructures such as carbon nanotubes (CNTs), graphene and its derivatives has offered a new era for enabling membrane science and technology with accomplished separation performance. The unique features of these materials have been promptly harnessed to design a radically new kind of inorganic membrane. Free standing inorganic membranes fabricated from nanomaterials such as CNT, graphene and MOF have been touted as the next big thing in membrane technology. The high permeability and selectivity of these membranes could allow higher water flux and show improved energy savings at high recovery. As such, these materials offer huge opportunities for the implementation of commercial plants with smaller footprint. Besides, the new structural concepts of these membrane materials are anticipated to ameliorate some existing issues found in the contemporarily used materials and accomplish the practically efficient, high productivity and energy saving desalination and wastewater treatment processes.

In the interest of the growing potential of inorganic membrane in desalination and wastewater treatment, this review compiles the contemporary advancement and remarkable achievements made in this field. The first part of this review focuses on the structural properties as well as the common design, fabrication and modification approaches for various inorganic membranes. The theoretical and empirical knowledge in the fluid transport phenomena and mechanisms of inorganic membranes is also covered to provide understanding on the flux and rejection behavior of the membranes. In the second part of this article, the performance evaluations of various inorganic membranes in desalination and several important wastewater treatments are highlighted. The potentials of ceramic membranes and carbon-based inorganic membranes in treating oily wastewater, dye and organic compounds, inorganic ions, heavy metal and radioactive ions are comprehensively elaborated based on the recent and current publications. Finally, the challenges, issues as well as the opportunities of inorganic membranes in advancing membrane technology are also addressed.

## 2. Inorganic membranes

### 2.1. Ceramic membranes

Ceramic membranes could be made from typical metal oxides (alumina, silica and zirconia) aluminasilicate zeolite as well as recently emerging metal organic framework. In this section, several classes of ceramic membranes made from different inorganic materials are discussed.

#### 2.1.1. Metal oxide membranes

Ceramic membranes with a wide range of insoluble oxides have shown interesting separation and processing properties. Out of these, mesoporous membranes zirconia and titania have been applied in the industry. Conventionally, a porous metal oxide based ceramic membrane is characterized by a multi-layered asymmetric structure which composed of a thicker support layer with relatively large pores ( $\sim 0.5 \mu\text{m}$ ) to provide mechanical integrity for the membrane systems, an intermediate layer to reduce pore size to mesoporous dimensions (2–5 nm) and a much thinner top layer with small and selective pores ( $< 1 \text{ nm}$ ) for selective separation [32,33]. Sol-gel is one of the most feasible methods for the preparation of metal oxide ceramic membranes. Depending on the media or solvent used, this technique can be further divided into the colloidal and polymeric sol-gel routes. A two-step process has been commonly applied to obtain colloidal sols from oxide precursors, i.e. precipitation of a condensed and hydroxylated species from hydrolyzed precursors followed by the transformation of precipitation into a stable sol through a peptization reaction [34].

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